UNIVERSITY OF SOUTHAMPTON

FACULTY OF ENGINEERING, SCIENCE AND MATHEMATICS

School of Civil Engineering & the Environment

Verification of methodologies for estimating human exposure to high levels of mercury pollution in the environment

by

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ABSTRACT

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VERIFICATION OF METHODOLOGIES FOR ESTIMATING HUMAN EXPOSURE TO HIGH LEVELS OF MERCURY POLLUTION IN THE ENVIRONMENT

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A considerable amount of work has been conducted developing exposure estimate models for quantitative evaluation of Hg intake and human health risks, but few have assessed the applicability and the validity for evaluating the risks posed by Hg in the environment and have achieved very mixed results. The present study focused on verifying the daily Hg intake estimates using exposure estimate models. Deterministic methods and the probabilistic methods (the Monte Carlo) were applied to simulate the daily Hg intake doses which were verified by comparing the estimates to those established from measured Hg concentrations in the hair of 289 participants. The results showed that the single-value deterministic method for simulating Hg exposure levels overestimated the level of risk by a factor of 1.5 when compared with the highest concentration of the Hg observed in the hair of the study population. The average daily Hg intake doses simulated using the probabilistic simulation were similar in distribution to the biomarker data, with the variability of 23%. The difference between the probabilistic simulation and the data derived from hair Hg levels was considered to be most likely due to the uncertainties in unconfirmed questionnaire-based survey data, small sampling sizes and the surrogates used in the exposure models. When the reference dose (RfD) of 0.1 µg/kg body weight/day was adopted as the acceptable dose for daily intake rate, there were approximately 19% estimated to have potential Hg exposure risks based on the Monte Carlo simulation. This percentage was favourably similar to the 17% determined from Hg concentrations in the hair samples. The findings implied that the existing exposure models together with the probabilistic approach were appropriate for the research of human exposure to Hg. On the other hand, low Hg levels in the participants' hair indicated that Hg accumulated in the study population was not very serious, probably due to the good Hg absorptivity of the on-site fly ash. However, it should be advised that consumption of river fish elevates the health risks to the local population.

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AUTHOR'S DECLARATION

I, Hui-Wen Hsiao, declare that the thesis entitled Verification of methodologies for estimating human exposure to high levels of mercury pollution in the environment

and the work presented in the thesis are both my own, and have been generated by me as the result of my own original research. I confirm that:

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1. INTRODUCTION

1.1 Background

Mercury (Hg) is released both naturally into the environment and by human activities. Its chemistry is complex being present in a number of chemical forms that can be readily transformed from one to another. The transformation into different chemical forms allows Hg to be transferred between soils, water, air and animals. This mobility has resulted in it becoming one of the most worldwide spread pollutants. There are three chemical forms of Hg: elemental Hg (Hg^0), inorganic Hg (Hg^+ and Hg^{2+}), and organic Hg compounds. The different forms of Hg have distinctive toxicological profiles which result in them having different clinical symptoms. Elemental Hg has the characteristic of having a high vapour pressure and poses a threat to human health mainly via inhalation. Inorganic mercurial salts, probably the commonest form of Hg in soils, vary in solubility and absorptive properties. It may cause toxicity to the kidneys and its ability to move via the placenta presents a high risk to the fetus (Gochfeld, 2003, Tchounwou et al., 2003). Organic forms of Hg, particularly methylmercury (MeHg) and its derivatives, are extremely neuro-toxic and are bioaccumulated in the food chain, resulting in them presenting a major health hazard. It is the major route of Hg exposure for humans.

High levels of exposure to Hg may result in neural problems (Bittner et al., 1998, World Health Organization (WHO), 1990, Clarkson et al., 2003b), and brain damage may occur in children due to prenatal or postnatal exposure (WHO, 1990). The ready transformation of Hg from a low toxicity form to the extremely highly toxic methylated form, which is bioaccumulated significantly, broadens the potential exposure routes and increases the risk to human health. In 1956, the most significant case of Hg poisoning occurred at Minamata, in Japan. The Chisso acetaldehyde plant released high levels of inorganic Hg into the sea via a river that drained into the nearby bay (Harada, 1995). Methylation within the sea resulted in MeHg accumulation to toxic levels in the fish and shellfish. The aquatic fish and shellfish was consumed raw by local inhabitants resulting in the death of 378 people by the end of 1980 and illness in many more (Tamashiro et al., 1984). In 1971, another major Hg incident occurred in Iraq, where wheat seeds treated with organic Hg fungicides were ground into flour, baked into bread, and consumed by the local people. Not only did the two cases arouse intense interest in the environmental fate of Hg and its ability to provide potential exposure pathways to humans, but they also drew attention to bioaccumulation and biomagnification of Hg in human bodies.

To reduce the risk of harmful effects on humans resulting from Hg exposure, the United States Environmental Protection Agency (USEPA) attempted to quantify the amount of Hg absorption in human bodies and recommends a safety level of the daily intake rate of less than a reference dose (RfD) of 0.1 μg/kg body weight/day (USEPA, 1997d). This catch all approach to minimising exposure risk to the population as a whole is clear to follow, but it gives limited help in managing the risk of exposure to individuals. The later task is made much more difficult by the fact that Hg may be present in the environment or exposure pathways in metallic, salts or organic form, the later being many times more toxic than the other forms. The catch all reference dose of 0.1 μg/kg body weight/day assumes that all Hg intake is in the organic form, as this is the form most likely to be absorbed by the body. To establish the true risk to individuals is therefore much more complex given the different Hg forms, polluted sites and exposure pathways. For this reason, a number of approaches have been developed for pathway assessment in the past 10 years (Tixier et al., 2002, Fryer et al., 2006, Boyce and Garry, 2002). Two general approaches have been developed and adopted for numerical estimations of risk levels to human Hg exposure. Deterministic methods that use single value input variables, which are often the higher observed exposure, give a single value for the worse case exposure level, and probabilistic methods which depend on deriving probability distribution functions for the different input variables to produce a stochastic evaluation of possible outcomes. With the ready availability of stochastic modelling tools for risk assessments, the probabilistic approach is becoming much more widely used as it provides a more realistic estimate of risk (Boon et al., 2003).

Since the aquatic food chains have been indicated as the primary Hg exposure route for humans (WHO, 1990), a number of studies have been carried out to assess the risk of exceeding intake doses of Hg via fish and shellfish consumption (Agusa et al., 2005, Boischio and Henshel, 2000a, Nasreddine and Parent-Massin, 2002, Tran et al., 2004, Ysart et al., 1999). Biases and errors are however often reported, and are mainly the result of both our poor understanding of Hg transformation forms in the aquatic environment and the unspecified exposure data traditionally collected by questionnaires, demographic or survey statistics, behaviour observation, or activity diaries. The under- or overestimated results may lead to misinterpretation and poor judgement since the major function of the assessment of human exposure is to help the decision-making process in risk management.

Paustenbach et al. (2006) considered that the validity of Hg exposure assessments is a challenge for the future that urgently needs attention. To verify the accuracy of estimates of Hg exposure evaluated on the basis of fish and shellfish consumption, several studies have compared the back-calculated Hg intake doses from biomarker data (Loranger et al., 2002, Canuel et al., 2006, Gosselin et al., 2006). The Hg level in hair is the most frequently used biomarker to determine exposure as it seems to reflect the actual burdens of the chemical levels in human bodies (Nieuwenhuijsen et al., 2006, Paustenbach and Galbraith, 2006). However, discrepancies have been found between biomarker data and exposure estimates based on questionnaire data. For example, Loranger et al. (2002) indicated that questionnaire-based Hg intake estimates were higher than that established from the concentrations of Hg in hair and that the reason might be due to self-reporting error of survey participants. A similar overestimate was observed by Canuel et al. (2006), and the considerable discrepancy between the estimated Hg intake by individuals and the amounts of Hg measured in hair samples was suspect due to the variability of pharmacokinetic constants within individuals and between ethnic groups. These studies implicated that the influence of uncertainty and variability generated from insufficient input parameter data to support traditional and deterministic exposure assessments as being a problem. As a result, probabilistic techniques, such as Monte Carlo, have been adopted in the assessments of human exposure to Hg (Chien et al., 2006, Hoover et al., 1997, Chien et al., 2007). Despite the use of the methodology in exposure risk assessments, none of the studies validated their results. Therefore, the applicability of existing Hg exposure models using the probabilistic technique is still unclear.

The present study aims to apply traditional deterministic and probabilistic risk assessment techniques to evaluate the risk posed by Hg waste from a disused acetaldehyde plant at Temirtau, in particular the risk to the residents of the town and the villages of the Nura valley.

In particular this thesis

- ❖ Adopts quantitative methodologies of deterministic and probabilistic approaches to assess Hg exposure risks for the possible targeted population.
- ❖ Verifies the validity of the deterministic and probabilistic approaches.
- ❖ Identifies the adoptability of these approaches and suggests improvements.
- ❖ Discusses the uncertainty and variability in the methodologies of exposure assessments.

1.2 Overview of the study

Chapter 2 Literature review

This chapter reviews the literature relating to the potential health impacts on humans due to Hg in the environment and the importance of different exposure pathways. This chapter also introduces the approaches that have been used by the preliminary studies associated with risk assessments for human exposure to Hg and discusses the validity of the assessments.

Chapter 3 Materials and methods

The study site is described together with survey procedures and analytical methodologies used in this study.

Chapter 4 Survey results

This chapter describes the results of the field study, in particular establishes the main sources and secondary sources of pollution and the levels of pollution. Results are also presented on Hg levels in potential exposure media together with the household survey information.

In addition, this chapter presents the results of the exposure levels of the study population via Hg concentrations in hair.

Chapter 5 Risk assessment for humans exposed to mercury

Based on the survey data in Chapter 4, exposure estimates are carried out in this chapter using both the methods of probabilistic and deterministic based on two assumptions with respect to different MeHg proportions in the exposure media. Hg exposure risks of the population are assessed according to the critical levels in various exposure pathways.

Chapter 6 Verification of modelled Hg exposure estimates with observed data

The verification of the Hg intake doses simulated using the probabilistic and deterministic methods is conducted by comparing the simulation results with that established from the observed Hg concentrations in the hair of the study population.

This chapter also discusses the uncertainty and variability originating from the exposure assessments and the validation processes.

Chapter 7 Exposure model for estimating mercury levels in hair

Statistical analyses are applied to examine the relationships between Hg concentrations in the individuals' hair and their demographic characteristics together with dietary intake patterns. A developed exposure model was used to explain the relationship between influential variables and the population's Hg exposure levels as shown via Hg concentrations in hair.

Chapter 8 Conclusions and the recommendations

2. LITERATURE REVIEW

2.1 Hg in the environment and human exposure to Hg

2.1.1 Hg in the environment

2.1.1.1 Three forms of Hg

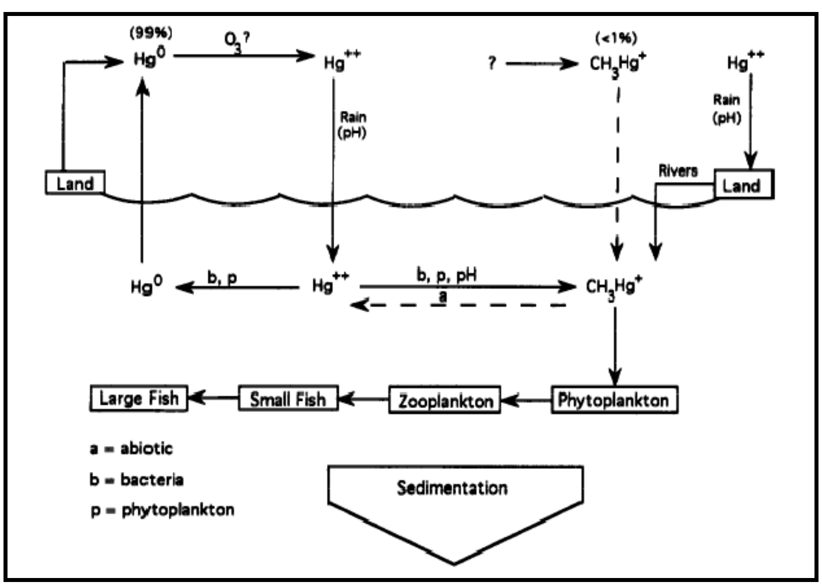
Three forms of Hg occur in the environment: elemental, inorganic and organic. Elemental Hg (Hg^0) is silvery in appearance and it is the only metal that is a liquid at room temperature. It is commonly called liquid silver, or quicksilver. Hg has a valency of 1 or 2, the atomic number of 80, atomic weight of 200.59, melting point of -38.87°C, boiling point of 356.58°C, density of 13.546 g/mL, and it has a relatively high vapour pressure of 13 mg/m^3 at 20°C (Agency for Toxic Substances and Disease Registry (ATSDR), 1999a). Elemental Hg is usually prepared from the major ore of Hg, cinnabar. Hg has been used by humans for at least two millennia. Today it is still used in thermometers, electric devices such as barometers and nanometers, medical equipment and the chemical industry (Clarkson et al., 2003a). Because of the relatively high vapour pressure, Hg^0 vapour emits from its liquid form and usually remains as gaseous Hg in our environment.

Hg also exists in the inorganic form. Inorganic mercurial salts (Hg^+ and Hg^{2+}) vary in solubility and absorptive properties (Gochfeld, 2003). Some of the most important Hg salts are mercurous chloride (Hg_2Cl_2, or calomel), occasionally used in medicines; mercuric fulminate [$Hg(OCN_2)$], explosives detonators; and mercuric sulfide (HgS), vermillion used in traditional oil paint (Tchounwou et al., 2003).

Lastly, organic Hg compounds are widely distributed in the environment. The methyl, ethyl, and phenyl series are the main organic forms that have been marketed as industrial compounds, primarily as biocides, pesticides and household antiseptics (Tchounwou et al., 2003). These uses have brought organic Hg to the environment. Among the organic compounds, MeHg is produced naturally through the methylation of inorganic Hg in aquatic sediments and continually accumulated in waters and rivers. It is the form most readily accumulated in the human food chains causing concerns about public health (Gochfeld, 2003, Clarkson et al., 2003b).

2.1.1.2 Fate and transport of Hg

The earth's crust contains 0.5 parts per million of Hg, which is released slowly from rocks and minerals as they erode under normal weathering condition. Other natural sources include forest fires and other wood-burning practices, volcanoes and hot springs. Once the vapour enters the atmosphere, it starts a global cycle and resides there for months or years (Clarkson, 1998). During this period, the Hg may not maintain the same form and it is transformed throughout its journey depending on the surroundings it encounters. Two main types of reaction in the Hg cycle determine the Hg transformation between the various forms: oxidation-reduction and methylation-demethylation (USEPA, 1997c). In oxidation-reduction reactions, Hg is oxidized to a higher valence state, for example, from Hg^0 to more reactive Hg^{2+}, and conversely Hg^{2+} is reduced to a low valence state. Likewise, in methylation processes, inorganic Hg can be transferred to and from MeHg by bio-reactions within bacteria. Figure 1 illustrates the Hg transport in various environmental media.



Source: Clarkson (1998)

Figure 1 Global cycling of Hg

More than 80% of gaseous Hg in the atmosphere is elemental Hg (Lindqvist and Rodhe, 1985, Goldblum et al., 2006, Saponaro et al., 2005). Because of its high volatility, elemental Hg travel in long range atmospheric transport for more than one year (Lindqvist and Rodhe, 1985, Wang et al., 2004, Clarkson et al., 2003a). In addition to gaseous elemental Hg, vapour forms of oxidised species of Hg and particulate Hg are present in the gaseous emissions (USEPA, 1997c). MeHg compounds are also detected in the atmosphere in very small amounts and with unclear origins (Clarkson, 1998). Gaseous Hg may be released naturally into the environment as a result of the reduction of Hg^{2+} by various mechanisms in water and soils or by artificial effluent from its widespread use in human activities. Winds transport Hg in the air and result in Hg being distributed throughout the global atmosphere (Fang et al., 2001).

Anthropogenic activities are the main source of Hg released into the atmosphere. Hg is widely used in the manufacture of dry batteries, fluorescent lamps, dental amalgam, alloying agent in gold mining, medical devices such as thermometers and sphygmomanometers, chlor-alkali industrials, tanning and leather industrials, pesticides among many others (Rivera et al., 2003, Zahir et al., 2005). Recent human activities such as metal smelting, coal combustion, chemical synthesis and use and waste disposal also produce a considerable amount of the Hg emission into the atmosphere (Clarkson et al., 2003a, Clarkson et al., 2003b, ATSDR, 1999a). As a result, Hg levels have increased dramatically and according to several studies around the world, modern deposition flux has been determined 3-24 times higher than pre-industrial flux (Wang et al., 2004, Heyvaert et al., 2000, Hermanson, 1998, Bindler, 2003).

Hg in the atmosphere can be deposited on lands or in water by wet and dry deposition. The wet deposition is by gaseous Hg^0 transforming to Hg^{2+} in both aqueous and particulate forms by cloud microphysics and precipitation. High atmospheric moisture content may increase the chance of the oxidation of Hg and speed up its deposition on land and aquatic systems (Lindqvist and Rodhe, 1985). This mechanism causes locally high levels of Hg (mainly Hg^{2+}) contamination, particularly in cold climates; most of the Hg being from anthropogenic sources. Deposition of gas phase Hg^{2+} is thought to be significant due to its reactivity with surface material (USEPA, 1997c). On the other hand, dry deposition of elemental Hg is seen on wet surfaces, but the quantity of the dry deposition is not comparable in magnitude to the cloud droplet mediated processes. Dry deposition is also likely to occur from ozone mediated oxidation although the process is not common (Clarkson, 1998).

Clarkson (1998) pointed out that Hg can form a stable monatomic gas whose characteristic is similar to the noble gases and its residence time in the atmosphere is of the order of a year or more. The author also added that, although Hg vapour is the main conduit for Hg transport, the actual concentrations in the air are low enough to be neglected as a contributor to overall human health. The gaseous Hg in the environment seems less harmful for the general public, except in contaminated working environments such as chlor-alkali plants. Nevertheless, cases of occupational or accidental exposures via the inhalation of Hg vapour still lead to undesirable effects on people.

Hg existing in surface soils is mostly ionic Hg^{2+}, 84-98% (Revis et al., 1990). Hg accumulates in soils generally via four routes - deposition of atmospheric Hg, agriculture activities, addition of sludge-amended soil and natural degradation of ferrallitic soils due to podzolisation (a natural process known to make the soil acidic) (Wang et al., 2004). Hg^{2+} has a strong affinity for sulphur-containing functional groups (USEPA, 1997c). Therefore, inorganic complexes of Hg are often seen in soils.

Divalent forms of Hg in soils are often linked with organic matter (mainly fulvic and humic acids) and mineral colloids, particularly the former, where it can be transformed to various oxidation states and compounds by both chemical and biological reactions. The consequence of various microbial processes acting on Hg^{2+} compounds under anerobic conditions is that it is likely to form MeHg, particularly in organic rich soils. The USEPA (1997c) estimated that the proportion of MeHg in surface soils is approximately 1-3% of the total Hg. However, the amount of MeHg varies with different physical and chemical properties of the soil. Cappon (1987) has demonstrated that MeHg is more frequently observed in certain soil conditions, such as garden soils with high organic content and under slightly acidic conditions.

Hg can also be passed to terrestrial plants. The presence of divalent Hg is thought to be readily transformed from the elemental Hg that is absorbed from the air and/or soils, but such transport between air and soils through plants is insignificant in terms of health risk concerns. The great proportion of Hg in plants is inorganic Hg^{2+} compounds, with little organic forms like MeHg. The fact that plants may have the ability of methylating Hg has been reported by the USEPA (1997c). Very high levels of MeHg in terrestrial plants are not commonly seen, except in highly polluted areas. For example, Horvat et al. (2003) found that in the Chinese province Guizhou, an area with very high levels of Hg production, there were high concentrations of MeHg up to 145 µg/kg, 25.5% of total Hg in the rice grains.

The water from rain, snow, or floods eroding and passing through Hg-containing soils is a major source of Hg pollution of aquatic resources. Hg is transported to groundwater systems from upper soil layers (Wang et al., 2004). As mentioned before, mercuric compounds are found bound to either dissolved organic carbon (FOC; consisting of fulvic and humic acids, carbohydrates, carboxylic acids, amino acids and hydrocarbons) or suspended particulate matter (Lindqvist and Rodhe, 1985, USEPA, 1997c). As a result, soils or sediments with abundant dissolved organic carbon elevate Hg contamination in water systems.

Hg can also be conveyed to water systems from the air. However, in contrast to the source of soil erosion, Hg deposition from the atmosphere occurs with a relatively low chance, except in cold climates.

The oceans contain vast amounts of Hg^{2+} compounds at low concentrations. When Hg is transported to the mixing layer at depths of less than 100 meters, the sediments and the water column near the oxycline area are suitable for methylation (USEPA, 1997c). The highest levels of Hg in the sea are found near or in estuaries. Most of the Hg in the sea is carried by polluted rivers from anthropogenic sources.

The Hg in water is methylated predominately through biotic processes (Ullrich et al., 2001). When the lipophilic MeHg is formed, it is bioaccumulated through the food web with concentrations increasing from microorganisms like plankton to herbivorous fish or insects and finally to the top fish predators, such as shark, swordfish, and to fish-eating mammals and biota (Clarkson, 2002, Clarkson et al., 2003a). People eat fish and this is the primary route of human exposure to the highly organic forms of Hg (WHO, 1990, USEPA, 1997f). In addition, Verdon et al. (1991) indicated that temporary or permanent flooding of vegetation and soils may potentially increase Hg mobilisation and consequently the concentrations in aquatic food chains. Such cases of Hg contamination in aquatic biota are particularly severe in recent flooded reservoirs.

2.1.2 Pathways of human exposure to Hg

The definition of human exposure to a chemical is ‘a condition of a chemical contacting the outer boundary of a human’ (USEPA, 1997a). The broad explanation of the outer boundary of the human body is the skin and the openings such as the mouth, nostrils, or punctures and lesions in the skin (Paustenbach, 2000). Exposure can be qualified and quantified referring to the intensity, frequency and duration of contact, the route of the chemical across the boundary (e.g. dermal, oral, or respiratory), the resulting amount of chemical actually crossing the boundary (dose) and the amount of chemical absorbed (internal dose) (USEPA, 1992a, Paustenbach, 2000).

Exposure pathways are the mechanisms by which humans are exposed to a polluted source. The USEPA (1989) defines that exposure points (points of potential direct contact with chemicals) and routes of exposure (e.g., ingestion, inhalation) are both considered as exposure pathways. Paustenbach (2000) identifies that the main pathways for human exposure to chemicals in the ambient environment are typically dust and vapour inhalation, dermal contact with contaminated soils or dust and ingestion of contaminated food, water, dust, or soils, whereas in the workplace, the predominant exposure route is usually inhalation, followed by dermal uptake and to a less extent dust ingestion due to hand-to-mouth contact. To date, the prevalent definition for exposure pathways is to classify them into direct and indirect. The following are examples of the most common exposure pathways (Nusslein et al., 1995, Valberg et al., 1996):

Direct:

a. Inhalation of airborne gases and particles, (if the following media are contaminated only by deposition of airborne emissions, they could be considered ‘indirect’ pathways rather than ‘direct’).

b. Incidental ingestion of contaminated soils, (the particle sizes greater than 10 µm in diameter (Department for the Environment Food and Rural Affairs (DEFRA), 2002a).

c. Ingestion of contaminated drinking water.

d. Incidental ingestion of contaminated surface water during recreation.

e. Contact with contaminated sediment.

f. Dermal absorption during recreational activities on contaminated soils or in contaminated surface waters.

Indirect:

a. Ingestion of locally grown vegetables (both above-ground and root vegetables).

b. Ingestion of locally produced dairy products (primarily milk).

c. Ingestion of meat products from meat and meat products.

f. Ingestion of fish (both finfish and shellfish).

g. Infant ingestion of mother's milk contaminated by mother's exposure to emissions (often not quantified due to uncertainly regarding relevant parameters).

The major direct exposure pathway that elemental Hg enters human bodies is inhalation, because elemental Hg is highly volatile and easily taken up in the mammalian respiratory tract. According to the ATSDR (1999b), vapour Hg can be largely absorbed by the lungs (about 80%). Compared with the gaseous form of Hg^0, the absorption of the liquid Hg^0 from the gastrointestinal index (GI) tract in most animal species is insignificant (Caussy et al., 2003). Although elemental Hg is found in plants due to the absorption through their foliages, this route of ingestion for Hg exposure is not common for general public and thus it is generally considered to be negligible (USEPA, 1997f, Wang et al., 2004).

Humans are most likely to be exposed to organic forms of Hg via ingestion (Nasreddine and Parent-Massin, 2002). Organic Hg compounds are readily absorbed by humans and animals via oral exposure, more than 80% (The Risk Assessment Information System (RAIS) website: http://rais.ornl.gov/tox/profiles/methyl_mercury_f_V1.shtml). Among the various forms of organic Hg, MeHg is the most commonly accumulated from aquatic food chains and hence MeHg poses a high level of risk to humans. It is therefore not surprising that the primary exposure pathway of human exposure to Hg is via fish and shellfish, or piscivorous mammals (usually sea mammals) and fish-eating-birds for some groups of people (USEPA, 1997d). MeHg is distributed to all tissues of the human body, including transport across the placenta to the fetus and through breast milk to the nursing infants. Hg exposure through the umbilical cord and breast milk can also be an important route in unborn and nursing children.

Inorganic salts of Hg are hardly absorbed (<10%) by gastrointestinal tract. In dietary uptake experiments using mice, soluble mercuric chloride was only one fifth uptake of MeHg and insoluble salt mercuric sulphide was much less (Schoof and Nielsen, 1997, Sin and Teh, 1992, Nielsen, 1992). As human exposure to Hg is likely to occur through the medium of soils, the ATSDR science panel integrates the bioavailability of Hg forms as 20-25% for mercuric chloride, nearly 100% for MeHg, 15% for mercuric nitrate, 0.01-0.5% for elemental Hg (Canady et al., 1997).

Unfortunately there is little data on the amount of uptake via dermal contact and data on the accurate absorption fractions of Hg compounds are scarce regardless human or animal experience. The defaults of 1% for organics and 0.1% for inorganics in general are adopted (The RAIS website: http://rais.ornl.gov/tox/profiles/methyl_mercury_f_V1.shtml).

2.1.3 Adverse health effects due to Hg exposure

Acute health effects of high concentrations of Hg vapour can give rise to severe pulmonary effects, sometimes leading to interstitial pneumonia. An exposure to high level Hg vapour of more than 1-2 mg/m^3 for a few hours was reported by Lilis et al. (1985) to cause acute mercurial pneumonitis in four patients. Metal fume fever may occur with symptoms similar to those of influenza, such as weakness, fatigue, anorexia, weight loss, and gastrointestinal disturbances (Clarkson, 1998, Tchounwou et al., 2003). Hand tremor and postural sway are also thought to occur as a result of exposure to elemental Hg (Iwata et al., 2007). Such exposure is now rare due to more careful control and usage. Nevertheless, subacute occupational poisoning because of elemental Hg sometimes occurs, typically giving rise to tremor, gingivitis and erethism (Clarkson, 1998).

Elemental Hg poses the greatest biohazard in contaminated workplaces, for example, the heating procedures used for its extraction from Hg-rich ores such as cinnabar (ATSDR, 1999a). Dentists are also in the high risk group as result of the use of Hg as a material in dental amalgam fillings (Ngim et al., 1992). A study reported that the dentists who worked in the environment of low levels of Hg for an average of 5.5 years performed less well in a battery of neuropsychological performance tests, compared with the control group of university employees. In addition to occupational cases, vapour Hg exposure is also frequently found in school science laboratories due to careless spills.

Inorganic mercuric complexes are likely to be directly accumulated in human bodies via ingestion or via conversion from other forms of Hg. Both metallic and organic Hg compounds are found to be oxidised to inorganic Hg in the liver. The principal syndrome of acute Hg salt poisoning is stomatitis or digestive upset. Hg salts are particularly toxic to the kidneys, causing acute tubular necrosis, immunologic glomerulonephritis, or nephrotic syndrome. According to the WHO (1991), the fatal levels of inorganic Hg are 10-14 ng/g in the kidneys. The half-life of mercurial salts is about 40 days in the body of an adult (Tchounwou et al., 2003).

Inorganic Hg transformation *in vivo* allows inorganic Hg to pass into the fetus via the placenta. A study of pregnant Swedish women showed that the median of inorganic Hg levels in the placenta at the time of delivery was four times higher (up to 7 µg/kg bw) than that in the maternal blood circulation (Ask et al., 2002).

Among organic Hg compounds, MeHg is the most hazardous to human health because it is almost 100% absorbed by human bodies (USEPA, 1997d, Clarkson et al., 2003a). When MeHg enters the body, it can stay in the blood of an adult with the mean half-life of 50-72 days (Aberg et al., 1969, Al-Shahristani and Shihab, 1974, Sherlock et al., 1984). Exposure of an adult to MeHg results in regional destruction of neurons in the visual cortex and cerebellar granule cells in the nervous system (Clarkson et al., 2003b). The neural problems includes visual-field constriction, ataxia, mental retardation, cerebral palsy, seizures. Ultimately death may occur with the fatal dose of MeHg ranged from 20 to 60 µg/g or the blood Hg level > 300 µg/L for 70kg-person (Bittner et al., 1998, WHO, 1990, Clarkson et al., 2003b). Several studies have also reported the statistical associations between cardiovascular disease and MeHg exposure (Clarkson et al., 2003b, Salonen et al., 1995, Stern, 2005).

Furthermore, damage to brain development was noted in infants if mothers encountered too much MeHg during their prenatal or postnatal periods (Albers et al., 1988, Choi et al., 1978, Tchounwou et al., 2003). It has been recognised that perinatal Hg burden of women is passed to the fetus (Chien et al., 2006, National Research Council (NRC), 2000).The clinical features include constriction of visual fields and hearing impairment (Murata et al., 1999, WHO, 1990). Similar to adults, the developing nervous system of children is vulnerable to MeHg. Risk to children of MeHg exposure is particularly acute during prenatal and postnatal periods when its toxic effect on the developing central nervous system can be severe (WHO, 1990, USEPA, 1997e, NRC, 2000, Davidson et al., 1998). The effects of MeHg on the exposed group may range from subtle delays of cognitive and motor development to cerebral palsy, depending on the amount and the period during the pregnancy that the MeHg is ingested by the mothers (Marsh et al., 1995a, Boischio and Henshel, 2000b). In addition to neurological development problems, Matamba et al. (2006) also indicated that Hg exposure resulted in the impairment of children's nervous system and immune and reproductive systems.

2.1.4 History of human exposure to Hg

2.1.4.1 Minamata Disease in Japan

Minamata Disease, which was the first recognised case of MeHg poisoning via bioaccumulation in aquatic food chains, was first noted in 1953 and the "Second Minamata" disease epidemic broke out in the Agano River region in Niigata Prefecture in Japan in 1965. The contaminated source was the Chisso acetaldehyde production plant, where inorganic Hg was used as a catalyst. The chemical plant drained the Hg-containing sludge and waste to a neighbouring river and eventually into Minamata Bay, causing Hg pollution in the environment and through the food chain resulted in MeHg accumulation in fish and other marine creatures (Harada, 1995, Rivera et al., 2003). The Japanese health authorities in Minamata had been aware for some time that fishermen and their families around Minamata Bay were suffering from a neurological disease exhibiting signs of incoordination, constricted visual fields, and numbness in the extremities. By later 1956, the research group determined the affected population was being poisoned by the ingestion of heavily

Hg-polluted fish and shellfish from the bay (Gochfeld, 2003). The Chisso company and the government undertook countermeasures. In 1959, the results of environmental monitoring showed that there were extraordinarily high concentrations of Hg, up to 2010 ppm in the mud near the drainage channels of the plant, and the Hg levels gradually declined with increased distances from the channel. Fish and shellfish in Minamata Bay were also shown to have high Hg contents, up to 11.4-29.0 ppm (Rivera et al., 2003). In 1962, it was proven that the methylation of the inorganic Hg in the wastewater from the acetaldehyde manufacturing process was the cause and in 1968 Minamata Disease was officially identified as MeHg poisoning.

The local people were exposed to MeHg by ingestion of Hg-contaminated fish for almost twenty years (at least from 1950 to 1968). The total Hg in the samples of 1644 residents of the coastal areas ranged from 0 to 920 µg/g with a median value of 23.4 µg/g in 1960, when the MeHg pollution reached a maximum (Ninomiya et al., 2005). The clinical symptoms of the acute poisoning cases included visual and hearing impairments, olfactory and gustatory disturbances, ataxic gait, clumsiness of the hands, dysarthria, and somatosensory and psychiatric disorders such as sensory changes, irritability and nervousness (Ekino et al., 2007). Moreover, fetuses were vulnerable due to maternal exposure to Hg. The exposed children were diagnosed to have blood-brain battier which resulted in 'infantile cerebral palsy' through the placental barriers. Those who survived became susceptible to other disease, like particularly pneumonia, which was the primary cause of death. There have been more than 200,000 suspected cases of MeHg poisoning since the initial outbreak of Minamata Disease in 1953. To date, about 17,000 people had applied for the recognition that they were poisoned but only 2264 people were recognised as victims of Minamata Disease, making them eligible for a variety of health benefits (Ekino et al., 2007).

2.1.4.2 Hg poisoning in Iraq

Another widespread Hg poisoning episode occurred in Iraq during the winter of 1971-1972. Wheat seeds intended for crop planting, which had been treated with MeHg as a fungicide, were distributed free in the rural area. The seeds were ground into flour by some farmers, baked into bread and consumed. As a result, there were

459 deaths and 6530 cases admitted to hospital with Hg poisoning, according to the official figures (Clarkson, 1998).

The susceptibility of the fetuses to in-utero exposure to MeHg was studied in this outbreak. MeHg was reported to inhibit neuronal migration of parentally exposed infants (Choi et al., 1978). Children with delayed motor development were found and the levels of Hg in maternal scalp hair during pregnancy were determined. It was established that Hg levels of 10-20 µg/g in mothers' hair indicated a relative high risk of having psychomotor retardation in the offspring (Rivera et al., 2003). Clarkson (1998) indicated that it was probably the first establishment of the relationship between peak concentrations of Hg in maternal hair during pregnancy and neurodevelopment impacts on infants.

The series of studies associated with prenatal exposure in Iraqi Hg poisoning case provided a new implication on the view of clinical and pathological human exposure to Hg. As a result, the reference dose (RfD) of 0.1 µg/kg body weight/day has been developed by the USEPA (1997d) for a 70-kg adult from the previous level of 0.3 µg/kg body weight/day, in order to sufficiently protect the most sensitive endpoint-fetal neurobehavioural development (Gochfeld, 2003).

2.1.5 Hg considered as a non-carcinogen

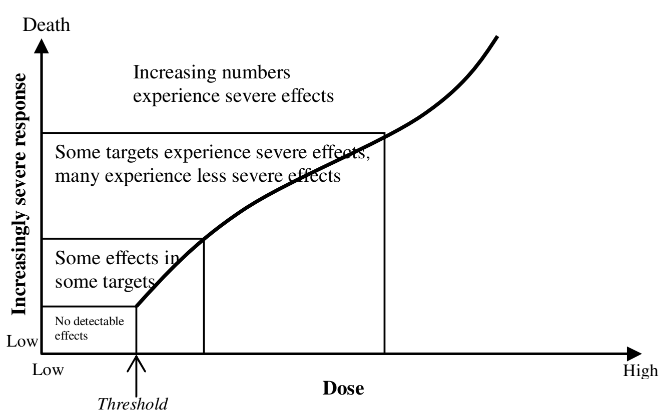
The USEPA has identified the toxicology of Hg for human health impacts by categorising the likelihood of getting cancer by a weight-of-evidence ranking. Elemental Hg is categorised as Group 'D' for little evidence of carcinogenicity. Inorganic and organic Hg are categorised as Group 'C', which indicates weak evidence from human and animal data that the chemicals have carcinogenic potential to humans (USEPA, 1997e). These three forms of Hg are noted as 'not likely to be a human carcinogen under conditions of exposure generally encountered in the environment'.

2.2 Assessment of Hg exposure risk using biomarker data

2.2.1 Exposure risk assessed using the dose-response relationship

Risk embodies the concepts of both severity of outcome and probability of outcome (Valberg et al., 1996). Assessing exposure risks consists of the estimation of the likelihood of harm and the severity of the consequence of some adverse health effects resulting from a certain level of exposure to a chemical for a certain length of time (Caussy et al., 2003, Holt et al., 2000). Currently, most risk assessments associated with human exposure to environmental chemicals rely upon a dose-response relationship. A dose-response relationship can be seen as a quantitative relationship between the internal dose of an environmental chemical and the intensity or occurrence of an adverse health effect resulting from the chemical (Petts et al., 1997).

Figure 2 depicts a typical dose-response curve, where dose is the amount of chemical that has been absorbed and is available for an interaction with a biologically significant receptor (e.g. target organs) (Paustenbach, 2000). There is a threshold level for exposure doses below which there is no detectable effect. Inversely, above this level, the adverse impacts become more severe. Note that the adverse impacts are very chemical specific. The risk effect is usually regulated at a level of one observable adverse effect per million cases and is expressed on a per-kg-body-weight basis (i.e., mg of chemical intake per kg body weight per day). The dose-response curve assumes that toxicity is dependent on lifetime average daily dose, not on dose rate or on dose distribution over a lifetime. Therefore, the dose-response curve may not be adaptable to describe some exposure cases where a very high dose may induce adverse effects by a single acute exposure, but exposure to the lower dose levels for a longer period of time may not be toxic (Valberg et al., 1996).



Source: Petts et al. (1997)

Figure 2 Curve of dose-response relationship

2.2.2 Biomarkers of Hg exposure

Biomarkers, the short name of biological markers, are sometimes called internal indicators, because they are able to present the past exposure rates of chemicals. Biological tissues or sera, such as blood, urine, breath, hair, nail, milk and adipose are commonly taken from humans and wildlife to reflect the absorbed fraction of the chemical that has successfully crossed physiological barriers to enter the organism (Nieuwenhuijsen et al., 2006). Biomarkers are used to quantify body burdens of hazardous chemicals only when the relationships between internal doses and the chemicals' levels in the biomarkers have been known (Paustenbach, 2000). The degrees of exposure shown via biomarkers demonstrate past exposure with an integration of exposure through all routes and sources. Biomarker exposure data have been thought to represent levels of exposure of individuals (Ponce et al., 1998).

Laboratory techniques have improved rapidly in recent decades to detect minimal amount of a chemical in biomarkers. However, there are limitations in the use of biomarkers. In the consideration of cost, the acquirement of biomarker data is sometimes expensive so that such data are only available for a few chemicals (Paustenbach, 2000). Since biomarker data are regarded as overall exposure levels in individuals, it may be difficult to understand whether the presence of the biomarker exposure is due to the hazard in ambient air, water, or diet (Valberg et al., 1996). Moreover, proportions of actual exposure doses being reflected via biomarker measurement have been reported to vary due to inhomogeneous biological characteristics of absorption and excretion among individuals (Nieuwenhuijsen et al., 2006, NRC, 1994).

Hg has been accurately measured in blood, hair, nails, urine and so on. Different Hg forms have been tested in the varied biomarkers. Cernichiari et al. (1995) reported that MeHg accounted for more than 80% of the total Hg in human hair and Barbosa et al. (2001) had a similar finding with a mean fraction of 71.3% of MeHg to the total Hg in hair. High fractions of MeHg to total Hg in hair are due to MeHg partly excreted through hair. Elemental and inorganic Hg, on the other hand, are normally found in urine and faeces (Johnsson et al., 2005, WHO, 1990, Malm et al., 1997, Carrier et al., 2001). Recognising the forms of Hg in those biomarkers is necessary in order to clearly identify the levels of human exposure to Hg and the exposure pathways.

A number of laboratory studies have found that Hg concentration in hair is proportional to its concentration in the blood (WHO, 1990, Lipfert, 1997, Phelps et al., 1980, McDowell et al., 2004, Johnsson et al., 2005). The WHO (1990) and the USEPA (1997e) reviewed the studies associated with the hair Hg (µg/g) and blood Hg (µg/mL) ratios within different exposure groups and suggested the ranges from 140: 1 to 370:1, a difference of about a factor of 3. Variability in the hair-blood relationship for Hg concentration is thought to result from the fact that unsegmented hair analysis gives a time-weighted average of Hg exposure, while analysis of Hg in blood reflects a much shorter period average of exposure (USEPA, 1997e). The widely used converse ratio at present is 250 studied by Ginsberg (2000) and Hightower et al. (2003), as the USEPA (1997e) concludes that 250 is a midpoint value and is acceptable for the purpose of estimating average blood levels in the Iraqi population. This ratio is also applied to derive the reference dose (RfD). Concentrations of the form of MeHg in blood have also been determined to have highly positive correlations with Hg concentrations in hair (IP et al., 2004, Lindberg et al., 2004, Sherlock et al., 1982).

Hg concentrations in blood and Hg concentrations in hair can be used as effective bio-indicators of Hg exposure, particularly MeHg exposure (WHO, 1990, Cernichiari et al., 1995, Grandjean et al., 1994). In comparison with blood samples, hair samples are easier to collect, to store and to transport and are less invasive to people (Dermelj et al., 1987). Hg concentrations in hair also reflect an integrated and long-term relationship with Hg exposure, based on the Hg levels tested from the different segments of the hair (Lipfert, 1997, WHO, 1990, Phelps et al., 1980). However, heterogeneity among individuals such as the speed of hair growth and the excretion rate of Hg yielded the variability to the levels of human exposure reflected by hair Hg concentrations. The hair growth rates conducted by Al-Shahristani and Shihab (1974) varied slightly from 0.9 to 1.5 cm per month, whereas the hair growth rate of about 1 cm per 30 days was defined by Grandjean et al. (2002). In other words, individual physio-characteristics contributed to variability in Hg exposure levels tested from Hg concentrations in hair.

2.2.3 One-compartment model to quantify Hg exposure doses from biomarker data

An empirical one-compartment kinetic model (Equation 1) has been developed to describe the relation of average daily Hg ingestion rates and Hg concentrations in blood (USEPA, 1997d, WHO, 1990). The USEPA (1997e) has applied the one-compartment model on Hg exposure research and found a reasonably good fit between Hg level changes in humans during and after consumption of MeHg-contaminated fish, while human kinetics for Hg was assumed to be steady-state. The one-compartment model is also a common tool to estimate the corresponding Hg average doses by tracing back from Hg levels in biomarkers. Many health organisations such as the WHO (1990) and the IRIS at the USEPA (1995) have used this pharmacokinetic model to develop acceptable Hg intake doses by converting Hg concentrations in hair to Hg concentrations in blood when the conversion ratio of Hg concentration in hair to in blood is determined. More recently, this model was also used by Yasutake et al. (2004) to estimate the risk of neurotoxic effects due to MeHg exposure to the populations in Japan, on the basis of the consumption rates of fish and shellfish of the study population.

$$d = \frac{c \times b \times V}{A \times f \times bw}$$

Equation 1

where:

d = daily Hg intake dose (µg/kg body weight/day)

c = Hg concentration in blood (µg/L)

b = elimination constant ($days^{-1}$)

V = volume of blood in the body (L)

A = absorption factor (dimensionless)

f = faction of daily intake taken up by blood (dimensionless)

bw = body weight (kg)

The parameters in the one-compartment pharmacokinetic model are often treated to be steady-state. In reality, however, the ratios of the biological parameters in this model vary within individuals. To examine the variation of the factors in the one-compartment model, several studies used distributed data as the inputs to demonstrate the physiological variability among individuals (Swartout and Rice, 2000, Stern et al., 2002, Stern, 1997). Swartout and Rice (2000) established a probabilistic model using the Monte Carlo technique and concluded that the variable of hair/blood ratio of Hg accounted for the most variance of the output in the one-compartment model. Swartout and Rice (2000) also acknowledged that the personal physiological characteristics were difficult to collect. Therefore, the personal physiological data were taken from published ranges, not individual-specific data which may possibly generate uncertainties in the application of the probabilistic model.

2.2.4 Critical levels of Hg exposure in biomarkers

2.2.4.1 Related studies to develop critical levels of Hg exposure

On the basis of the findings from the severe poisoning episodes in Japan and Iraq, a hair Hg concentration of 50 µg/g is associated with approximately a 5% risk of paresthesias in adults (Mahaffey, 2000). However, several more recent studies regarding Hg as a devastating neurotoxic agent to humans have reported that the harmful impacts on human health occur at lower doses of Hg. In the studies for Amazonian adult population, the impact of chronic low-doses of MeHg exposure below 50 µg/g in hair was defined as causing vision impairment (reduced contrast sensitivity and reduced peripheral vision) and neuromotor effects on adults (Lebel et al., 1998, Lebel et al., 1996). Herada et al. (1999) indicated that neurological symptoms particularly sensory disturbances such as glove and stocking types were seen at hair Hg levels of less than 10 µg/g as a result of a long term produced low Hg exposure through fish eating. The studies carried out in the Cree Indians had similar evidence with respect to low levels of Hg exposure resulting in neurological impairment. The exposure group had significantly different indices of static tremor and kinetic tremor and different performance on a test of rapid and precise proximodistal movements from the control group with hair Hg levels greater than 24 µg/g (Beuter et al., 1999b, Beuter et al., 1999a, Beuter and Edwards, 1998).

As the neurodevelopment effects may occur in fetuses due to prenatal and postnatal Hg exposure, women's hair Hg concentrations are critical indicators during periods of pregnancy and breast milk feeding. A study associated with the case of Hg poisoning in Iraq indicated that there were adverse fetal effects when mothers' hair Hg concentrations were as low as 20 µg/g (Marsh et al., 1995b). In 1977-1978, a study in New Zealand emphasised the effects of *in utero* exposure to MeHg from maternal fish consumption. The study found that a statistically significant decrease in test performance on Denver Development Screening Test Scores within children at age both four and six years whose mothers were found to have MeHg concentrations in hair exceeding 6 µg/g. Moreover, two large longitudinal studies associated with children exposed to Hg were carried out in the Republic of Seychelles and the Faroe Islands. The Seychelles studies reported that no adverse effects on child development were seen in the mother group with the mean Hg concentration of 6.8 µg/g in hair and the children at 66 months of age, with the mean Hg concentration of 6.5 µg/g (Davidson et al., 1998). By contrast, among the Faroese cohort, maternal hair Hg concentrations as low as 3-10 µg/g were related to children's neuropsychological dysfunction. The adverse impact was most pronounced in the domains of language, attention and memory and to a lesser extent in visuospatial and motor functions (Mahaffey, 2000, Myers et al., 2000, Grandjean et al., 1998, Grandjean et al., 1997).

These findings support the view that unborn children are affected via mothers exposed to low Hg levels. Therefore, the tolerable limit of daily Hg intake has been revised for women of childbearing age and infants, the group most sensitive to Hg expose (Tchounwou et al., 2003). Some health authorities like the United States Food and Drug Administration (USFDA) (The USFDA referencing system http://www.cfsan.fda.gov/~lrd/tphgfish.html), the European Food Safety Authority (2004) and Swedish National Food Administration (Johnsson et al., 2005) have set advice on the safety levels of fish consumption particularly for pregnant and lactating women.

2.2.4.2 Critical levels for humans exposure to Hg

The reference dose (RfD) is defined by the USEPA (2001a) as 'an estimate (with uncertainty spanning perhaps an order of magnitude) of a daily exposure to the human population (including sensitive subgroups) that is likely to be without an appreciable risk of deleterious effects during a lifetime'. The RfD for MeHg is set as 0.1 µg/kg body weight/day with an uncertainty factor of 10, based on the study reported by Marsh et al. (1987) for Iraqi children of *in utero* exposure to MeHg. The RfD for mercuric chloride is 0.3 µg/kg body weight/day with an uncertainty factor of 1000, based on rat feeding studies. No RfD for elemental Hg is defined but the reference concentration (RfC) for Hg inhalation is 0.0003 mg/m^3 (USEPA, 1997e). Although a number of states in the US develop two-tier fish consumption advisories in order to protect the more sensitive population including women of childbearing age and young children, the USEPA still maintains the single recommended level, since some studies have also found cardiovascular effects in men with Hg concentrations in hair as low as below 3 µg/kg (Rice, 2004).

The United States Food and Drug Administration (USFDA) uses an action level based on consideration of the tolerable daily intake (TDI) for MeHg and information on seafood consumption and associated exposure to MeHg. The TDI is interpreted as the amount of MeHg that can be consumed daily over a long period of time with a reasonable certainty of no harm to adults. The USFDA in cooperation with the WHO develops a TDI with a weekly tolerance of 0.3 mg of total Hg per person, of which no more than 0.2 mg should be presented as MeHg. These amounts are equivalent to 5 µg/kg body weight/per week and 3.3 µg/kg body weight/per week (0.47 µg/kg body weight/day), respectively. For a 70-kg person, the TDI for MeHg would thus correspond approximately to an intake of 33 µg/day, or 230 µg/week. Nevertheless, due to the uncertainties associated with the Iraqi study, the USFDA has chosen not to use that study as a basis for revising its action level at present (USEPA, 2001b). Instead, the USFDA suggests consumption of no more than 12 ounces of cooked fish on average per week as a safety level and to select a variety of fish species but not large fish species including shark, swordfish, king mackerel and tilefish (USFDA referencing system: http://www.cfsan.fda.gov/~lrd/tphgfish.html). No harm is expected with this portion of fish unless high levels of Hg are found in fish.

The ATSDR first published a Minimal Risk Level (MRL) for Hg in 1993. The MRL which was derived from the concept of the RfD can be defined as an estimate of the daily human exposure to a hazardous substance that is likely to be without appreciable risk of adverse non-cancer health effects over a specified duration of exposure (USEPA, 2001b). Goldblum et al. (2006) added that the MRL is calculated to ensure a substantial margin of safety. In 1999, the ATSDR (1999a) adopted the Seychelles Islands study (Davidson et al., 1998) about the correlation between subtle neurological effects and low-dose chronic exposure to MeHg and revised the MRL of MeHg. The ATSDR consequently established a MRL of 0.3 μg/kg body weight/day, based on an acceptable dose of 1.3 μg/kg body weight/day which reflects the average concentration of the upper quintile of the exposed population but does not necessarily correspond to a no-observed-adverse-effect level (NOAEL).

More recently, in the 61st meeting of the Joint FAO/WHO Expert Committee on Food Additives (JECFA) (2003), a provisional tolerable weekly intake (PTWI) of 1.6 μg/kg body weight/week for MeHg was recommended. The collection of acceptable Hg exposure for non-carcinogenic oral criteria doses by governments or international agencies is showed in Table 1.

Table 1 Non-carcinogenic oral criteria of acceptable and maximum doses for human exposure to Hg

Agency	Hg compounds	Acceptable level μg/kg body weight/day	Uncertainty factor	Referred study
USEPA	Total Hg	<0.1	10	Iraq study
USEPA	MeHg	0.1	10	Iraq study
USEPA	Mercury chloride	0.3	1000	Iraq study
USFDA	MeHg	0.47	10	Waiting for Seychelles study
USFDA	Total Hg	0.5	10	Waiting for Seychelles study
ASTDR	MeHg	0.3	4.5	Seychelles study
JECFA	Total Hg	5 μg/kg bw/week	10	Seychelles and other studies
JECFA	MeHg	3.3 μg/kg bw/week	10	Seychelles and other studies
Canadian Council of Ministers of the Environment (CCME)	Total Hg	0.2	5	Seychelles, New Zealand

Source: Revised from the NRC (2000)

The concept of ‘uncertainty factor’ initially commented by the USEPA is applied during the development of threshold levels including oral RfD and inhalation RfC to compensate for the insufficient data in risk assessments of chronic exposure. Because the RfD and RfC are estimates derived from a lowest-observed-adverse-effect-level (LOAEL) or no-observed-adverse-effect-level (NOAEL), uncertainties result from the insufficient data. Hence, an uncertainty factor is applied to cover the uncertainties inherent in the fields consisting of a) average human to sensitive human, b) animal laboratory data to human, c) subchronic to chronic effects, and d) lowest-observed-adverse-effect-level (LOAEL) to no-observed-adverse-effect-level (NOAEL) (Hattis et al., 1996, USEPA, 1997e). The range of the uncertainty is assumed to be distributed log normally. The uncertainty factor of 10 is typically used to derive the RfD and considered to have uncertainty spanning at least an order of magnitude (Dourson et al., 1996). The larger uncertainty factors, the more overprotective the subthreshold doses for harmful exposure are likely to be.

The RfD of 0.1 μg/kg/day for MeHg exposure developed by the USEPA (2006) is based on the benchmark dose and the consideration of the uncertainty factor. The benchmark dose procedure is based on developmental neurological abnormalities in infants exposed *in utero* as the crucial health effect in the human epidemiological studies. However, the average daily intake rates were not wstablished through the measurement of Hg concentrations in children’s hair samples but the mothers’. The RfD for MeHg was therefore divided the estimated intake dose of 1.1 μg/kgbody weight/day by the uncertainty factor of 10, where 1.1 μg/kgbody weight/day was the benchmark dose, lower 95% confidence limit on the dose associated with a 10% extra risk) (USEPA, 1997e, Dourson et al., 1996). This uncertainty factor in the exposure research relates to two generations and actually contains two parts. The first part of the three-fold uncertainty factor was a standard ten-fold uncertainty factor by half and it was applied for interindividual variability in the toxicokinetic field, including known variation in the biological half-life of MeHg and variation in the hair/blood ratio of Hg. Another part of the three-fold uncertainty factor was due to possible postdevelopment sequelae and the lack of a two-generation reproductivity study (USEPA, 1997e, Dourson et al., 1996, Stern, 1997).

2.2.5 Factors in predictive models for Hg concentrations in hair

2.2.5.1 Studies of fish and shellfish consumption and increased hair Hg

A positive correlation between hair Hg levels and fish and shellfish consumption has been demonstrated by several studies (Airey, 1983a, Babi et al., 2000, Birke et al., 1972, Dickman and Leung, 1998, Hightower and Moore, 2003, Johnsson et al., 2004, McDowell et al., 2004, Morrissette et al., 2004, Sherlock and Quinn, 1988, Shipp et al., 2000, Tran et al., 2004, Lipfert, 1997). Frequent fish eaters were found to have elevated Hg levels in hair compared to those consuming less fish.

Varied levels of Hg exposure were associated with consumption of different fish species (Lebel et al., 1997, Johnsson et al., 2004, Boischio and Henshel, 2000a, Kehrig and Malm, 1999, Luk and Wai, 2006). The prominent study was conducted by Weihe et al. (2005) for whale consumers and fish consumers in the Faroe Islands. They found that women consuming whale meat had Hg concentrations in their hair about 60% higher than those living in the community without local access to whale meat. This was because the whale is high up the food chain. Fish species, together with Hg contents in various fish species, should therefore be taken into account when the influence of fish consumption is investigated in human exposure studies.

In order to protect fetuses and children from the adverse health effects of fish consumption, the Swedish National Food Administration had issued dietary intake recommendations for freshwater fish species for pregnant and lactating women. Bjornberg et al. (2003) found that Hg levels in some women's hair were also locally elevated when they consumed fish species that had been identified as problematic by the local authorities but high Hg concentrations were also observed in hair of the pregnant women who had consumed deep-frozen fish (e.g. cod, saithe), pickled herring and fresh marine fish.

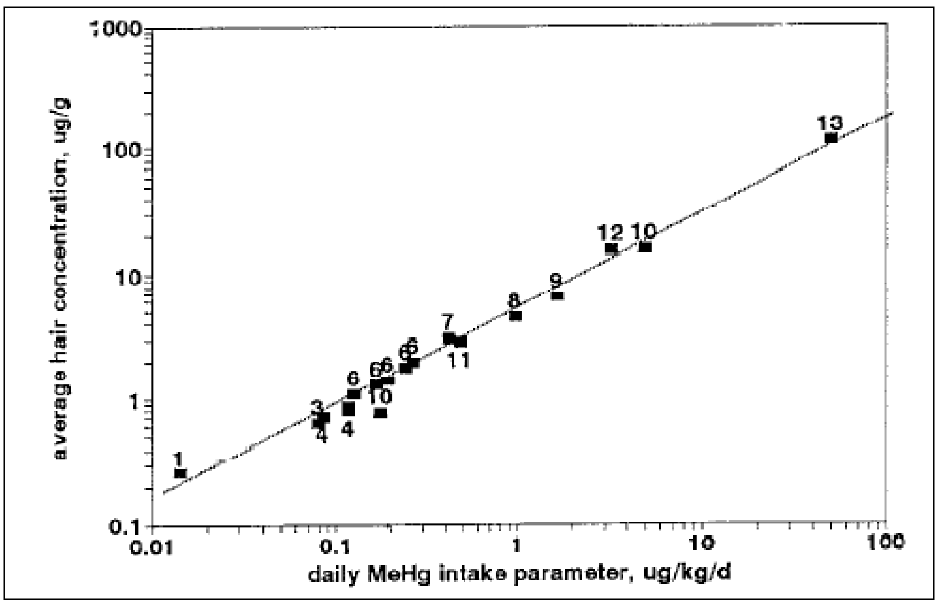
Table 2 summarises the models developed in the above studies between hair Hg levels and Hg intake via fish and shellfish consumption. The fact that increasing intake rates or frequencies of fish and shellfish elevated Hg levels in hair was consistently reported by studies. Fish and shellfish consumption significantly accounted for the majority of variance of Hg concentrations in hair in the study of Holsbeek et al. (1996) who found that the monthly fish consumption of the population accounted for approximately 77% of variation of their Hg levels in hair. These study results implicated fish and shellfish consumption as the key driver of elevated Hg concentrations in hair. The relationships between Hg concentrations in hair and Hg intake via aquatic food chains were described with either linear or non-linear equations for different exposed groups.

Table 2 Previous correlations and models for hair Hg and fish consumption

Study site (Reference)	Subject	Methodology	Model: Dependent variable	Model: Independent variable	Remark
13 countries (Airey, 1983b)	43 locations	-	Hair Hg	Fish consumption; GNP	a. Correlation between daily Hg intake and hair Hg = 0.54** b. R^2 = 0.694**
Bangladesh (Holsbeek et al., 1996)	219 males (13-69 years)	Unpublished Bangladeshi statistical data	Hair Hg	Monthly fish consumption	a. r = 0.88*** b. R^2 =0.773
Brazilian Amazon (Passos et al., 2003)	26 adult women	Daily food diary, questionnaire	Hair Hg	Annual fish consumption***; annual banana consumption**	a. R^2 =0.50*
Brazilian Amazon, non-impacted communities (Santos et al., 2002)	321 individuals	Questionnaire	Hair Hg	Weekly fish intake	a. r = 0.15*** b. R^2 =0.033
China (IP et al., 2004)	137 children (4-11 years)	Questionnaire	Hair Hg	Fish consumption frequency	a. r = 0.51***
Canada (Legrand et al., 2005)	91 people	FFQ and 24-h recall questionnaire	Log(hair Hg)	Daily Hg intake	a. r = 0.47**
Japan (Dakeishi et al., 2005)	327 mothers	FFQ	Log(hair Hg)	Log(daily Hg intake)***; artificial hair-waving***; hair colouring/dyeing; residence; working status; age	a. Correlation between daily Hg intake and hair Hg = 0.25*** b. No R^2 info.
Faroe Islands (Budtz-Jorgensen et al., 2007)	1022 mothers	Questionnaire	Log(hair Hg)	Log(Fish dinner frequency)	a. r = 0.25***
Sweden (Bjornberg et al., 2003)	123 pregnant women	FFQ	Log(hair Hg)	Consumption of deep-frozen fish, pickled herring and fresh marine fish	a. r = 0.2-0.3* b. R^2 =0.2***

*: significant level < 0.05
**: significant level < 0.01
***: significant level < 0.001

Lipfert (1997) summarised these finding in his comprehensive review. His studies of human exposure to Hg in different contaminated sites argued exposure levels spanning almost four orders of magnitude in total Hg intake but only three orders in hair Hg concentrations (Figure 3). The diversity of models for predicting Hg concentrations in hair may imply that the dose-response relationships are specific and heterogeneous among the Hg-exposed populations. On the other hand, the varied mathematical models or relationships could indicate originated uncertainties due to dietary data. People may report incorrect dietary data, mainly because of recall difficulty, in the investigations. Such data error problems have been observed and reported by several related studies (Berry, 1997, IP et al., 2004, Tran et al., 2004).



Data number 1-13: referring to Table II in Lipfert (1997)

Figure 3 Relationship between average hair Hg concentrations from various epidemiological studies of fish-eating populations and their respective average Hg dietary intake rates

2.2.5.2 Studies of other factors influencing Hg levels in hair

Gender

The finding of higher Hg levels in the scalp hair of males than females was consistently demonstrated in several studies (Foo et al., 1988, Yasutake et al., 2003, Johnsson et al., 2004, Lee et al., 2000, Airey, 1983b, Barbosa and Dorea, 1998a, Barbosa et al., 2001, Barbosa et al., 1998b, Knobeloch et al., 2007, Shimomura et al., 1980). High Hg levels in males' hair are generally thought to be as a result of higher amounts of food consumed by males. However, Airey (1983a) explained the finding from a biological point of view. Lower levels of Hg in women's hair are potentially due to females having a shorter biological half life of Hg than in males, as a result of the fact that a woman monthly loss blood may create distinct hormonal and biochemical mechanisms from men and result in lower levels of Hg exposure in women than in men.

Barbosa and Dorea (1998a) and Barbosa et al. (1998b) stated a different explanation for lower Hg levels in women's hair. The authors collected the maternal hair in a polluted area in the Amazon basin and demonstrated the reduction of the Hg concentrations in the women's hair during pregnancy and lactation. Hg appeared to be transferred to the infants via the umbilical cord or breast milk. As a result, the study teams suggested that childbearing women should consume small amounts of fish and shellfish.

On the other hand, a study (Yasutake et al., 2004) in Japan reported that the differences of hair Hg levels between genders were partly attributed to Japanese women's hair being artificial waved. Some Hg in females' hair was removed during the waving process as a result of thioglycolate in the lotion. Nevertheless, after adjusting for this factor, the male population in that study still had higher levels of Hg than the female population.

Age

Hair Hg concentrations have been found to vary between different age groups. Higher Hg concentrations tended to be found in tests on the hair samples of older populations than younger ones (Agusa et al., 2005, Dickman and Leung, 1998). A survey for the adult population carried out by the USEPA Mercury Study Report to Congress found that higher Hg levels in blood were seen in people older than 45 years compared with the population aged 15- to 44-year-old due to the higher fish consumption of the older people (Mahaffey and Rice, 1996). A study of the population in Wujiazhan, Northeast China observed that the people aged 46-55 years old had the higher concentrations of Hg in their hair (Li et al., 2006). The reason for the greater

Hg accumulation in the older population was considered to be likely to be low Hg excretion rate in old people.

Similarly, the study in Japan reported that Hg concentrations in the hair of males aged in their 50s or 60s were almost twice as high as children (Yasutake et al., 2004). Nagakawa (1995) explained that the varied Hg exposure levels in hair among different generations was likely to be associated with dietary behaviour. In Japan, the elderly people ate traditional meals with a lot of fish, whereas the younger population preferred more western food with less amounts of fish.

However, increased hair Hg concentrations did not always yield a consistent correlation with the increasing ages of individuals or groups. Li et al. (2006) discovered that the positive correlation between age and Hg levels in hair was found only in the population below age 30. Yasutake et al. (2004) demonstrated that Hg levels in hair of the population aged 50s and 60s were higher than those in the hair of the children, but the finding was only seen in the male subgroup.

Contrarily, De Oliveira Santos et al. (2000) found hair Hg levels in the elderly group were lower than the juvenile group. The study for the Amazon communities close to gold mining areas indicated that individuals under 25 years old had significantly higher hair Hg than other age groups due to *in utero* exposure. In this case, chronic exposure for young people played the important role in the severity of Hg exposure.

Proximity to Hg sources

Regional differences on Hg concentrations in hair have been seen in epidemiological studies. Populations living in the areas adjacent to contaminated sources tended to have higher Hg concentrations in hair than those living further away from polluted sites (De Oliveira Santos et al., 2000, Al-Shahristani and Alhaddad, 1973, Harada, 1995, Suckcharoen et al., 1978). These results seemed reasonable as the on-site residents were likely to be exposed to the polluted media during their daily activities. With respect to this plausible result, however, Airey (1983a) suggested that dietary behaviour like fish consumption was still the key factor causing the differences of hair Hg levels between locations. In other words, the population with high Hg concentrations in their hair were likely to be exposed to Hg via frequent contaminated fish consumption in their diet rather than via other contaminated environmental media in their residences. Agusa et al. (2005) also discovered that the geographic distance from the polluted source to the exposed area was less important in terms of the influence on human exposure to Hg. The authors added that a number of factors may exist behind the ‘distance’ factor, such as Hg transformation in environmental media and diverse ethnicities or dietary behaviour of the different inhabitations.

In short, geographic location may provide an initiative to investigate the levels of Hg exposure for the potentially exposed population. This factor should also be considered as a potential confounding effect in the estimates of human exposure (Budtz-Jorgensen et al., 2003). The root cause of elevated hair Hg levels should be clarified during the field research process.

Chemical treatment on hair

Chemical treatment on hair was found to influence Hg concentrations in humans’ hair. Foo et al. (1988) reported that lower Hg levels were found in hair that had chemical waving treatment than in untreated hair. Dakeishi et al. (2005) also studied the effect of hair waving on Hg levels in hair and also concluded that on average hair waving treatment using thioglycolate reduced hair Hg concentrations by approximately 30%. These studies confirmed that artificial hair treatment confounded the use of Hg levels in hair as a biomarker. Therefore, Mahaffery and Mergler (1998) named artificial hair treatment as a preanalytical factor. Dakeishi (2005) indicated that the levels of human exposure to Hg may be mistaken if this factor was neglected in Hg exposure studies.

Other factors

Different ethnicities were studied and found to have show differences between the Hg concentrations in the hair of the different racial populations (Foo et al., 1988, Batista et al., 1996, McDowell et al., 2004), but this is also likely to be linked to racial differences in levels of fish and shellfish consumption. Study population’s social profiles, such as GNP (Airey, 1983b) and educational level (Lebel et al., 1997) were also found to contribute to the statistical differences on Hg levels in hair, but again this most probably reflects dietary intake.

2.3 Exposure estimate model for assessing Hg exposure risk

2.3.1 Introduction to exposure estimate models

Exposure can be estimated by evaluating, at various levels of detail, the degree and connectivity between a contaminated source and the concentrations of a hazard and a receptor (e.g. humans) in the environment via various exposure pathways and routes (Nieuwenhuijsen et al., 2006, Environment Agency, 2001, Paustenbach, 2000). The environmental media via which individuals may be exposed to the chemical of concern include air, water, soils, house dust and diet, through the exposure routes of inhalation, dermal contact and ingestion. Exposure estimation is applied to anticipate what might happen or estimate what has happened or did happen in the past (Georgopoulos and Lioy, 1994, Paustenbach et al., 1991). Evaluating exposure, even potential exposure, is essential for receptors, as estimates of the absorbed doses can be used to define the likelihood and extent of exposure risk (Paustenbach, 2000).

Estimating human exposure levels can be conducted using mathematical equations/models based on exposure activities. The equations/models require a diversity of exposure data including characteristics and concentrations of the chemical in media or contaminated sources and the information with the time that receptors are exposed to the chemical. The exposure data applied in the equations/models are traditionally collected using questionnaires, demographic data, survey statistics, behaviour observation, activity diaries, activity models, or, in the absence of more substantive information, assumptions about behaviour (USEPA, 1992b, Nieuwenhuijsen et al., 2006, Paustenbach, 2000). When there are no specific data, users can input the data of hypothetical or potential exposure events for exposure simulating.

Exposure assessments have been widely used in the fields of epidemiology, industrial hygiene and health physics since the early twentieth century, or perhaps earlier (Paustenbach, 2000). With information such as possible exposure behaviour which may result in populations encountering a contaminant, and estimation of the amounts of a contaminant in the exposure receptors, the estimation approach provides initial and virtual help to risk management. Today, exposure assessments are considered as an appropriate approach for the assessment of exposure levels and are commonly accepted in the United States and Europe (USEPA, 1992b, Nieuwenhuijsen et al., 2006, Paustenbach, 2000).

2.3.2 Equations for evaluating exposure dose

Levels of exposure to a chemical can be evaluated using mathematical equations. The general equation used to calculate chronic daily intake doses (Ruby et al., 1999, USEPA, 1989). Chronic / subchronic average daily dose is used to predict or assess the non-carcinogenic effects of a chemical on human health. Averaging time (AT) in this equation is the period of exposure, usually 365 days/year (USEPA, 1997a).

$$\text{Chronic average daily dose} = \frac{C \times IR \times FI \times EF}{BW \times AT}$$

Equation 2

where

C = chemical concentration

IR = ingestion rate

FI = fraction ingested from contaminated source

EF = exposure frequency

BW = body weight

AT = averaging time

In contrast to chronic average daily dose, a lifetime average daily dose expresses the primary health risk of carcinogenic or other chronic chemical effects. The equation to evaluate the lifetime average daily dose is shown as Equation 3.

$$\text{Lifetime average daily dose} = \frac{C \times IR \times FI \times EF \times ED}{BW \times AT \times LT}$$

Equation 3

where

ED = exposure duration

LT = lifetime

and the rest of the parameters are as defined previously.

The meaning of lifetime (LT) in the equation is time, averaging the cumulative dose over a lifetime, usually 70-year. The parameter of exposure duration (ED) is also a time factor in the equation, but ED may be shorter than LT.

Exposure assessments require good quality exposure data, including concentrations and characteristics of the hazard, time and activities of the population in order to obtain a reliable and accurate estimate. Nieuwenhuijsen et al. (2006) suggested that actual measurements of contamination levels are better for exposure estimation than the generic and proxy values, since exposure assessments are very site-specific. For example, concentrations of chemicals may have temporal and spatial variability or they may decline non-linearly with the increase of the distance from contaminated sources. In these cases, extrapolation may not be appropriate to estimate the concentrations of the chemicals. To lead to a better and more valid exposure estimate, the NRC (1991) recommended to quantify area measurements in the vicinity of the sites of activity (Table 3).

Table 3 General hierarchy of exposure measurements with respect to the true exposure fixed source contaminants

Type of data	Approximation to actual exposure
1. Quantified personal measurement	Best type of data for estimating actual exposure
2. Quantified area measurements in the vicinity of the residence or sites of activity	↓
3. Quantified surrogates of exposure (e.g. estimates of drinking water use)	
4. Distance from the site and duration of exposure	
5. Distance or duration of residence	
6. Residence or employment in geographical area in reasonable proximity to the site where exposure can be assumed	
7. Residence or employment in a defined geographical area (e.g. a county) of the site	Worst type of data for estimating actual exposure

Source: The NRC (1991)

2.3.3 Quantification of non-carcinogenic exposure risk

Adverse health risks due to human exposure to non-carcinogenics can be presented as a ratio of the average daily exposure dose (ADD in this study, µg/kg body weight/day) relative to the reference dose (RfD; µg/kg body weight/day). The ratio is named hazard quotient (HQ). A hazard quotient is appropriately applied for a quantitative evaluation of exposure risks through ingestion and dermal contact, whereas the ratio of a chemical's concentration relative to the reference concentration (RfC; µg/m^3) is applied for the exposed route of inhalation. The general formula of hazard quotient is shown in

Equation 4 (USPEA, 1999).

$$HQ = \frac{ADD}{RfD} \quad \text{for exposure via ingestion and dermal contact}$$

or

$$\frac{\text{Concentration}}{RfC} \quad \text{for exposure via inhalation}$$

Equation 4

A hazard quotient is dimensionless. It represents the ratio of the estimated intake level from exposure risk at the exposed site to the RfD. A hazard quotient less than one means that the average daily exposure dose is still under the safe level and adverse health impacts are less likely to occur. In contrast, a hazard quotient greater than one is interpreted that adverse health effects are likely to, or will occur, on the basis of the exposure scenario or the current toxicity data. A hazard quotient is not a probability of harm. For example, a hazard quotient of 0.01 does not mean that there is a one in one hundred chance of the adverse effect occurring.

Hazard index (HI) is a total value of exposure risk by summing the hazard quotients of the individual pathways. Like a hazard quotient, a hazard index is also dimensionless and is not a measure of the probability of harm. Hazard index calculation is a simple tool to estimate exposure risks of populations. It should be examined in light of the uncertainties and assumptions in the entire risk assessment, in order to obtain the comprehensive details of exposure risks in a contaminated site (Goldblum et al., 2006). For example, in a study associated with human exposure to non-carcinogens, Hoffman et al. (1993) defined that a hazard index greater than 1.0 was treated as high-priority for screening and the hazard index value 0.1 can be treated as a low-priority for screening. The screening approach effectively gives advice on the highest priority for appropriate remediation efforts.

2.3.4 Deterministic and probabilistic approaches for exposure estimates

There are two classes of methods used in today's numerical exposure risk characterisation: deterministic and probabilistic methods (Williams and Paustenbach, 2002). Deterministic methods have been traditionally and broadly used in the estimation of environmental exposure risk studies. Single values are used for input variables to produce point estimates of potential exposure. The values are often referred to upper-bound values (i.e. values at the 97.5th percentile), in order to ensure the inclusion of the extreme exposure risk. However, such scenarios generally result in the worst case scenario due to the sums of the multiple commodities and multiple chemicals in the evaluations (Ferrier et al., 2002). The evaluation results are conservative to present only the experience of the population exposed to the high-level. In other words, the exposure risks evaluated by the deterministic method are likely to overestimate the actual exposure. In spite of overestimates, the simple and clear evaluation steps of the single-value method are easily conducted. Thus, the deterministic methods are widely accepted by both regulators and industry to assess exposure risks.

Probabilistic or stochastic techniques use probability distributions of possible occurrences as selected input parameters in mathematical algorithms and equations established to describe the relationships between the parameters. A probabilistic simulation is calculated with data selected repeatedly from the multiple values of the parameters. The simulation result is represented by a distribution of all possible outcomes, which are performed as a cumulative probability distribution function or a probability density function. These may show several types of distributions (e.g.

normal, log-normal, uniform, triangular, etc.) (Schuhmacher et al., 2001, Glorennec, 2006, Fan et al., 2005). Moreover, outcomes of probabilistic simulations are reproducible (Lunchick, 2001).

Uncertainty and variability generating from input data can be considered and identified in probabilistic simulations. ‘Uncertainty’ can be regarded as a lack of knowledge about the true facts of these input parameters. It is reducible by collecting additional information. ‘Variability’ can be described as the heterogeneity and diversity existing in individuals of a population. Because variability is inherent from distinct personal physiological behaviour, it is difficult to eliminate (Schuhmacher et al., 2001). In addition to uncertainty and variability, a comprehensive sensitivity analysis is provided by probabilistic methods. Sensitivity analysis determines how much uncertainties of individual inputs affect the outcome of the simulation and ranks the input contributions with respect to the uncertainty and variability of the outcome (Ma, 2002a, USEPA, 1997b). The information provided by sensitivity analysis is helpful to improve the quality of input data and to reduce the uncertainty and variability in the model.

The characteristics of probabilistic approaches are appropriate to be used in exposure risk assessments because they consider varying exposure data associated with the contaminants and the populations (Petersen, 2000, Ma, 2002b). For example, at a polluted site, a contaminant is likely to present in varying concentrations in a single exposure medium. Variations among personal exposure activities are possibly seen in practical exposure surveys. In addition, body weight and food portions of individuals are varied individually. Even the diets of the same person are varied every day. The features implicate not only the complexity of exposure estimates but also the adaptation of probabilistic assessments in estimates of exposure.

The advantages and disadvantages of both deterministic and probabilistic approaches are listed in Table 4. Compared with generic and deterministic risk characterisation methods, probabilistic and stochastic approaches require a relatively large amount of data and complex statistical techniques. This may be the main reason that regulators and advisory committees are less inclined to carry out and trust the probabilistic outputs if they are unfamiliar with the probabilistic approaches. Nowadays, there are developed computer programmes available for conducting the calculation. The tools can enhance the efficiency of risk assessments as well as risk-based decision-making processes using probabilistic methods. Two commonly used software for probabilistic approaches of the Monte Carlo method and the Latin Hypercube sampling is Crystal Ball® (Version 7.2.1, Decisioneering, Inc., Devor, CO, USA) and @Risk (Version 4.5, Palisade Corp., Newfield, NY, USA).

Table 4 Advantages and disadvantages of deterministic and probabilistic approaches

Approach	Advantages	Disadvantage
Deterministic	▪ Guidelines exist for the risk assessment process. ▪ Less need for extensive databases to support the input variables. ▪ Standard default assumptions can be used. ▪ Relatively easy to carry out. ▪ Single-risk estimate output is easy to understand and interpret. ▪ Industry and regulations are familiar with this approach.	▪ Not all the available (and potentially value) data for each variable are used. ▪ Knowledge and data on patterns of use and exposure potential are not used. ▪ Variability and uncertainty are not reflected. ▪ Exposure estimate can be wrongly assumed as the average or the 75th percentile. Repeated use of the 50th percentile does not produce an estimate of the 50th percentile of the population ('hidden compounding of conservatism').
Probabilistic	▪ All available knowledge and data are used and the probability of a value occurring can be investigated. ▪ Exposure estimate is presented as a distribution, with each value having a probability attached to it. ▪ Probability of potential exposure can be accurately shown, accompanied by extensive information for decision-making. ▪ Variability and uncertainty can be quantified.	▪ Relatively complex to perform with more labour, expertise and resources needed. ▪ Most extreme exposure estimates can be seen from the distribution, allowing consideration, quantification and undue focus on very unlikely events. ▪ Acceptance problems are compounded by the unfamiliar process, the lack of precedents and guidelines, and risk management decisions having to be based on probabilities and large amount of information.

Source: Lunchick (2005)

2.3.5 Earlier studies estimating Hg exposure levels and risk

The deterministic method has been frequently used in studies associated with human exposure to Hg. A study carried out by Goldblum et al. (2006) at the Hg-polluted site of Fort Totten near New York evaluated the doses of chronic daily intake (CDI) via several exposure pathways, based on the site-specific exposure factors and the reasonable maximum exposure strategy (RME, the upper 90^{th}-95^{th} percentiles). The exposure risks of adverse heath impacts on the study population were found mainly via ingestion of finfish, as this exposure pathway was considered to have the highest hazard quotient value of 0.73 among the evaluated exposure pathways. That study also addressed the potential health concern on the child population at their study site.

A similar approach to estimate extreme risk was adopted by Taylor et al. (2005) in Tanzania, where fish consumption was demonstrated to be the most critical route of Hg exposure. In addition to fish consumption, young children were also assessed to be at risk due to the likely ingestion of contaminated soils, although the inadvertent ingestion presently performed a lower risk than other dietary sources (water, rice, and fish).

There have been several studies typically using the deterministic methods and the hazard quotient / hazard index indications to assess the non-carcinogenic health risks for Hg-exposed populations in China (Horvat et al., 2003), Philippines (Appleton et al., 2006), Indonesia (Castilhos et al., 2006) and Spain (De Miguel et al., 2007). The worse-case assumptions were commonly carried out in these risk assessments to achieve single-point results of exposure risks due to Hg exposure.

There are well-developed models available for human exposure assessment, like CalTOX, developed by California Environmental Protection Agency (California EPA, 1997), Risk-Based Corrective Action (RBCA), established by the American Society for Testing and Materials (American Society for Testing and Materials, 2000), Total Risk of Utility Emissions (TRUE), developed by Office of Air Quality Planning and Standard of the USEPA (Seigneur et al., 1996, Constantinou and Seigneur, 1994) and Risk Integrated Software for Clean-ups (RISC4), developed by BP Oil Internation Limited (BP Oil International Limited, 2004). The software efficiently handles a number of distributions of the input data and contains models of multimedia transport and transformation of contaminants, and has scenario models for simulating site-specific pollution.

Some recent studies have assessed Hg exposure levels using the probabilistic technique. In a study of Taiwanese infants exposed to Hg, the probability simulation of Hg exposure risk was conducted using the Monte Carlo technique (Chien et al., 2006). The results found that 12.9% of urban babies and 18.8% of fishermen's babies were evaluated to have the hazard quotient exceeding one, indicating those children having the potential adverse health effects due to Hg exposure. The babies in the fishing village were simulated to have higher levels of Hg exposure than those in the city. That study also demonstrated that the exposure pathway of milk breast ingestion accounted for the greatest proportion of daily Hg intake doses. In the study of Chien et al. (2007), it was indicated by the sensitivity analysis that the parameter of Hg concentration in fish accounted for the most significant contribution to exposure risk.

Using the existing simulation models of a) the transport, transformations and deposition of Hg in the atmosphere, b) hydrology and water quality in terms of Hg bioaccumulation in lakes and c) Hg intake by the local population via fish consumption, Seigneur et al. (1997) conducted a Monte Carlo simulation to assess the Hg exposure levels of potentially exposed populations living in the Great Lakes region of the US. Based on the Hg contamination levels in the fish in the study's Hg-rich lakes, the simulated daily Hg intake doses ranged from 0 to 5.7×10^{-4} mg/kg/day with a mean of 8.9×10^{-6} mg/kg/day. The researchers found that although the probabilistic approach had covered the variability of the data associated with environmental characteristics and personal exposure activities, the major uncertainties still originated from the input parameters in the involved simulation models.

The probabilistic analysis methods used in earlier studies contain the full ranges of input data to conduct assessments which provide comprehensive information with realistic estimates regarding potential health concerns to risk managers. However, they require robust data and sophisticated computer techniques, which may pose a difficulty to previous researchers. As a result, probabilistic techniques are still uncommonly seen in studies associated with human exposure to Hg.

2.3.6 Validity of exposure estimates and related studies

To verify the prediction of harmful exposure doses that a chemical reaches vulnerable populations, a direct measurement at the point of contact seems the best way to obtain accurate exposure values (USEPA, 1992b). The approach of the point-of-contact is therefore carried out directly at the time of exposure. The chemical concentrations at the interface between a person and the environment are measured as a function of time to produce an exposure profile. Although the point-of-contact approach obtains accurate and updated exposure doses of individuals, such measurement is often expensive and the measurement devices and techniques are not available for all chemicals.

Valberg et al. (1996) suggested that the real impact on human health is an ideal index to verify the estimates of exposure levels. However, toxicological or epidemiological studies need a long period to observe if adverse clinical health effects are likely to occur. In some exposure cases, low exposure doses of individuals may not cause any symptoms, or the harmful contaminant-related disease cannot be detected. So far, the levels of a chemical tested in human tissue (biomarkers) are considered to provide a picture of the amount of that chemical actually absorbed into the body (Paustenbach and Galbraith, 2006, Ponce et al., 1998). The verification using biomarker data for risk assessment models is also recommended by Petersen (2000).

Some studies have carried out verification for the estimates of human exposure to Hg by comparing the estimated doses to the observed biomarker data such as Hg concentrations in hair or blood of the exposure populations. For example, Canuel et al. (2006) estimated the Hg concentrations in the hair of the study population in the Innu community in east Canada based on the survey data of fish consumption and current knowledge of Hg metabolism. They developed one-compartment model containing pharmacokinetic parameters such as half-life of Hg in hair, number of teeth with amalgam fillings and body absorption rate of inorganic Hg^{2+} to calculate the corresponding Hg concentrations in an individual's hair. The estimated Hg concentrations in hair were then verified by comparing with the actual Hg concentrations measured in hair. The estimated hair Hg concentrations were up to 14-fold higher. The discrepancies between the two data sets were thought to have resulted from the differences in individual genetic characteristics and/or interactive effects of other dietary components.

Gosselin et al. (2006) took the biological samples of hair and blood from indigenous women of the Inuvik region in Canada and reconstructed the personal history of MeHg intake rate using the toxicokinetic model established by Carrier et al. (2001). In comparison with simulated MeHg levels in blood considering interindividual variability of MeHg blood elimination half-life, MeHg concentrations in hair were more sensitively influenced by the variability as in addition to half-life of MeHg, blood:hair conversion ratio was attributed to the variability. Furthermore, in order to verify the modelled MeHg uptake, the simulation data were compared with daily MeHg intake doses estimated from the food frequency questionnaire. The study found that the simulated Hg intake rates based on hair Hg data (mean: 0.03 µg/kg body weight/day) were lower than those estimated from the questionnaire data of fish and sea mammal consumption (mean: 0.20 μg/kg body weight/day). The result implicated that that the daily Hg intake rates back-calculated from the collected hair strands were potentially affected by variable eating habits of the exposed individuals.

Loranger et al. (2002) assessed the potential risk of MeHg exposure for recreational anglers of hydroelectric reservoirs situated in the James Bay territory in Canada where several fish species were increased MeHg concentrations due to recent reservoir filling. The toxicokinetic model of Carrier et al. (2001) was adopted to estimate 94 participants based on their body weights and their fish consumption behaviour collected via questionnaires. The estimated MeHg concentrations in the participants' hair were found to be higher, compared with the actual MeHg concentrations in their hair, with 11% of the cases overestimated by over an order of magnitude. Loranger et al. (2002) commented that direct measurement of Hg concentrations in hair could provide reliable MeHg exposure levels in human bodies than the exposure estimates based on questionnaire data, because recall errors and self-report biases in the questionnaire-based method were found to result in the overestimation in their study.

2.4 The need for validation of risk based studies of human Hg exposure

Exposure assessments are an important analytical tool for evaluating the likelihood and extent of actual or potential exposure of populations to a chemical hazard (Nieuwenhuijsen et al., 2006). Exposure assessments also characterise the crucial component of health risk for the exposure populations by estimating the intake of the contaminant of concern via different environmental media such as air, water, soil, food and exposure routes of inhalation, dermal contact through skin and ingestion. Such assessments relying on environmental measurements of the chemical and exposure models to evaluate the health risk of the population are available and economical for risk assessors to evaluate exposure levels. This exposure estimation method has been commonly adopted in the studies associated with human exposure to Hg (Appleton et al., 2006, Castilhos et al., 2006, Goldblum et al., 2006, Horvat et al., 2003, Taylor et al., 2005, De Miguel et al., 2007).

When the traditional deterministic approach is applied for exposure estimation, however, the single-point input data do not take into account of differences in exposure activities, exposure pathways and physiological conditions of the exposed individuals. In order to protect humans from the adverse health effects, exposure data are usually the statistical data taken from the upper bound value of datasets (e.g. the 97.5^{th} percentile) which may result in the worst-case estimates. Although the deterministic approach has been considered to be conservative and overestimate, most studies with respect to human exposure to Hg used this approach to conduct Hg exposure levels of study populations.

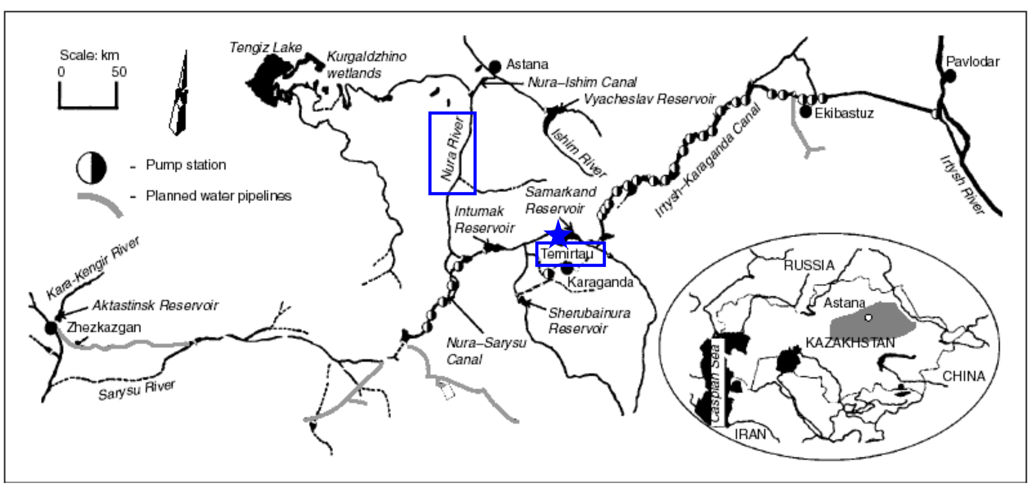
In contrast to deterministic estimates of exposure, exposure assessments using probabilistic technique involves the use of distributions in place of single-point estimates to represent exposure input data. This technique is much more resource intensive approach than in the deterministic approach, but has the essential advantage over the probabilistic estimate for risk assessors as it identifies a probabilistic distribution. The probabilistic approach has been applied on exposure assessments. Although the probabilistic approach is becoming more widely used in chemicant exposure studies and has been used by several researchers in assessing exposure to Hg, the number of studies that have tried to verify the results within the target population has been very low and those have got very mixed results. The aim of the present study is to quantify the validity of the exposure simulation for the estimation of human exposure to Hg pollution in comparison with a reference data of Hg concentrations in hair collected from the exposure population.

3. MATERIALS AND METHODS

3.1 Study site and population

3.1.1 Physical description of the site

The study site was the residential area of Temirtau town and the River Nura and its floodplain villages, Chkalovo, Gagarinskoye, Samarkand and Rostovka, in North Central Kazakhstan (50º02′00″/50º08′42″N and 72º40′12″/72º58′30″E, Figure 4 and Figure 5). It has been estimated that between the period of the mid 1950s and the mid 1970s, a now abandoned acetaldehyde factory, AO Karbide (Figure 6), discharged an annual average of 22-24 tonnes of Hg into the River Nura and polluted the ground below the factory and the wastewater treatment works. Appreciable levels of Hg continued to be released into the environment until the later 1990s, when the factory was closed (Heaven et al., 2000b).



Source: Tanton et al. (2001)

Figure 4 Map of the River Nura flowing areas

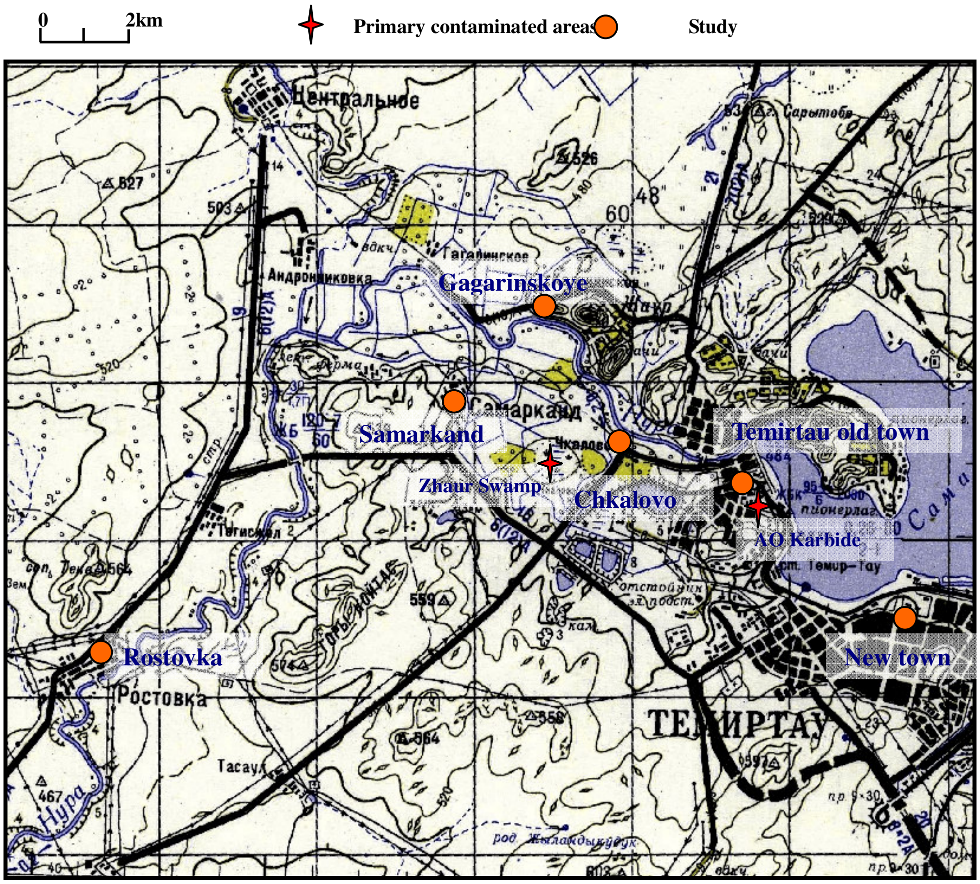


Figure 5 Map of primary contaminated areas and study town and villages

Figure 6 Picture of AO Karbide

The River Nura (Figure 7) is the only perennial water supply in an area of the size of France. It is 978 km long and typically 40-50 m wide and located in the north central massif of Kazakhstan (Heaven et al., 2000b). The head waters of the river are in the Karkaralinsk mountains in the north-east of the region of this dry steppe. The river terminates in the internationally important Kurgaldzhino wetlands (Figure 4). The sources of the runoff for the River Nura are mainly winter snow melt and early spring rains. Precipitation in this semi-arid region is between 250 and 300 mm/year with 80% of the annual flood discharge occurring in less than one month during the spring thaw, while the average annual potential evaporation of this area is 1000 mm/year (Tanton et al., 2001). The River Nura's waters are essential for the needs of the heavily industrialised towns of Karaganda and Temirtau in the centre of the catchment and for agriculture, as well as by the new rapidly developing capital city of Astana, in the lower catchment, some 260 km downstream.

Figure 7 Picture of the River Nura

As a result of the low precipitation and the high evaporation rate, there is no significant groundwater in most of the catchment, although gravel deposits exist in the valley bottom to form some shallow alluvial aquifers with a depth of 0.4 to 10-11 m, fed from the river (Van Epp, 2002). Below the Samarkand dam, there are infiltration galleries in the river gravels where water is abstracted for the domestic water supply for western Temirtau (Figure 11). In the villages further downstream, the groundwater is abstracted directly from the river gravel for household use.

However, Hg concentrations in the local groundwater were found to be insignificant (0.12-0.7 μg/L) (Posch & Partners Consulting Engineers, 2004, Heaven et al., 2000a). It was thought to be related to the various characteristics of the soils at the study site. The floodplain is chestnut soils, permeable and having a good affinity for Hg. The acetaldehyde factory lies on the western edge of town on a low lying hill which is very heavily polluted with Hg. Fortunately, the soils of the hills are made of impermeable clays. This explains why previous studies have not found polluted groundwater from anthropogenic sources moving beyond the boundaries of the factory.

Zhaur Swamp is a low-lying depression on the floodplain to the west of the factory site (Figure 5). This depression was used for disposal of extremely high levels of Hg contaminated waste. Therefore, very high levels of Hg (mean: 306.7 mg/kg) were found in the topsoils (Heaven et al., 2000a). The possibility of infiltration from the relatively shallow depression is not clear but since it is not saline (all non draining depression lakes are saline in the area), there must be some groundwater flows out of the lake. Luckily, because of a layer of peat and clay underlying the polluted depression, Hg is prevented from contaminating river gravel groundwater.

During 50-year operation of the acetaldehyde factory, it discharged Hg pollution into the soils of its surroundings, the wastewater treatment works, the River Nura sediments, floodplain soils, and the old town of Temirtau, all of which are now highly polluted (Tanton et al., 1999, Heaven et al., 2000b, Heaven et al., 2000a). However, because of a need for a perennial water supply there are several residential areas situated in the neighbourhood of the contaminated plant and alongside the river. Except for Temirtau, the people live in the riverine villages, the closest of which are Chkalovo, Gagarinskoye, Samarkand and Rostovka (Figure 8). The residents are generally poor. Some residents maintain their livelihoods by working in the industries in Temirtau, while others tend to consume home-produced food such as vegetables and beef and locally caught fish. The River Nura is either directly or indirectly the main source for water. These villages, together with Temirtau town, were the study populations, whose geographic locations and the lifestyle are described as follows.

Figure 8 Pictures of riverine villages

The town of Temirtau (Темцр-Тау) has been developed as an industrial town with the population of approximately 180,000. Industries include a coal-fired power station, the world largest steel works, the now abandoned chemical production plant of AO Karbide (e.g., calcium carbide, organic synthesis products, synthetic rubber), foundries and forges, cement and asbestos cement plants, and coal mines (Yanin, 1997). Rural and urban lifestyles are observed in the town. The residents of the northwestern Temirtau (the north from 50°03′54″ N approximately), where the polluted acetaldehyde production factory was situated, live in a mixture of flats and traditional small wooden houses surrounded by gardens in which they grow fruit and vegetables (Figure 9). Domestic water supply was from both the Ishim-Karaganda canal and from an infiltration gallery just below the Samarkand dam. A preliminary study determined that the Hg contents in the domestic supply in Temirtau were below the Kazakh maximum allowance Hg level of 0.5 μg/L (Heaven et al., 2000a).

Figure 9 Residential area in Temirtau old town

On the other hand, the eastern area so-called 'new town' has been developed as a modern urban connabation with apartments (Figure 10). Many local people grow fruits and vegetables at dacha (small summer homes), private gardens or farms near the Samarkand Reservoir (Figure 11 and Figure 12). The Samarkand Reservoir has been indirectly used for the purpose of the domestic drinking water and proven to contain low Hg concentrations in a preliminary study (Heaven et al., 2000b).

Figure 10 Picture of Temirtau new town

Figure 11 Map of dachas at the study site

Figure 12 Picture of dacha area

The village Chkalovo (Чкалово) (location shown in Figure 4) is adjacent to the west of Temirtau and close to Zhaur Swamp, the heavily Hg-polluted site in Temirtau old town and 1.5 km from the polluted factory, where wastewater with very high levels of Hg was discharged. The residents work in Temirtau and have a similar lifestyle to the people living in the town, except that they tend to grow vegetables and/or to keep a few animals. Water from the highly Hg-polluted sewage outfall drain is frequently used for irrigation. The second village Gagarinskoye (Гагаринское) is situated approximately 7.6 km downstream from the plant close to the River Nura. The villages of Samarkand (Самарканд) and Rostovka (Ростовка) are sited further downstream. Samarkand is situated on the left river bank and 8.5 km west from the pollution source, while Rostovka is the furthest downstream village with a distance of some 35 km from the outfall. At the time of the study, the typical lifestyle of the three villages is subsistent farming and fishing. The residents generally pump or divert the water from the River Nura or its tributaries for irrigation and acquire their drinking water from wells in the river gravels. The populations of these riverside villages Gagarinskoye, Samarkand, and Rostovka were less than 2000 people. The population of Chkalovo was the largest being about twice the size of the others, but this residential area was observed to be quite and desolate, as they tended to have their social and commercial activities in the near town of Temirtau.

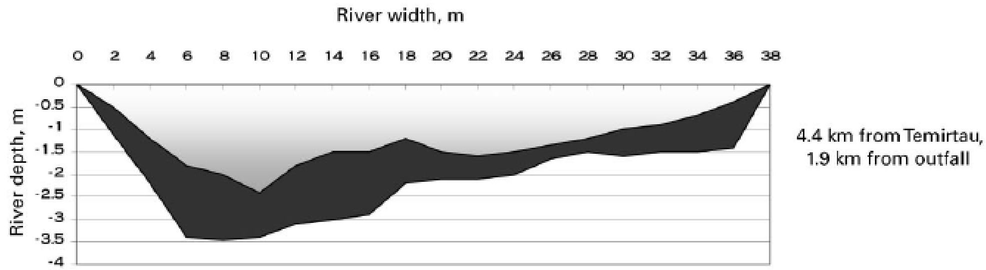
Domestic water supply for most residents was from a central water supply, whereas some families in rural areas still used private wells or boreholes. Nevertheless, the origins of the well water were groundwater flow from the River Nura.

3.1.2 Hg pollution at the study site

The Hg-polluted wastewater was released by the acetaldehyde factory AO Karbide directly into the River Nura from 1950 until the mid 1970s. From the mid 1970s to the mid 1990s, the wastewater was treated by neutralisation, sulfidisation, and magnetic treatment. The treated wastewater was then sent to the municipal sewage works where much of the remaining Hg were bound to the organic solid waste fraction (Yanin, 1997). The wastewater was eventually discharged into the River Nura, 9 km below the Samarkand Reservoir, although it still contained unacceptably high levels of Hg. Despite the closure of the plant in the late 1990s, the Hg in the sediments of the main discharge drain from the sewage works remains high and as a result, it has high concentration of Hg in the drain water. There are also very high levels of Hg pollution at the wastewater treatment work. The outflow of the main drain flows at 1 m^3/s with an annual discharge of approximately 50 kg/year of Hg to the river (Heaven et al., 2000b). As a result of these activities, there were very high levels of Hg in the river sediments below the wastewater outfall. The levels of Hg pollution were the worst in the 25 km downstream from the water outfall at Temirtau, with the average total Hg concentration of 150-240 mg/kg in this section and the Hg levels in excess of 200 mg/kg in the first 9 km from the source (Heaven et al., 2000b). In addition, high levels of the pollution occur again at approximately 75 km downstream of Temirtau when the river enters the Intumak Reservoir and sediment settles (Heaven et al., 2000b).

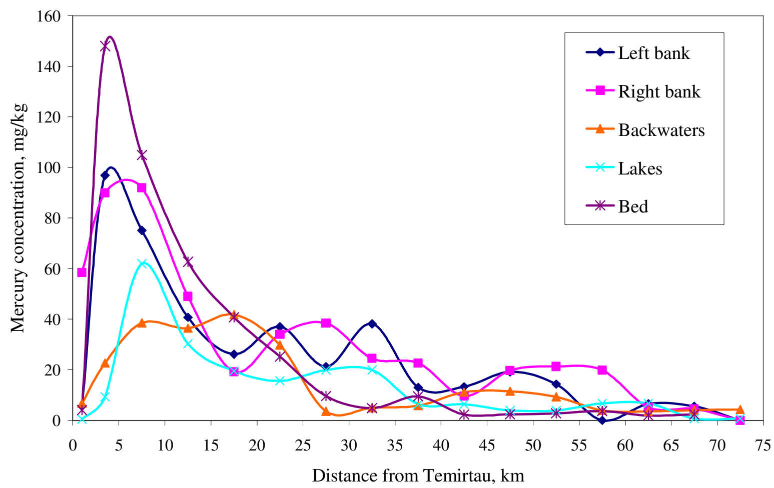
During the operation of the factory, the Hg-contaminated wastewater was $Ca(OH)_2$-rich. This hydroxide transformed to calcium carbonate ($CaCO_3$) into the CO_2-rich water and was deposited in the sediments. At the same time when the waste was discharged into the river, some 5 million tonnes of fly ash was dumped into the River Nura 1 km upstream of the wastewater outfall by a coal-fired power station (KarGRES-1) from 1950 to 1968. The ash is now disposed into lagoons although it is occasionally discharged into the river (Tanton et al., 2001). The fly ash, calcium carbonate and the Hg salts formed a man-made sediment rich in Hg. The Hg was absorbed on the ash at the alkali pH of the river (Yanin, 1997). A previous study has shown that at the pH of the river, the Hg on the sediments is fairly stable (Heaven et al., 2000b). Hence, the technogenic silts have a typical appearance of the local coal-fired ash, a grayblue colour, as shown in Figure 13, accumulated in pockets 1-3.5 m thick on the riverbed and banks and are easily identifiable in contrast to the yellow soils (Figure 14). Heaven et al. (2000b) showed the silts with high concentrations of Hg up to 147 mg/kg distributed in the riverbed, the river banks, and the floodplain in the first 30 km below the outfall (Figure 15).

Figure 13 Pictures of technogenic silts



Source: Heaven et al. (2000b)

Figure 14 Typical profile of technogenic silt distribution in the River Nura



Source: Stratienko (2004)

Figure 15 Hg content in technogenic silts of riverbed, river banks, backwaters, and oxbow-lakes of the Rive Nura

Away from the factory site and the wastewater treatment works, the contaminant was limited to the river and its floodplain (Figure 16). Hg pollution was widespread in the floodplain of the River Nura below Temirtau, with the river bed, the river banks and the oxbows being heavily contaminated. Approximately 50% of the heavily contaminated silts were located in the river in the first 25 km below the outfall, more than 70% of the total Hg was located in the topsoils (0-20 cm) and over 90% of Hg deposited on the river banks was within 2 km of the outfall (Heaven et al., 2000b, Heaven et al., 2000a). These studies estimated that there were more than 9.4 tonnes of Hg in the silts of the section of the river from the polluted factory site to the Intumak

Reservoir, some 75 km downstream.

The Zhaur Swamp was used for emergency discharges of mostly elemental Hg and Hg sulphate from the periodic flushing of the reactors and as a result it is highly contaminated (Heaven et al., 2000a). Very high levels of Hg were measured in the upper 20 cm of the soils in this area, with Hg content reducing with increased depth. According to the study of Heaven et al. (2000a), the mean total Hg concentrations was 306.7 mg/g in the 0-20 cm topsoils, decreasing gradually to 54.6, 18.5 and 9.5 mg/kg at 20-40, 40-60 and 60-80 cm depths, respectively. The residents of Chkalovo intensively use the soils for agriculture.

During spring floods, elevated levels of Hg were found in the waters of the River Nura due to re-suspended contaminated sediments. Data of the 1997 flood showed that Hg levels in the unfiltered water rose from 0.5 to 1.25 µg/L (Stratienko, 2004, Heaven et al., 2000b). Floods containing high levels of contaminated sediments have deposited vast depositions on the banks of the river, especially the low-lying land on the inside of banks and widely across the floodplain downstream of the outfall (Figure 16). Heaven et al. (2000a) have studied the distribution of Hg on the river banks and the floodplain and the results of which are summarised in Table 5. Ninety-four percent of the total Hg deposition was discovered in the first 30 km section of the river down the outfall (Heaven et al., 2000a).

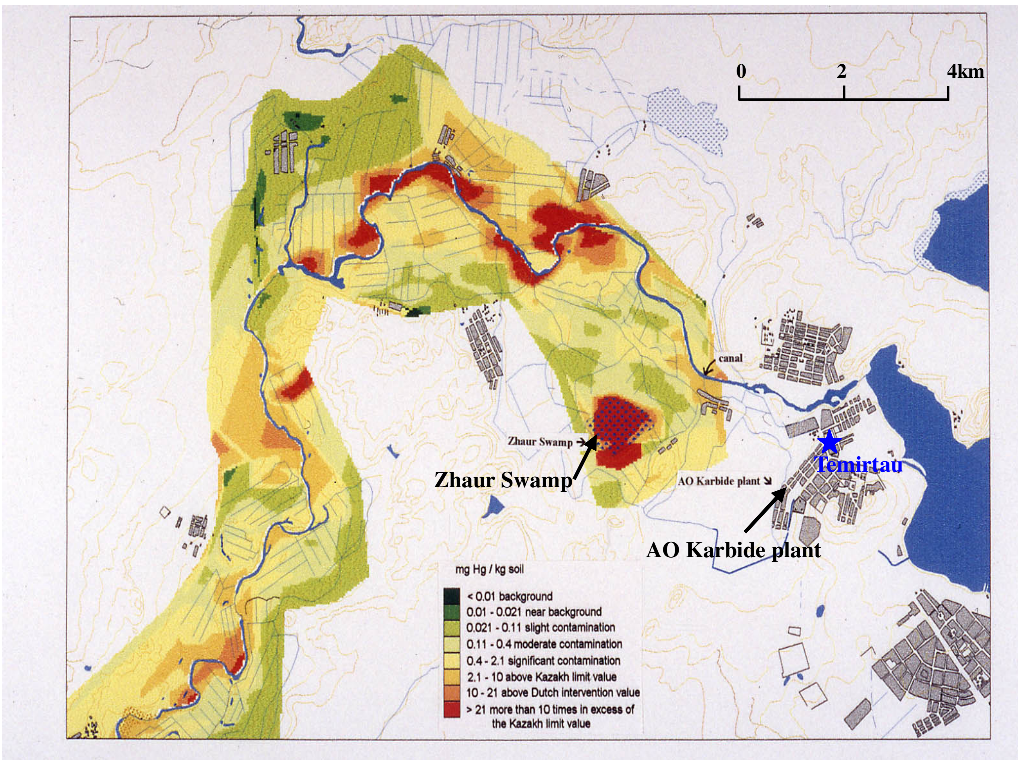
Table 5 Measured total Hg concentrations (mg/kg) in silt deposits along the river and floodplain soils

Distance from Temirtau (km)	**River bank deposits**				**Floodplain topsoils**			
	n	Mean	S.D.	Range	n	Mean	S.D.	Range
1-12.5	24	73.3	7.6	0.47-138.4	195	5.9	1.2	0.001-123.1
12.5-25	26	30.6	2.4	3.62-54.3	272	2.1	0.4	0.004-43.7
25-37.5	26	32.2	2.6	10.7-70.2	204	1.4	0.3	0.006-27.6
37.5-50	22	23.0	2.3	0.06-45.4	252	0.64	0.14	0.001-18.5
50-62.5	16	22.2	1.9	10.6-39.8	106	0.34	0.12	0.001-11.7
62.5-75	3	13.4	4.0	5.5-18.1	67	0.29	0.07	0.002-2.9

Source: Heaven et al. (Heaven et al., 2000a)

The topsoils of the floodplain from Temirtau down to the Intumak Reservoir contained high Hg concentrations, ranging from near background concentration (0.001 mg/kg in Central Kazakhstan) to more than 100 mg/kg. There was an area of

7.54 km^2 (9.9%) of the irrigated or formerly irrigated agricultural areas in the floodplain with Hg concentrations higher than 2.1 mg/kg, the Kazakh maximum legally acceptable level of Hg in soils (Heaven et al., 2000a). The most polluted topsoils were in the first 12.5 km downstream from Temirtau (Figure 16) (Heaven et al., 2000a, Heaven et al., 2000b).



Source: Heaven et al. (Heaven et al., 2000a)

Figure 16 Spatial distribution of total mercury in the topsoils of the floodplain in the first 25 km from Temirtau

Much of these contaminated areas are cropped and in summer the soils are very dry, giving the possibility of Hg-rich dust being transported to the residential areas by the winds and on the crop. Yanin (1997) found the Hg concentrations ranging from 1.02 to 58.13 μg/m^3 in the air at the territory of the plant. Compared with the reference concentration (RfC) 0.3 μg/m^3 recommended by the USEPA, the pollution was very high. These high levels of Hg vapour near the location of the chemical plant were also attributed to the vaporisation of elemental Hg and Hg-rich waste complexes from fabric of the building and soils around the plant site. A more recent survey during June and August in 2004 confirmed that the high Hg levels in the atmosphere were high but only at the joint effluence discharge area (Posch & Partners Consulting Engineers, 2004). Nevertheless, both of these surveys reported that Hg concentrations in the air fell rapidly with distance from the source. Furthermore, Hg contaminations in the air were only high in summer and at the site close to the most contaminated areas, with no significant vaporisation likely to take place in winter due to very low temperature (-18°C on average).

3.1.3 MeHg in environmental media

Most of the data of Hg in the Temirtau region only reported total Hg. In reality, only a small proportion is expected to be MeHg. A previous study reported that there were low rates of methylation in the soils, the sediments, the hydrological systems and the atmosphere in the neighbouring environment of the study site (Posch & Partners Consulting Engineers, 2004). Tanton et al. (1999) found that the methylation rate given by laboratory work was 0.13%. Ullrich et al. (2007c) indicated that methylation effects in the sediments of the River Nura were generally less than 0.1% with the range of 0.01-0.11%. Low fractions of MeHg in sediments of Hg contaminated areas were generally less than 1% (Ullrich et al., 2007c, Ullrich et al., 2001). Although the fractions of MeHg to total Hg were sometimes found to be as high as 13% in the research of Fischer and Gustin (2002), lower methylation rates, less than 1%, have been reported by several studies associated with Hg-pollution in sediments (Heyes et al., 2004, Bloom et al., 1999, Mason et al., 1999, Mason and Lawrence, 1999, Benoit et al., 1998). Therefore, in this study, the fraction of MeHg to total Hg in soils was conservatively presumed to be 1.5% for risk assessment purposes.

The methylation rate in surface water at the study site has not been confirmed yet. Nevertheless, based on Beniot et al. (1998) who found a statistically positive correlation between methylation rate and total Hg in water, the fraction of MeHg in water was referred to the previous studies in other contaminated sites and having the similar Hg-polluted levels in the aquatic environment to the study site. Horvat et al. (2003) reported that in the province of Guizhou, in China, human activities have caused high levels of Hg contamination, up to 1830 ng/L near the wastewater outfall and 450 ng/L at 2.5 km downstream from the main polluted chemical factory. Under such contamination levels, which were similar to that measured in the River Nura, the methylation rates in non-filtered water and filtered water were reported to be 0.003-3.72% and 0.009-3.484%, respectively. In another study associated with severe Hg pollution due to mining industry in the Sacramento River Basin in the USA, the highest level of Hg in non-filtered water was 2248 ng/L, the net amount of MeHg being less than 0.6 ng/L in non-filtered water with a median of approximately 0.1 ng/L (Domagalski, 2001). Methylation rates in surface water with such levels of Hg contamination seem less than 5%. Low methylation rates in general in water systems have been reported by several preliminary studies (Benoit et al., 1998, Coquery et al., 1997, Mason et al., 1999, Horvat et al., 2003). As a result, the percentage of MeHg in surface water was thought to be less than as 5%.

The proportion of MeHg to total Hg in fruit and vegetables together with the methylation rates in the soils at the study site were also unclear. Based on the studies of Cappon (1981) in New York, the US, the percentages of MeHg in total Hg in fruit and vegetables growing in soils of clay loam range from 0 (green bush beans, cucumbers and summer squash) to larger proportions of MeHg in leafy vegetables such as Swiss chard (30.2%). The concentration data of different Hg forms reported by the USEPA (1997d) agreed with the study results of Cappon (1981) and stated that the percentages of MeHg in total Hg in territorial plants were generally low, from 4% in potatoes to 21% in leafy vegetables. In addition, Qiu et al. (2006) surveyed the vegetables in the long-historic mining district of Lanmuchang in China and found high ratios (up to 6%) of MeHg to total Hg in vegetables. Results have shown that in terms of beef and diary product, the fractions of MeHg to total Hg in these foodstuffs are even lower, approximately 19% (USEPA, 1997d). Based on those relevant studies, this study assumed that a rate of 25% was the percentage of MeHg in the local foodstuffs to ensure a safe margin of error.

3.2 Survey of households

The fieldwork was carried out during June/July, 2005. Three fluent Russian speakers were trained as interviewers. The interviewers explained the purpose of the survey to the interviewees prior to their answering the questionnaires and after obtaining consent from the household heads. Questions included the locations of houses, the age and any rebuilding in the previous two years, family members, the present or absence of pets, and ways of reducing contaminant track-in (e.g. shoe removal, door mats) and they were typically completed within ten minutes. The household questionnaire used is given in Appendix A.

Eleven households participated in the survey in Temirtau old town and new town, Chkalovo, and Gagarinskoye, respectively (Table 6). The locations of the households are shown in Appendix D, as these residences were in the most contaminated areas of the floodplain.

Table 6 Sampling size of survey for households from different locations

Area	n
Temirtau old town	11
Temirtau new town	11
Chkalovo	11
Gagarinskoye	11
	44

3.3 Soils and loose dust collection and analysis

Twenty-seven soil samples were collected from the gardens or children's play areas in the residential areas, whilst one was taken from the riverside where the children of Samarkand went swimming frequently in summer and the other one was from the agricultural soils in dachas or gardens (Table 7). Soils were sampled from the top 5 cm of bare soils with a stainless-steel spatula. The coordinates of the soil samples are shown in Appendix D.

Thirty-eight loose dust samples were collected from the residential areas of Temirtau, Chkalovo, and Gagarinskoye, samples being collected from the main streets of the communal areas, the front entrances and the interior entryways of the buildings where the local people frequently visited like local shops and pharmacies (Table 8). Few dust samples were taken from Chkalovo. It was considered that Chkalovo residents generally worked and shopped and/or went to school in Temirtau. In addition to the four residential areas, exterior loose dust samples were collected at the chemical plant and a children's summer camp in Temirtau old town. A dust pan and bush set was used to collect loose dust at those selected locations. The data of the sites such as the coordinate, the name, the substrate and the size of the area were noted at the time of collection (see Appendix B). The coordinates of the dust samples are shown in Appendix D.

Table 7 Number of samples of soils from different locations

Area	Sampled position	n
Temirtau old town	Children's play areas	2
	Garden	1
Temirtau new town	Children's play areas	3
	Garden (dacha)	1
Chkalovo	Children's play areas	1
	Gardens	4
Gagarinskoye	Children's play areas	2
	Gardens	10
Samarkand	Garden	1
	Riverside soils at swimming spot	1
Rostovka	Garden	1
	Total	27

Table 8 Number of samples of loose dust from different locations

Area	Sampled position	n
Temirtau old town	Main streets	3
	Front entrance of shops or pharmacies	3
	Internal entryway of shops or pharmacies	3
Temirtau new town	Main streets	3
	Front entrance of shops or pharmacies	3
	Internal entryway of shops or pharmacies	3
Chkalovo	Main streets	2
	Front entrance of shops or pharmacies	1
	Internal entryway of shops or pharmacies	1
Gagarinskoye	Main streets	4
	Front entrance of shops or pharmacies	4
	Internal entryway of shops or pharmacies	4
Chemical plant	Front entrance	1
	Internal entryway	1
Summer camp in	Front entrance	1
Temirtau old town	Internal entryway	1
	Total	38

Each dust and soil sample was placed in polypropylene bags, sealed, labelled and double-bagged. Dust and soil samples were dried at room temperature and sieved to < 2 mm with a stainless steel sieve. Sample analysis was conducted in the chemical laboratory in Almaty using cold vapour atomic absorption spectroscopy (CVAAS). Heaven et al. (2000b) described the details of analytical methods. Sediment samples were dried in the dark at 20-25°C and were digested in a mixture of HNO_3 and H_2SO_4, according to a method adapted from Hatch and Ott (1968), followed by NaBH4 reduction and AAS detection. Total, dissolved and suspended mercury in water samples was determined directly on site after Au pre-concentration and $SnCl_2$ reduction, using a portable AAS with a detection limit of 1 ng Hg/L. The test procedure followed the standard document of EPA-821-R-01-013.

Quality assurance was carried out by blind determination of standard reference materials, reagent blanks and duplicate samples. Approximately 10% of samples were tested as interlaboratory controls.

3.4 Water collection and analysis

The water samples from the household taps were collected and usage notes (i.e. for drinking, cooking, washing, or irrigating) (Appendix A). Samples of groundwater from taps or wells were collected when water temperature became stable. Water was collected from all the main drinking water supply. Fourteen groundwater samples were collected from the residential areas of Temirtau old town and new town, Chkalovo, Gagarinskoye, Samarkand and Rostovka (Table 9).

Table 9 Number of samples of groundwater from different locations

Area	Sampled position	n
Temirtau old town	Drinking water supply well	1
	Tap water (source: Nizhny Bief canal)	1
Temirtau new town	Drinking water supply well	1
	Tap water (source: Irtysh-Karaganda canal)	1
Chkalovo	Boreholes	3
	Well	1
Gagarinskoye	Drinking water supply wells	3
	Tap water	1
Samarkand	Pumping central water supply	1
Rostovka	Borehole for central water supply	1
	Total	14

Hg levels in surface waters between the Samarkand Reservoir down to Intumak Reservoir have been regularly monitored between 2001 and 2005. The sampling points were selected by British Gas Chair of Environmental Technology in Institute of Power Engineering and Telecommunications (AIPET) and the mapping locations were in Heaven et al. (2000b). In addition to the data, three sites were chosen to measure Hg levels in surface water in this fieldtrip. One was located in the Samarkand Reservoir at the north part of Temirtau where there was a swimming area for school children arranged by a summer camp. The other two were the fishing lakes in the neighbourhood of the village of Gagarinskoye. The coordinates of the collected samples are in Appendix D. Samples were collected in 500 ml polypropylene bottles that were washed and dried. Two replicates were taken from each site and analysed in their laboratory using atomic-fluorescence spectrophotometer (AFS) with cold vapour technique. The analytical procedure and quality assurance are described by Ullrich et al. (2007a). Samples were oxidized with BrCl immediately after delivery to the laboratory and were left standing overnight for complete digestion. Excess bromine was destroyed with hydroxylamine hydrochloride and total Hg was determined by CV-AFS after $SnCl_2$ reduction (PSA (PS Analytical), 2001) on a PSA10.025 Millenium Merlin System (PS Analytical, Kent), using high purity argon (99.99%) as the carrier gas. Quality assurance for water analysis included daily instrument calibration, analysis of blank samples, spike additions and analysis of reference water samples (ORMS-2, National Research Council, Canada). Calibration standards were prepared fresh every day from two working solutions (50 and 100 μg/L) prepared by dilution of a mercury standard solution (1000 mg/L, Merck SpectrosoL). Ultra-pure water (Fistreem Multipure, Fisher Scientific) was used for all dilution purposes. Two field blanks were prepared daily and were treated and analysed in the same way as ordinary water samples, to check for potential contamination. The method detection limit was 2 ng/L.

3.5 Food collection and analysis

Fish

Twenty-one fish samples collected in the fieldwork in 2005 are listed in Table 10. The Hg content of fish was established by sampling locally obtained pike (*Stizostedion lucioperca*), zander (*Stizostedion lucioperca*), bream (*Abramis brama*), roach (*Rutilus rutilus*), carp (*Cyprinus carpio* or *Carassius auratus*), perch (*Perca fluviatilis*), crayfish (*Orconectes rusticus*) and shellfish. The sources of fish (i.e. whether the fish was caught locally or sold in the markets) were provided by the interview participants. The coordinates of the fish samples are in Appendix D.

Table 10 Numbers and collected sites of fish samples

Origins	Fish samples		
	Area	**Items**	**n**
Self-caught	Temirtau old town	Pike	4
		Zander	3
		Perch	1
	Temirtau new town	Carp	2
	Gagarinskoye	Zander	1
		Bream	1
		Roach	3
		Carp	3
		Perch	1
		Gudgeon	1
	Rostovka	Carp	1
	Total		21
Market	Temirtau old town	Pike	1
		Zander	1
		Carp	2
		Crayfish	2
		Catfish	1
	Total		5

The length of the fish was measured before a small part of the fish muscle was taken for analysis. The fish and shellfish samples were stored in plastic bags, labelled, frozen, and transported to the laboratory for chemical analysis. The samples were tested for total Hg contents using atomic-fluorescence spectrophotometer (AFS) Millennium Merlin (PS Analytical, UK) with the cold vapour technique. The details of the analytical procedure and laboratory quality control were reported in the study of Ullrich et al. (2007b). Samples of fish muscle tissue were taken with a stainless steel knife from the posterior part of the fish on the left hand side of the body. Tissue samples were digested in a mixture of HNO_3 and H_2SO_4. This method was found to give better recoveries than either nitric peroxide or aqua regia digestion. Rates of recovery were evaluated using the certified reference material DORM-2 (dogfish muscle tissue; National Research Council Canada). About 1.0 g of tissue was digested with 7 ml conc. HNO_3 and 3 ml conc. H_2SO_4 on a water bath for 1–2 h until the tissue was completely dissolved. The solution was chilled and diluted with ultrapure water to 100 ml. The mixture was quantitatively transferred to a 0.5 l vessel containing 150–200 ml of ultra-pure water. 25 ml conc. HCl, 10 ml 0.2 M KBr and 10 ml 0.2M $KBrO_3$ solution were added and the volume was made up to 0.5 L. Samples were set aside overnight for complete digestion. The solution was then analysed in the same way as described for water samples. The accuracy of the method was assessed by analysis of DORM-2 (certified value 4.64±0.26 mg kg/L), which gave an average recovery of 101.5±1.3% (n=14). All fish samples were analysed in duplicate. All tissue concentrations are reported as wet weight concentrations.

Other food items

The Hg content of the local foodstuffs including tomatoes, cucumbers, potatoes, cabbages, beef and dairy product were measured for a limited numbers of samples (Table 11). The origin of the samples was established, i.e. whether the food was grown in household gardens or purchased from the local markets. The coordinates of the collected samples are in Appendix D. The food samples, as well as fish and shellfish samples, were stored in plastic bags, labelled, frozen and transported to the laboratory for chemical analysis. These food samples were analysed for total Hg contents using atomic-fluorescence spectrophotometer (AFS) Millennium Merlin (PS Analytical, UK) with the cold vapour technique. The samples were digested and analysed by the same method as described for fish samples using the HNO_3/H_2SO_4 digestion method. The details of the analytical procedure and laboratory quality control were reported in the study of Ullrich et al. (2007b). All Hg concentrations are reported as wet weight concentrations.

Table 11 Numbers and collected sites of other food samples

Origins	Food samples		
	Area	**Items**	**n**
Self-produced	Temirtau old town	Potato	1
	Temirtau new town (dacha)		1
	Chkalovo	Potato	3
	Gagarinskoye	Potato	3
	Samarkand	Potato	2
	Rostovka	Potato	1
	Chkalovo	Tomato	2
	Gagarinskoye	Tomato	1
	Temirtau old town	Cucumber	1
	Samarkand	Cucumber	1
	Chkalovo	Cabbage	1
	Chkalovo	Milk	1
Market	Temirtau old town	Potato	1
		Tomato	1
		Cucumber	2
		Beef	2

3.6 Food frequency questionnaire survey

A food frequency questionnaire (FFQ) was designed to establish the dietary behaviour of the residents. The original questionnaire was modified after pre-testing to ensure the questions were understood and relevant. The questionnaire included questions on common food items, food sources, average consumption of frequencies and sizes (by weight). It typically took fifteen minutes to finish the questionnaire. The details of the questionnaire are given in Appendix C.

The questionnaire was also designed to establish participants' biographical details, including name (optional), gender, ethnicity, occupation, number of siblings and residential area. Age and body weight were specified or classified into groups (less than age 16, age 16-30, age 31-45, age 45 above and less than 40 kg, 40.01~60.00 kg, 60.01~80.00 kg, 80.01~100.00 kg, more than 100 kg, respectively). Participants were also asked about hair treatment, i.e. do they use artificial colouring and when, and finally women were asked if they were pregnant or not.

Two hundred and thirty-two questionnaires were completed in this survey. The sample size in each residential area is shown in Table 12. Among these interviewees, some also participated in the household survey and the collection of soils, water, or foodstuffs. The coordinates of the interviewees are given in Appendix D.

Table 12 Sampling sizes of food frequency questionnaire in different residential areas

Area	n
Temirtau old town	45
Temirtau new town	49
Chkalovo	30
Gagarinskoye	47
Samarkand	31
Rostovka	30
Total	232

3.7 Hair collection and analysis

Samples of hair for determination of Hg concentrations were taken from the 1.5 cm close to the scalp of the heads and then placed on plastic bags with the root end stapled. The samples were sent to the National Institute for Minamata Disease in Japan for Hg analysis using Rigaku Mercury Analyzer SP-3 or MA-2 (Nippon Instruments Co., Tokyo, Japan) with a detection limit of 0.1 ng/g and expressed as total Hg concentrations. The DORM-2 (National Research Council of Canada) was adopted as analytical quality control samples for the determination of total Hg (National Institute for Minamata Disease (NIMD), 2006).

The hair samples were taken from the participants of the food frequency questionnaires. If hair samples from other members of the same family were willingly given, they were collected together with their personal data. There were 289 individuals from 232 families providing hair samples in the survey. The distributions of the hair samples from the six residential areas were 50 in Temritau old town, 52 in Temritau new town, 45 in Chkalovo, 67 in Gararinskoye, 42 in Samarkand and 32 in Rostovka. The coordinates are shown in Appendix D.

3.8 Equations for Hg exposure estimation

Equation 5 is adopted for average daily doses of Hg via accidental ingestion of soils (Ruby et al., 1999, USEPA, 1989).

$$ADD_{ingS}\ (\text{mg/kg/day}) = \frac{CS \times IR_S \times ADJ_S \times FC_S \times EF_{ingS} \times BIO_{oral}}{BW \times AT} \times 10^{-6}\ \text{kg/mg}$$

Equation 5

where

ADD_{ingS} = average daily dose via soil ingestion (mg/kg/day)

CS = measured concentration in contaminated soil (mg/kg)

IR_S = ingestion rate of contaminated soil (mg/day)

ADJ_S = adjustment of soil-to-vegetable uptake factor (after wash) (in range 0.0-1.0)

FC_S = fraction of chemical in contaminated soil (mg/mg)

EF_{ingS} = exposure frequency of ingestion of soil (day/year)

BIO_{oral} = oral bioavailability of Hg

BW = body weight (kg)

AT = averaging time (day/year)

Similar to Equation 5, Equation 6 was used to calculate average daily doses of Hg via ingestion of beef, dairy, tomatoes and cucumbers.

$$\mathrm{ADD_{ingF}}\ (\mathrm{mg/kg/day}) = \sum_{i=1}^{4} \frac{\mathrm{CF}_i \times \mathrm{IR}_i \times \mathrm{ADJ}_i \times \mathrm{FC}_i \times \mathrm{EF_{ingF}} \times \mathrm{BIO_{oral}}}{\mathrm{BW} \times \mathrm{AT}} \times 10^{-6}\ \mathrm{kg/mg}$$

Equation 6

where

ADD_{ingF} = average daily dose via ingestion of beef, dairy, tomatoes and cucumbers (mg/kg/day)

CF_i = measured concentration in contaminated foodstuffs (beef, dairy, tomatoes and cucumbers) (mg/kg)

IR_i = ingestion rate of contaminated foodstuffs (beef, dairy, tomatoes and cucumbers) (mg/day)

ADJ_i = adjustment of edible part of foodstuffs (beef, dairy, tomatoes and cucumbers) (in range 0.0-1.0)

FC_i = fraction of chemical in contaminated foodstuffs (beef, dairy, tomatoes and cucumbers) (mg/mg)

EF_{ingF} = exposure frequency of ingestion of foodstuffs (beef, dairy, tomatoes and cucumbers) (day/year)

i = foodstuffs of beef, dairy, tomatoes and cucumbers

and the rest of the parameters are as defined previously.

The average daily dose of Hg via fish and shellfish consumption was calculated from the sum of the average daily dose of Hg ingested through different species of fish and shellfish from different origins. The equation was modified from Equation 6.

$ADD_{ingFish}$ (mg/kg/day) =

$$\sum_{j=1;k=1}^{8;2} \frac{\text{CFISH}_{jk} \times \text{IR}_{\text{Fish}\,jk} \times \text{ADJ}_{\text{Fish}\,jk} \times \text{FC}_{\text{Fish}} \times \text{EF}_{\text{Fish}} \times \text{BIO}_{\text{oral}}}{\text{BW} \times \text{AT}} \times 10^{-6} \text{ kg/mg}$$

Equation 7

$ADD_{ingFish}$ = average daily dose via ingestion of fish and shellfish (mg/kg/day)

$CFISH_{jk}$ = measured concentration in contaminated fish or shellfish (mg/kg)

IR_{Fishjk} = ingestion rate of contaminated fish or shellfish (mg/day)

ADJ_{Fishjk} = adjustment of edible part of fish or shellfish (in range 0.0-1.0)

FC_{Fish} = fraction of chemical in contaminated fish or shellfish (mg/mg)

EF_{Fish} = exposure frequency of ingestion of fish or shellfish (day/year)

j = food item (e.g. pike, zander, beef, etc.)

k = origin of food item (e.g. commercial food in the market, self-caught from the river or self-produced)

and the rest of the parameters are as defined previously.

If a meal was normally prepared for a whole family, the average meal size was evaluated by the division between all family members. Some of the interviews clearly had problems with estimating portion size, for instance, an estimated size of 2500g carp per day for a person, was unlikely to occur in the normal dietary intake. Four out of 232 participants were found to have the portions of fish and shellfish exceeding 500g per day on average, which were overestimated and beyond the 95th percentile of the accumulated frequency distribution. These values were treated as outliers and excluded.

A similar equation (Equation 8) was used for the average daily dose entered through water ingestion.

$$\text{ADD}_{\text{ingWater}} \text{ (mg/kg/day)} = \frac{\text{CW} \times \text{IR}_{\text{ingWater}} \times \text{FC}_{\text{W}} \times \text{EF}_{\text{ingW}} \times \text{BIO}_{\text{oral}}}{\text{BW} \times \text{AT}}$$

Equation 8

where

$ADD_{ingWater}$ = average daily dose via water ingestion (mg/kg/day)

CW = measured concentration in water (mg/L)

$IR_{ingWater}$ = ingestion rate of water in L/day

FC_{W} = fraction of chemical in contaminated water (mg/mg)

EF_{ingW} = exposure frequency of ingestion of water (day/year)

and the rest of the parameters are as defined previously.

Some exposure occurs due to water ingestion during swimming. In this case, the ingestion rate becomes accidental surface water ingestion (IR_{sw}). The time factor of ET is considered as the exposure time of swimming. The equation for accidental water ingestion during swimming is shown in Equation 9.

ADD_{ingSW} (mg/kg/day) =

$$\frac{CSW \times IR_{ingSW} \times ET_{ingSW} \times FC_{SW} \times EF_{ingSW} \times BIO_{oral}}{BW \times AT} \times 10^{-3}\,\text{L/mL}$$

Equation 9

where

ADD_{ingSW} = average daily dose via water ingestion during swimming (mg/kg/day)

CSW = measured concentration in surface water (mg/L)

IR_{ingsw} = ingestion rate of surface water in ml/hr

ET_{ingSW} = exposure time of water ingestion during swimming (hrs/day)

EF_{SW} = exposure frequency of swimming (days/year)

and the rest of the parameters are as defined previously.

The absorption is likely to occur when the chemical passes across the skin and into the blood stream. Equation 10 was therefore used to calculate the absorbed dose as a result of dermal contact of contaminated soils (USEPA, 1989). The adhesion factor (AF) is related to soils types and body parts.

$$ADD_{derS}\ (mg/kg/day) = \frac{CS \times SA_{derS} \times AF \times FC_S \times EF_{derS} \times BIO_{der}}{BW \times AT} \times 10^{-6}\ kg/mg$$

Equation 10

where

ADD_{derS} = average daily dose via dermal contact of soils (mg/kg/day)

SA_{derS} = surface area of skin exposure (cm^2)

AF = adhesion factor (amount of soils adhering to skin) (mg/cm^2/event)

EF_{derS} = exposure frequency of dermal contact of soil (day/year)

BIO_{der} = bioavailability of Hg in soils adhering to skin that is absorbed (mg/mg)

and the rest of the parameters are as defined previously.

Similarly, contaminated water is possibly absorbed by humans via dermal contact during swimming, wading, bathing, or showering. In those cases, Equation 11 was adopted to evaluate the exposure doses. The difference between Equation 11 and Equation 10 is that the absorbed dose through dermal contact of water is calculated without considering the amount of chemicals in contact with the skin (AF). Instead, dermal permeability constant (PC) is taken into account (Valberg et al., 1996).

$$ADD_{derSW}\ (mg/kg/day) = \frac{CSW \times SA_{derSW} \times PC \times ET_{derSW} \times EF_{SW} \times BIO_{der}}{BW \times AT} \times 10^{-3}\ L/cm^3$$

Equation 11

where

ADD_{derSW} = average daily dose via dermal contact of water during swimming (mg/kg/day)

SA_{derSW} = skin surface area available for contact (cm^2)

PC = chemical-specific dermal permeability constant (cm/hr)

ET_{derSW} = exposure time of dermal contact of water during swimming (hrs/day)

and the rest of the parameters are as defined previously.

The inhalation dose of indoor/outdoor air is calculated using Equation 12.

$$ADD_{inhA}\ (\text{mg/kg/day}) = \frac{CA \times IR_{inhA} \times FC_A \times ET_{inh} \times LRF \times EF_{inh} \times BIO_{inh}}{BW \times AT}$$

Equation 12

where

ADD_{inhA} = average daily dose via air inhalation (mg/kg/day)

CA = measured concentration in the air (mg/m^3)

IR_{inhA} = inhalation rate of contaminated air (m^3/hr)

FC_A = fraction of chemical in contaminated air (mg/mg)

ET_{inh} = exposure time of air inhalation (hrs/day)

LRF = lung retention factor (dimensionless)

EF_{inh} = exposure frequency of air inhalation (day/year)

BIO_{inh} = bioavailability of Hg via inhalation (mg/mg)

and the rest of the parameters are as defined previously.

3.9 Probability distributions and Monte Carlo simulations

To assess Hg exposure dose and exposure risks, a probabilistic approach using the Monte Carlo technique was adopted. Crystal Ball software (version 7.2.1, Decisioneering, Denver, CO, USA) was used to process Monte Carlo simulations, which propagated the uncertainty and variability of the parameters throughout the calculation of the risk. The simulations were calculated from 10,000 iterations using randomly selected values derived from each probability distribution of the exposure parameters. The analytical results were then output in the data of distributions with corresponding probabilities at the percentiles of the 0, 10^{th}, 20^{th}, 30^{th}, 45^{th}, 50^{th}, 60^{th}, 70^{th}, 80^{th}, 90^{th}, 95^{th} and 100^{th}. The sensitivity analysis used to rank the simulation's input assumptions with respect to their contribution to model output variability or uncertainty was also performed using Crystal Ball software.

The data for exposure parameters including contamination levels in soil, water, air and foodstuffs, ingestion rates of foodstuffs and body weight were collected in the fieldtrip in 2005. Those site-specific data were conducted to optimise the goodness of fit by the chi-square and Anderson-Darling statistics using Crystal Ball software. Proxy values shown in Table 13 were used for several parameters whose exposure data were not extensively available, such as daily average ingestion rates of soils, skin surface areas during swimming, shower duration time and etc. The distributions and the curves of the exposure parameters for the Monte Carlo simulations are in Appendix E. The single values and the distributions of the parameters were referred to the literature values and experts' views. Parameters including Hg concentrations in multi-media, food intake rates and food consumption frequencies are introduced in Chapter 4 based on the data collected in the fieldtrips during 1997 and 2005.

Table 13 Exposure parameters for the population living alongside the River Nura for Monte Carlo simulations

Parameter	Symbol	Type	Distribution	Reference
Soil exposure				
Ingestion rate for soil (mg/day)	IR_S	Lognormal	65 ± 82	Sander et al. (2006)
Total skin surface area (cm^2)	SA_{derS}	Lognormal	1800 ± 170.0	Sander et al. (2006)
Soil/skin adherence factor (mg/cm^2)	AF	Lognormal	0.81 ± 8.3	Finley et al. (1994)
Soil-to-vegetable uptake factor (after wash) (dimensionless)	ADJ_S	Constant	0.002	Department for Environment Food and Rural Affairs (2002)
Exposure frequency for soil ingestion (days/yr)	EF_{ingS}	Constant	350	USEPA (1997a)
Exposure frequency for dermal contact soil (days /yr)	EF_{derS}	Constant	350	USEPA (1997a)
Exposure frequency for vegetable intake (events/yr)	EF_{ingF}	Constant	350	USEPA (1997a)
Water exposure				
Ingestion rate for drinking water (L/day)	IR_{ingW}	Lognormal	1.366 ± 0.728	Roseberry (1992)
Ingestion rate while swimming (ml/hr)	IR_{ingSW}	Constant	50	Paustenbach (2000)
Total skin surface area (cm^2)	SA_{derSW}	Lognormal	19400 ± 37.4	USEPA (1989)
Skin permeability constant (cm/hr)	PC	Constant	0.0017	USEPA (1992a)
Time spent swimming (hrs/day)	ET_{derSW}	Constant	3	Survey data
Exposure frequency for drinking water (days/yr)	EF_{ingW}	Constant	350	USEPA (1997a)
Exposure frequency for swimming (days/yr)	EF_{SW}	Triangular	0-60	Hertwich et al. (1999)
Outdoor air exposure				
Inhalation rate outdoors (m^3/hr)	IR_{inhA}	Constant	15.2	USEPA (1997a)
Time spent outdoors (hrs/day)	ET_{inh}	Constant	1.75	Hertwich et al. (1999)
Exposure frequency for outdoor air (days/yr)	EF_{inh}	Constant	350	USEPA (1997a)
Food ingestion				
Ingestion absorption adjustment factor for fish (dimensionless)	$ADJ_{FishJ=1-8; K=1-2}$	Constant	0.5	USEPA (1997a)
Ingestion absorption adjustment factor for beef, diary, tomatoes and cucumber (dimensionless)	$ADJ_{i=1-4}$	Constant	1	USEPA (1997a)
Exposure frequency for food ingestion (event/yr)	EF_{ingF}, EF_{Fish}	Constant	350	USEPA (1997a)
General parameters				
Bioavailability of ingestion for MeHg (dimensionless)	BIO_{oral}	Uniform	(0.7, 0.9)	Assumption
Bioavailability of dermal contact for MeHg (dimensionless)	BIO_{der}	Constant	0.2	The RAIS website*
Hg^0 bioavailability of inhalation (dimensionless)	BIO_{inh}	Constant	1	Assumption
MeHg adjustment factor in soil (%)	FC_S	Constant	100 or 1.5	Assumption
MeHg adjustment factor in water (%)	FC_W	Constant	100 or 5	Assumption
MeHg adjustment factor in beef, diary, tomatoes and cucumber (%)	$FC_{i=1-4}$	Constant	100 or 25	Assumption
MeHg adjustment factor in fish and shellfish (%)	FC_{Fish}	Constant	100	Assumption
Body weight (kg)	BW	Site-specific	36-120	Survey data

*: The RAIS website: http://rais.ornl.gov/tox/profiles/methyl_mercury_f_V1.shtml

3.10 Biomarker-based exposure estimation and verification

The validity of Hg exposure levels in human bodies estimated using the equations in Section 3.8 was verified against the observed Hg concentrations in the hair of the study population. To process the verification, the observed Hg concentrations in individuals' hair were converted to their average Hg daily intake doses using the one-compartment model, as shown in Equation 13. The conversion process assumed that the biological parameters with respect to human exposure to Hg were steady-state. The well established input parameters shown in Table 14 were applied in the one-compartment model, according to the recommendations of the health organisations (USEPA, 1997d, WHO, 1990).

$$\text{d (daily Hg intake dose, µg/kg body weight/day)} = \frac{r \times HHg \times b \times V}{A \times f \times bw}$$

Equation 13

Table 14 Summary of variables in the one-compartment model for converting average Hg daily intake dose

Variable	Symbol	Value
Hair/blood ratio (µg/g/µg/mL)	r	250
Hg concentration in hair (µg/g)	HHg	Individual-specific
Elimination constant (days^{-1})	b	0.014
Volume of blood in the body (% of body weight)	V	9
Absorption factor (dimensionless)	A	0.95
Faction of daily intake taken up by blood (dimensionless)	f	0.05
Body weight (kg)	bw	Individual-specific

Q-Q plots were shown to indicate normality of exposure doses of Hg simulated using the Monte Carlo technique and that derived from observed Hg concentrations in hair. A magnitude of the variation of an exposure simulation was called 'variability' in this study. It was regarded as a ratio of the difference between the mean of the 'simulated' exposure data and the mean of the 'observed' exposure doses derived from the Hg concentrations in hair to the mean of the 'observed' exposure doses (Equation 14). Unlike the probabilistic method that produced a range of outputs, the deterministic method only created single-point outputs in the exposure evaluations.

Hence, the single-point results were directly used as the 'mean of simulated value' in the equation of variability. The variability value could be negative, if the mean value of Hg exposure simulation was lower than the mean of the actual exposure data. The variability value equal to zero represented the accurate simulation of Hg intake doses for human exposure to Hg.

$$\text{Variability (\%)} = \frac{\text{(Mean of simulated values) - (Mean of actural value)}}{\text{(Mean of actural value)}} \times 100\%$$

Equation 14

3.11 Statistical Analysis

Descriptive statistics were used to describe the Hg concentrations in the various media and in human hair, as well as lifestyle and dietary behaviour. Q-Q plots, visual inspection for normality, were drawn to show normality of daily intake doses of Hg and Hg concentrations in hair simulated by the Monte Carlo or estimated based on questionnaires. A number of statistical tools were adopted to analyse the data sets. Analysis of variance (ANOVA) was used to compare differences between inter-groups of a number of the variables together with Scheffe post-hoc analysis.

Pearson's correlation coefficient (r) was used to assess a) the linear dependence within dietary intake rates of different food items, b) the linear dependence between dietary intake rates of different food items and Hg levels in hair, c) the validity of the dose-response relationship established using the questionnaire-based simulations of Hg exposure doses and Hg concentrations measured in hair samples.

Lastly, multiple regression analysis was used to build the model for the description of the relationship of the dependent variable Hg concentrations in humans' hair and the independent variables of daily Hg intake doses via food chains sex, age, residential area and fishery occupation. In addition, the multiplicative terms were adopted in the model in order to investigate the interactive effects of independent variables on the dependent variable in the regression equation. The dependent and independent variables in the model were tested normality, homoscendasticity and collinearity, in order to avoid serious bias of misleading of the model. All statistical analyses were performed using the statistical package SYSTAT (version 15, SPSS, Chicago, IL, USA) and the *p.* value less than 0.05 was determined statistical significance.

4. SURVEY RESULTS

4.1 Hg concentrations in key environmental media

4.1.1 Hg concentrations in soils

In general, the soils collected in 2005 from household gardens, children's play areas and loose dust in the communities of the residential areas, contained less than 1 mg/kg of Hg (Table 15). Hg levels in agricultural soils ranged from 0.006 to 0.581 mg/kg, being higher than the background concentration in Central Kazakhstan of 0.001 mg/kg but lower than the Kazakh limit value of 2.1 mg/kg. Compared with a previous much larger sampling-size survey which found 0.05-2.3 mg/kg in the top 20-30 cm of agricultural soils in the regions near the villages Samarkand and Rostovka (Posch & Partners Consulting Engineers, 2004), Hg contamination in the agricultural soils in 2005 survey were lower.

Table 15 Hg concentrations (mg/kg) measured in different types of soils at different residential areas in 2005 survey

	Garden soils		Play area soils		Loose dust	
	n	**Mean (range)**	**n**	**Mean (range)**	**n**	**Mean (range)**
Temirtau old town	1	0.396	2	0.005 (0.004-0.006)	9	0.185 (0.070-0.350)
- Chemical plant	0	-	0	-	2	0.988 (0.994-1.033)
- Summer camp	0	-	0	-	2	0.203 (0.119-0.286)
Temirtau new town	0	-	3	0.006 (0.003-0.007)	9	0.080 (0.030-0.175)
- Dacha	1	0.321	0	-	-	-
Chkalovo	4	0.369 (0.224-0.581)	1	0.017	4	0.104 (0.057-0.176)
Gagarinskoye	10	0.096 (0.029-0.292)	2	0.012 (DL-0.013)	12	0.129 (0.021-0.496)
Samarkand	1	0.067	0	-	0	-
Rostovka	1	0.006	0	-	0	-
Total	18	0.182 (0.006-0.581)	8	0.008 (DL-0.017)	38	0.177 (0.021-1.033)

DL: < detection limit

The garden soils collected from Temirtau old town, Chkalovo and the dacha gardens had higher Hg levels than the other areas, whereas one garden sample collected from the downstream village Rostovka had a very low contamination level of 0.006 mg/kg. Nevertheless, since Hg concentrations are reported to be particularly high on the site of the polluted AO Karbide factory and the river bank within the first 25 km section of the River Nura (Yanin, 1997, Heaven et al., 2000a), severe Hg pollution in the garden soils was expected in the areas adjacent to the heavily contaminated sites as the residents used irrigation water from the Main drain and the river to grow their crops. Heaven et al. (2000a) also indicated that highly polluted irrigated land was in the floodplain of the River Nura and when it floods, it deposits technogenic silts from the river on the land. Since this contained large amounts of Hg, much of the floodplain was contaminated (see Figure 16 in Section 3.1.2). As a result, Hg concentrations in the agricultural soils were found to be high in Temirtau old town, Chkalovo and the dacha but low in the downstream villages.

The lower levels in the garden soils collected in 2005 could be as a result of a small sampling size or the fact that the large-scale survey (Posch & Partners Consulting Engineers, 2004) and the study of Heaven et al. (2000a) were carried out on much more extensive areas.

Hg contamination in the soils of children's play areas was minimal, with the mean of 0.008 mg/kg and with the levels in the play areas of Gagarinskoye below the detection limit. In the survey, the local people reported that the soils in children's play areas in the residential areas were conveyed mostly from outside the polluted site, thus explaining the result.

The lowest level of Hg in the loose dust was in Temirtau new town where the mean of Hg concentrations in the dust was 0.08 mg/kg. It is likely that the contamination of land from Hg particles blowing from the heavily contaminated sites is low and/or high summer soil temperatures lead to volatilisation. However, relatively high concentrations of Hg were found in the loose dust collected from the concrete slabs at the gates of the polluted plant, up to 1.033 mg/kg. This is not surprising given to very high levels of Hg pollution at the factory site. Heaven et al. (2000a, b) observed Hg levels in the topsoils of up to 6467 mg/kg (Table 16). They also observed similar high levels at wastewater treatment plant and Zhaur Swamp through a large scale of survey. Hg levels at those polluted sites are extremely high.

Table 16 Hg concentrations (mg/kg) in the soils at the polluted sites

	0-10cm topsoils		10-20cm topsoils	
	n	**Mean (range)**	**n**	**Mean (range)**
AO Karbide	75	346.98 (2.39-6467.30)	75	223.41 (0.53-4504.00)
Waste treatment plant	10	81.37 (8.83-588.10)	10	56.13 (0.61-384.8)
Zhaur Swamp	28	289.04 (1.15-1974.17) [a]		

a: 0-20cm topsoils
Source: revised from Heaven et al. (2000a, b)

A more recent and more extensive survey of the levels of Hg contamination in Temirtau old town and Chkalovo gave a mean Hg content of 2.521 and 2.931 mg/kg, respectively (Table 17). This survey had one sample from close to the main wastewater discharge drain near the village of Chkalovo (50.6 mg/kg). Except for this Hg-rich sample, the range of Hg concentrations in this area was from 0.007 to 15.350 mg/kg. Of the 94 samples, there were 28 exceeding the Kazakh limit value of 2.1 mg/kg and 5 exceeding the Dutch intervention value of 10 mg/kg. In spite of that, it was seen that the Hg concentrations in the soils some metres away from the most polluted sites reduced dramatically.

Table 17 Previous survey of Hg concentrations (mg/kg) in topsoils in the regions of Temirtau old town and Chkalovo

	0-20cm topsoils	
	n	**Mean (range)**
Temirtau old town	37	2.521 (0.238-14.933)
Chkalovo	57	2.931 (0.077-50.550)

Source: unpublished data from Ilyushchenko (2005)

Compared with the data collected in the survey 2005, the Hg contamination levels in soils in the previous data were higher (Figure 17). High Hg concentrations exceeding the Kazakh limit value of 2.1 mg/kg were tested in some soil samples, whereas Hg concentrations were lower than 2.1 mg/kg in the soil and dust samples collected in 2005.

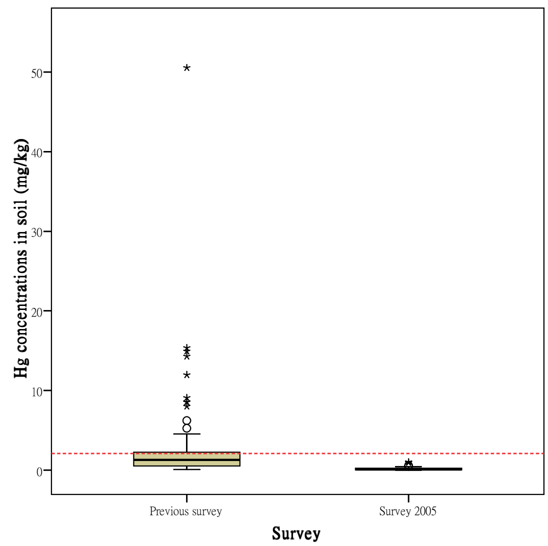


Figure 17 Comparison of Hg concentrations in the soils collected from two periods of surveys

Although the previous soil contamination data (as seen in Table 17) were not conducted specifically in the residential areas, the sampling points were in the irrigated farmland, cattle pasture and steppe areas in close proximity the residences and hence had the potential to impact on the residents' health, particularly via ingestion and dermal contact of soils. As a result, those data, together with the samples of soils and dust collected in the survey in 2005 (as seen in Table 15) were incorporated in the inputs for the human exposure model, as both samples were analysed in the same laboratory. Table 18 shows the distributions of the 158 samples presenting Hg concentrations in the soils or dust of the six residential areas and the 95th percentile values. The distribution curves are illustrated in Appendix F. For those Hg concentrations below the detection limit, the value derived from division of the detection limit by two was used.

Table 18 Distributions of Hg concentrations in soils at different residential areas

Media/Area	n	Distribution type (site-specific)*	Distribution parameters (site-specific)*	95th-tile*
Soils (unit: mg/kg)				
Temirtau old town	53	Gamma	Location = -0.02; scale = 2.54; shape = 0.75	8.781
Temirtau new town	13	Beta	Min = 0; max = 0.43; α = 0.53; β = 1.92	0.321
Chkalovo	68	Lognormal	Mean = 2.40; std. dev. = 7.29	8.396
Gagarinskoye	24	Lognormal	Mean = 0.11; std. dev. = 0.13	0.292
Samarkand	1	Constant	0.067	0.068
Rostovka	1	Constant	0.056	0.058
Total	158	Lognormal	Mean = 2.27; std. dev. = 11.85	8.255

*Data from Table 15 and Table 17

4.1.2 Hg concentrations in surface water

A wide range of the Hg concentrations were found in the samples of surface water from below the detection limit to exceeding 2000 ng/L (Table 19). Based on the regular surveys carried out in the period of 2001 and 2005, relatively high Hg pollution in surface water was seen near the village of Chkalovo. The heavily contaminated points were at the discharge of the wastewater treatment plant (2160 ng/L) and the outfall into the River Nura (1645.01 ng/L). The two samples greatly exceeded the threshold level of 1 μg/L recommended by the WHO (1990) for drinking water. There were fifteen out of thirty-two surface water samples exceeding the Kazakh acceptable level of 0.5 μg/L for drinking water. In short, higher Hg levels were found in the river water of the neighbouring region of Chkalovo than the other areas.

Table 19 Hg concentrations (ng/L) in surface water in different residential areas

2001-2005 survey			2005 survey		
Area	**n**	**Mean (range)**	**Area**	**n**	**Mean (range)**
Temirtau old town	8	81.35 (DL-645.48)	**Summer camp at Samarkand Reservoir**	1	DL
Temirtau new town	7	4.62 (DL-15.96)	-		-
Chkalovo	32	522.49 (16.96-2185.00)	-		-
Gagarinskoye Samarkand	26	326.84 (112.69-1043.56)	**Oxbow lakes**	2	53.16 (DL-105.32)
Rostovka	14	173.00 (41.70-451.25)	-		-
Total	87	325.52 (DL-2185.00)	**Total**	3	35.77 (DL-105.32)

DL: < detection limit

In the fieldtrip in 2005, the residents of Chkalovo reported using the water from the main wastewater drain for irrigation. Since the water has been heavily polluted, the Hg was suspected to be absorbed by the farmed crops.

The following high levels of Hg contamination in surface water were found in Gagarinskoye. The sample collected from the river near Gagarinskoye and 9 km downstream from the Main drain was found to contain Hg as high as 1043 ng/L in the 2002 survey. Except for this sample, the average Hg concentration in this area was 298.68 ng/L. Of twenty-six water samples, there were one above the acceptable drinking water level of 1 μg/L and three above the level of 0.5 μg/L. The most likely reason of this pollution is the high Hg contamination in river sediments, which resulted in Hg concentration up to 104.4 ng/L in the river water between 5 to 10 km from the polluted plant or 3.5 to 8.5 km from the Main drain (Heaven et al., 2000b, Ullrich et al., 2007c).

One sample collected from a lake near Gagarinskoye in the 2005 fieldtrip was 105 ng/L. The lake was a former oxbow lake.

This study used the identical Hg contamination data of surface water for Gagarinskoye and Samarkand. The reason was because Samarkand is situated on the left band of the River Nura and with a distance of approximately 1.5 km away from both the river and Gagarinskoye. Although Gagarinskoye was just located alongside the river and the local people in Gagarinskoye were observed to rely on the river more intensively than those in Samarkand did, a popular swimming site was observed for the school children from Samarkand in this fieldtrip in 2005. As a result, the potential risk posed from the surface water was considered to be common for both Gagarinskoye and Samarkand. Hg exposure from swimming in contaminated water may have been more significant for children as they were often seen swimming in the River Nura in summer. Swimming in contaminated water poses a potential route of exposure to Hg via accidental ingestion or skin dermal contact of water.

Although Temirtau new town was situated geographically adjacent to the contaminated area, the samples of the surface water contained low levels of Hg. Some were even under the detection limit.

Table 20 Distributions of Hg concentrations in surface water at different residential areas

Media/Area	n	Distribution type (site-specific)*	Distribution parameters (site-specific)*	95^{th}-tile
Surface water (unit: ng/L)				
Temirtau old town	9	Lognormal	Mean = 26.52; std. dev. = 148.63	378.861
Temirtau new town	7	Lognormal	Mean = 4.62; std. dev. = 5.32	12.578
Chkalovo	32	Weibull	Location = 10.31; scale = 530.86; shape = 1.10	1289.518
Gagarinskoye	26	Maximum Extreme	Likeliest = 211.40; scale = 150.81	808.43
Samarkand	2	Uniform	Min = 0; max = 512	464.539
Rostovka	14	Lognormal	Mean = 173.83; std. dev. = 141.82	403.424
Total	90	Gamma	Location = -1.80; scale = 481.67; shape = 0.66	956.94

*Data from Table 19

The distributions of the data of Hg contamination in surface water (see Table 19) are listed in Table 20. The data were used in for the exposure risk simulations for the local population using the Monte Carlo technique. The distribution curves are given in Appendix F. The 95th percentile data in Table 20 are adopted in the deterministic evaluation. For those Hg concentrations below the detection limit, the division of the detection limit by two was used.

4.1.3 Hg concentrations in groundwater

Hg levels were below the detection limit in most groundwater samples collected in the fieldtrip in 2005 (Table 21), but relatively high Hg concentrations were measured in samples collected from a drinking water well (5 m deep) in Temirtau old town, the tap water in Gagarinskoye and the central water supply pump in Samarkand, 33.48, 38.73 and 51.09 ng/L, respectively. Nevertheless, those samples were below the international guideline levels and the Kazakh acceptable level of 0.5 μg/L for drinking water.

The data of Hg concentrations in groundwater in this survey were comparable to the results obtained by the preliminary survey conducted in 2002 at the riverine villages including Chkalovo, Gagarinskoye and Andrennikovka by the BCEOM project (unpublished data of the Nura-ishim Basin Environmental and Rehabilitation Project). Among the ten samples of groundwater (tap water or water from wells), Hg concentrations were generally below the detection limit, whereas one sample with a Hg concentration of 362.4 ng/L was collected from the well at the border of Chkalovo where the water was originally groundwater flow from the Main drain. The Hg concentrations in the groundwater samples at the study site were similar to the uncontaminated groundwater level of 10-400 ng/L in Germany.

Table 21 Hg concentrations (ng/L) in groundwater in different residential areas

	2002 survey		2005 survey	
	n	**Mean (range)**	**n**	**Mean (range)**
Temirtau old town	-	-	2	17.05 (DL-33.13)
Temirtau new town	-	-	2	DL
Chkalovo	10	37.14 (DL-362.42)	4	1.35 (DL-2.32)
Gagarinskoye	5	6.01 (DL-17.27)	4	10.43 (DL-38.73)
Samarkand	-	-	1	51.09
Rostovka	-	-	1	DL
Total	15	26.77 (DL-362.42)	14	9.69 (DL-51.09)

DL: < Detection limit

Two periods (year 2002 and 2005) of data were adopted to present the distributions and the 95th percentile values of the Hg levels in groundwater in the different residential areas (Table 22). Because the groundwater system in Temirtau old town was lacking, as discussed before, Hg contamination data in the groundwater in Chkalovo was used for Temirtau old town. The distribution curves are illustrated in Appendix F. For those Hg concentrations below the detection limit, the division of the detection limit by two was used.

Table 22 Distributions of Hg concentrations in groundwater in different residential areas

Media/Area	n	Distribution type (site-specific)*	Distribution parameters (site-specific)*	95th-tile
Groundwater (unit: ng/L)				
Temirtau old town **Chkalovo**	16	Lognormal	Mean = 7.49; std. dev. = 28.61	115.713
Temirtau new town	2	Constant	1.000	1.000
Gagarinskoye	9	Gamma	Location = 0.94; scale = 25.36; shape = 0.28	30.145
Samarkand	1	Constant	51.090	48.586
Rostovka	1	Constant	1.000	1.000
Total	39	Lognormal	Mean = 7.69; std. dev. = 25.31	46.146

*Data from Table 21

4.1.4 Hg concentrations in the air

No data regarding Hg contamination in air were collected in the 2005 fieldtrip. According to the previous survey by Yanin (1997), Hg concentrations ranged from 30 to 575 ng/m^3 in the vicinity of the wastewater treatment plant and from 40 to 199 ng/m^3 in western Temirtau, whilst the highest Hg level were found in the surrounding of AO Karbide with 580 ng/m^3. Posch & Partners Consulting Engineers (2004) updated the finding with Hg levels of 75-290 ng/m^3 in the air near the joint wastewater effluent. Hg contamination levels in the atmosphere were below the WHO safety guideline of 1,000 ng/m^3 (WHO, 2000), the reference concentration (RfC) of 300 ng/m^3 developed by the USEPA (1997e) and the Kazakh acceptable limit of 300 ng/m^3. Low levels of Hg in the air declined rapidly with the distance further from the polluted plant.

Table 23 shows the distributions of Hg concentrations in the air in the different residential areas. Based on the previous survey at the contaminated site, two types of data were determined to describe Hg pollution in the air. For the residents in Temirtau, Hg levels in the air were high due to the polluted plant, the contamination data were applied as a lognormal distribution ranging from 75 to 290 ng/m^3 with a mean value of 45 ng/m^3. For the populations of Chkalovo, Gagarinskoye, Samarkand and Rostovka, the levels of Hg were at a constant 20 ng/m^3. The distribution curves of these distribution types are illustrated in Appendix F. For those Hg levels below the detection limit, the division of the detection limit by two was used.

Table 23 Distributions of Hg concentrations in air in different residential areas

Media/Area	Distribution type (site-specific)	Distribution parameters (site-specific)	95th-tile
Air (ng/m^3)			
Temirtau old town	Lognormal	Mean = 45; std. dev. = 20	290
Temirtau new town			
Chkalovo	Constant	20	20
Gagarinskoye			
Samarkand			
Rostovka			
Total	Lognormal	Mean = 45; std. dev. = 20	290

4.2 Hg concentrations in food

4.2.1 Hg concentrations in fish and shellfish

Hg concentrations in various river fish species found in the 2005 fieldtrip ranged from 0.016 to 0.516 µg/g (Table 24). The samples of bream and gudgeon had relatively high levels of Hg in muscle tissues, compared with Hg levels in predatory fish. Nevertheless, the most likely cause of this was the small sample size of pike and zander that might not have been caught in the River Nura. Most river fish samples in this survey contained Hg concentrations of less than the threshold level of 0.5 µg/g that was suggested by both the Environment Canada (website: http://www.ec.gc.ca/MERCURY/EH/EN/eh-hc.cfm?SELECT=EH) and European Standards (Crepet et al., 2005), except for one carp sample caught near Gagarinskoye. However, the more strict acceptable level of 0.3 µg/g was set by the Kazakh authority. As a result, one third of the river fish samples were above the acceptable level.

In the more extensive survey by Ullrich et al. (2007b), Hg contamination levels in the river fish were found to be appreciably higher than in the 2005 survey (Table 25 and Figure 18). This survey of Hg in the fish in the River Nura between the drain outfall and the Intumak Reservoir found that the means of Hg concentrations ranged from 0.325 to 0.923 µg/g in various non-predatory fish species (Figure 19). Several fish samples had higher Hg concentrations than the critical level. Approximate 84% of river fish samples exceeded the Kazakh safety level of 0.3 µg/g, whilst about 33% contained higher Hg than the threshold levels of 0.5 µg/g. The result indicated the bioaccumulation of Hg in the aquatic food chain. Nevertheless, the Hg loads in the fish caught from the Nura River were lower than that tested in the Minamata City in Japan in 1959, where Hg levels were reported to be 20.0 µg/g in shortneck clams, 24.1 µg/g in sea bream and 10.6 µg/g in grey mullet (Harada, 1995).

Table 24 Hg concentrations (µg/g) in river fish and market fish

Origins	Fish samples		
	Items	**n**	**Mean (range)**
Self-caught	Pike	4	0.127 (0.045-0.290)
	Zander	4	0.160 (0.054-0.404)
	Bream	1	0.390
	Roach	3	0.243 (0.198-0.309)
	Carp	6	0.250 (0.016-0.516)
	Perch	2	0.163 (0.149-0.178)
	Gudgeon	1	0.339
	Mean	21	0.211 (0.016-0.516)
Market	Pike	1	0.054
	Zander	1	0.015
	Carp	2	0.046 (0.031-0.060)
	Crayfish	2	0.034 (0.026-0.043)
	Catfish	1	0.063
	Mean	7	0.042 (0.015-0.063)

Table 25 Mean Hg concentration (µg/g) and sampling size in the fish of the River Nura

	Outfall	Near outfall	Mill house dam	Tegiz Zhol	Intumak Reservior outflow
Bream	0.325 (2)	0.506 (3)	0.557 (7)	-	-
Roach	0.485 (2)	0.379 (19)	0.422 (6)	0.383 (5)	0.482 (5)
Gudgeon	0.359 (1)	0.436 (16)	0.425 (2)	-	-
Dace	-	0.093 (1)	-	-	0.306 (1)
Perch	-	-	0.923 (5)	0.703 (1)	0.544 (14)

Source: revised from Ullrich et al. (2007b)

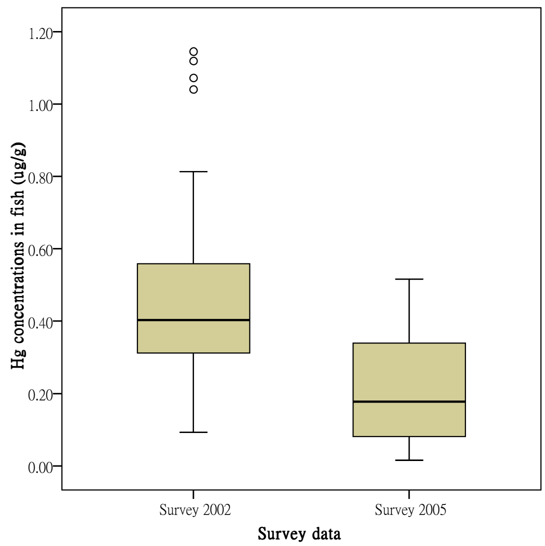
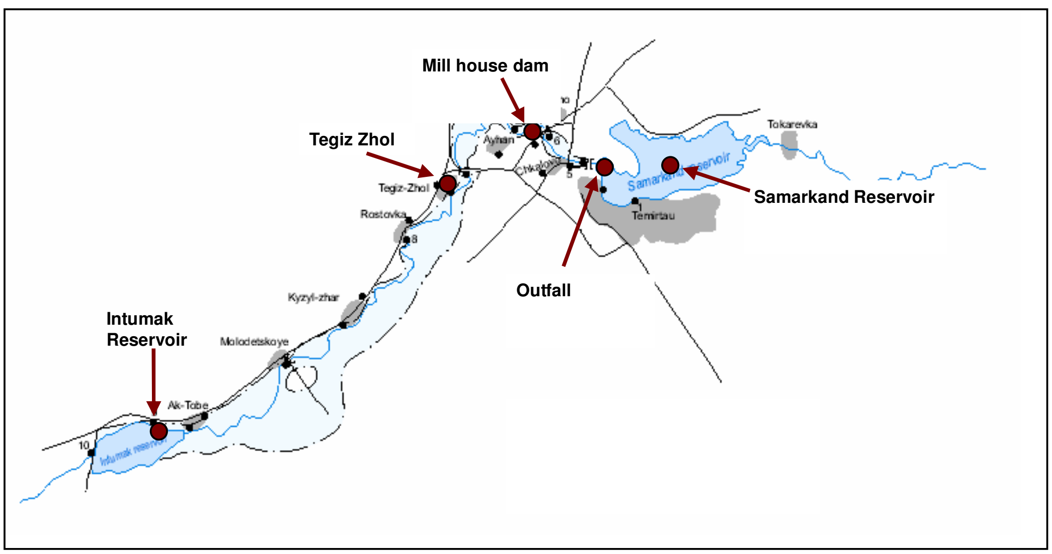


Figure 18 Comparison of Hg levels in fish samples collected from surveys in 2002 and in 2005



Source: revised from Ullrich et al. (2007c)

Figure 19 Map of fish collection in the survey 2002

Perch, a low-level predator, were observed to have higher levels of Hg than other species. Three out of five perch sampled at the mill house dam (14.2 km downstream from the outfall) had Hg concentrations exceeding 1 μg/g. Perch is omnivorous and commonly seen in the River Nura. As a result of Hg accumulation via the aquatic food chains, perch were expected to bioaccumulate a higher amount of Hg at the study site than some of the other species. The fact of higher levels of Hg in river fish were not seen in the data of the 2005 fieldtrip was probably a result of no high-level predators being captured. The mean of Hg concentrations in these perch samples was as low as 0.163 µg/g, one order lower than that in the perch samples collected in the survey in 2002. Variation in Hg levels between the two groups of data is likely to be due to different sampling locations. The samples in this latest fieldtrip were collected from the upstream areas of Temirtau and Gagarinskoye, whereas the sampling areas in 2002 were distributed including the downstream from the polluted sources.

On the other hand, Hg levels in seven samples of market sold fish and shellfish varied from 0.015 to 0.063 µg/g (Table 24), the levels being much lower than river fish and similar to the uncontaminated fish in the UK which has a the mean of 0.054 µg/g (Ysart et al., 1999). In other words, market fish contributed insignificant Hg intake to the local people's diet, compared with river fish. The implication was that, in addition to fish meal size/frequency, the source of the fish was also a crucial factor in assessing the level of human exposure to Hg. Consumers with meals of commercial fish typically had a lower possibility to encounter the adverse health effects due to Hg exposure, compared with the population eating fish caught from the River Nura.

Table 26 shows the distributions of Hg concentration in various fish species from the river and the markets, based on the surveys in 2002 and 2005 for river fish and the survey in 2005 for market fish. Ten fish samples collected at the Intumak Reservoir outfall in Table 25 were not included in the data regarding river fish contamination in this study, as the area was less likely to be a fishing place for the study population. There were no river fish samples of bream and roach collected from Temirtau due to an urban residence, the data of Hg concentrations in river fish in this area was identical with Chkalovo. Likewise, few samples of river fish were collected from Rostovka; the fish data were assumed to be the same in Samarkand and Rostovka. The presumption was based on the geographic locations of the residences. No regional difference was seen with respect of Hg contamination levels in pike, zander, carp, crayfish, shellfish and unclassified fish species, probably because the fish samples were small. Moreover, as Hg concentrations in market fish were low and nearly constant, a uniform distribution from 0.013 to 0.068 µg/g was assumed. The detail distributions for the separated residential areas and the distribution curves are listed in Appendix F.

Table 26 Distributions of Hg concentrations in fish in different residential areas

Media/Area	n	Distribution type (site-specific)*	Distribution parameters (site-specific)*	95^{th}-tile
River fish: pike, zander, crayfish, shellfish and other fish species (unit: mg/kg)				
Total	29	Minimum Extreme	Likeliest = 0.42; scale = 0.14	0.555
River fish: bream and roach (unit: mg/kg)				
Temirtau old town, Temirtau new town, Chkalovo	4	Beta	Min = 0.24; max = 0.61; α = 0.94; β = 1.15	0.803
Gagarinskoye	26	Gamma	Location = 0.19; scale = 0.15; shape = 1.21	0.715
Samarkand, Rostovka	18	Weibull	Location = 0.12; scale = 0.38; shape = 1.49	0.723
Total	48	Gamma	Location = 0.13; scale = 0.13; shape = 2.20	0.803
River fish: carp (unit: mg/kg)				
Total	6	Gamma	Location = 0.02; scale = 0.46; shape = 0.51	0.501
River fish: perch (unit: mg/kg)				
Temirtau old town, Temirtau new town, Chkalovo, Gagarinskoye	2	Constant	0.163	0.176
Samarkand, Rostovka	6	Maximum Extreme	Likeliest = 0.77; scale = 0.21	1.139
Total	8	Beta	Min = 0.03; max = 1.19; α = 0.81; β = 0.58	1.077
Market fish (unit: mg/kg)				
Total	**7**	Uniform	Min = 0.013; max = 0.068	0.065

*Data from Table 24 and Table 25

4.2.2 Hg concentrations in other foodstuffs

Although only limited sampling of home-grown vegetables were collected, mainly because of the time limit of the fieldwork, Hg contamination levels were lower than 0.1 mg/kg, except for one sample of cucumber having an appreciably higher level of Hg (Table 27). The cabbage sample had a low concentration of Hg, 0.008 mg/kg. Hg levels in the vegetable samples as a whole at the study site ranged from the detect limit to 0.013 μg/g. These are similar to those found in the vegetables from a metropolitan and non-anthropogenic Hg contaminated city in China (Li et al., 2006). Nevertheless, in comparison with the Hg levels in the unpolluted vegetables ranged from 0.0005 to 0.005 μg/g from the Hg-free city of Catalonia in Spain (Falco et al., 2005), Hg contamination in the vegetables of the study areas were higher. In comparison with the acceptable level of 0.01 mg/kg recommended by the Kazakh authority and the Chinese National Standard Agency (Qiu et al., 2006), four out of ten potato samples contained Hg concentrations that just exceeded the limit value. Hg concentrations in the samples of tomatoes and cucumbers were all above the Kazakh acceptable level, regardless of wherever they were collected from the local households or the markets.

Table 27 Hg concentrations (mg/kg) in vegetables at different residential areas

	Potatoes		Tomatoes		Cucumbers		Cabbage	
	n	**Mean (range)**	**n**	**Mean (range)**	**n**	**Mean (range)**	**n**	**Mean (range)**
Temirtau old town	1	0.010	-	-	1	0.064	-	-
Temirtau new town	1	0.011	-	-	-	-	-	-
Chkalovo	3	0.009 (0.008-0.010)	1	0.017	1	0.120	1	0.008
Gagarinskoye	3	0.008 (0.007-0.010)	1	0.016	-	-	-	-
Samarkand	2	0.012 (0.010-0.014)	-	-	-	-	-	-
Rostovka	1	0.013	-	-	-	-	-	-
Mean	11	0.010 (0.007-0.014)	2	0.017 (0.016-0.017)	2	0.092 (0.064-0.120)	1	0.008
Market	1	0.005	2	0.017 (0.016-0.019)	2	0.058 (0.055-0.064)	-	-

Home-grown potatoes and cucumbers had slightly higher Hg concentrations than market product, but the sample numbers were too few to test the statistical difference. Hg concentrations in four samples of market tomatoes and cucumbers exceeded the Kazakh acceptable level. During sample collection procedure, the origins of the market vegetables were not confirmed from the vendors. The vegetables sold in the market were possibly grown in the local farms. The Hg contaminated levels were similar to those home-grown samples, which confirms this assumption.

Hg levels in the beef from the two market venders were found to be lower than the detection limit 0.002 mg/kg.

Table 28 shows the assumed values of Hg concentrations in various food types for the probabilistic simulations. Hg concentrations in the vegetables and beef from the local markets and households were treated as being the same as the differences between the two sources were very minimal. The 95th percentile values were the input data in the single-value simulations. For those Hg levels below the detection limit, the division of the detection limit by two was used. In addition, due to the limited food samples, Hg contamination in food was assumed to be the same among the six residential areas.

Table 28 Distributions of Hg concentrations in food in different residential areas

Media/Area	Distribution type	Distribution parameters	95th-tile
Beef and dairy: ***home-fed and commercial (unit: mg/kg)***	Constant	0.001*	0.001
Tomatoes: ***home-grown and commercial (unit: mg/kg)***	Constant	0.017	0.019
Cucumbers: ***home-grown and commercial (unit: mg/kg)***	Constant	0.075	0.113

*: <Detection limit

4.3 Survey results of dietary intake patterns

4.3.1 Dietary intake patterns on fish and shellfish

Table 29 gives the frequency distribution of reported consumption of different fish species by the residents of the study region. Carp was the most commonly consumed fish, with approximately 80% of the population having eaten carp in the previous year. Crayfish, shellfish, perch and others non-categorised species were not a significant part of the local diet of many people, with more than 50% of the population having not consumed those species in the past year. Only three people reported having eaten shellfish in the past year.

Table 30 reports the sources of the different fish species. More than 50% of the bream, roach, carp and perch were reported to be caught from the River Nura or the neighbouring lakes. These fish species were a subsistent food caught from the local river or the lakes. By contrast, the fish species such as pike, zander, crayfish, shellfish and herring tended to be purchased from the markets or be consumed from tins. This is due to these fish species not living in the River Nura. The local people purchased the commercial fish in the markets or the local shops.

Table 29 Frequency of fish consumption reported by interviews at the study site

Frequency	Pike/zander	Bream/roach	Carp	Crayfish
	n (%)	n (%)	n (%)	n (%)
Did not eat fish in the past year	91 (39.22)	85 (36.64)	48 (20.78)	188 (81.03)
1~6 times per year	82 (35.34)	50 (21.55)	69 (29.87)	33 (14.22)
1 time per month	29 (12.50)	31 (13.36)	43 (18.61)	6 (2.59)
2~3 times per month	19 (8.19)	23 (9.91)	30 (12.99)	2 (0.86)
1 time per week	4 (1.72)	19 (8.19)	18 (7.79)	2 (0.86)
2 times per week	3 (1.29)	12 (5.17)	12 (5.19)	0 (0.00)
3~4 times per week	3 (1.29)	9 (3.88)	9 (3.90)	0 (0.00)
5~6 times per week	0 (0.00)	0 (0.00)	0 (0.00)	0 (0.00)
1 time per day	1 (0.43)	3 (1.29)	2 (0.87)	1 (0.43)
Total	232 (100.00)	232 (100.00)	231 (100.00)	232 (100.00)
Frequency	**Shellfish**	**Herring**	**Perch**	**Other fish**
	n (%)	n (%)	n (%)	n (%)
Did not eat fish in the past year	229 (98.71)	115 (49.57)	208 (89.66)	200 (86.21)
1~6 times per year	1 (0.43)	26 (11.21)	4 (1.72)	17 (7.33)
1 time per month	2 (0.86)	41 (17.67)	7 (3.02)	7 (3.02)
2~3 times per month	0 (0.00)	29 (12.50)	5 (2.16)	1 (0.43)
1 time per week	0 (0.00)	19 (8.19)	2 (0.86)	3 (1.29)
2 times per week	0 (0.00)	1 (0.43)	2 (0.86)	2 (0.86)
3~4 times per week	0 (0.00)	0 (0.00)	3 (1.29)	2 (0.86)
5~6 times per week	0 (0.00)	0 (0.00)	0 (0.00)	0 (0.00)
1 time per day	0 (0.00)	1 (0.43)	1 (0.43)	0 (0.00)
Total	232 (100.00)	232 (100.00)	232 (100.00)	232 (100.00)

The average frequency of fish meals and/or shellfish consumption was 0.23 meals/day. The residents in Samarkand consumed the most fish with a mean of 0.29 meals/day (Table 31). The town of Temirtau and Rostovka had comparable fish consumption frequencies, eating fish and/or shellfish approximately every four days on average. In contrast, the residents in Chkalovo consumed fish much less frequently, once per nine days on average.

Fish consumption of the local people was generally less than two meals per week. Within six residential areas, there were differences in the sources of fish consumed (river caught fish and commercial fish). In Temirtau and Chkalovo, more than 50% of the fish meals were bought from the local markets. Contrarily, self-caught fish were the primary source for the people in the villages, particular for those living in Samarkand and Rostovka where more than 80% of the fish meals were caught from the river or the lakes by themselves. The residents in Gagarinskoye, Samarkand and Rostovka consumed significantly more locally caught fish than commercial purchased fish. The population as a whole consumed river fish more frequently than market products.

Table 30 Reported sources of the fish consumed

Fish species	% of people eating	Source of fish eaten	%
Pike or zander	60.8	Over 50% river fish	35.3
		Over 50% market fish	53.0
		50% river fish, 50% market fish	10.9
		Canned fish	0.8
		Total (%)	100.0
Bream or roach	63.4	Over 50% river fish	53.1
		Over 50% market fish	38.5
		50% river fish, 50% market fish	6.9
		Canned fish	1.5
		Total (%)	100.0
Carp	79.2	Over 50% river fish	51.2
		Over 50% market fish	41.5
		50% river fish, 50% market fish	7.3
		Canned fish	0.0
		Total (%)	100.0
Crayfish	19.0	Over 50% river fish	45.9
		Over 50% market fish	51.4
		50% river fish, 50% market fish	2.7
		Canned fish	0.0
		Total (%)	100.0
Shellfish	1.3	Over 50% river fish	0.0
		Over 50% market fish	100.0
		50% river fish, 50% market fish	0.0
		Canned fish	0.0
		Total (%)	100.0
Herring	50.4	Over 50% river fish	0.0
		Over 50% market fish	83.9
		50% river fish, 50% market fish	0.0
		Canned fish	16.1
		Total (%)	100.0
Perch	10.4	Over 50% river fish	60.0
		Over 50% market fish	40.0
		50% river fish, 50% market fish	0.0
		Canned fish	0.0
		Total (%)	100.0
Other fish species	13.8	Over 50% river fish	12.0
		Over 50% market fish	64.0
		50% river fish, 50% market fish	0.0
		Canned fish	24.0
		Total (%)	100.0

Table 31 Fish and shellfish intake frequencies from different sources at the study site

Food intake frequency (meal/day)	**Mean±SD**		
	Total	**River fish**	**Market fish**
Temirtau old town	0.27±0.34	0.10±0.22	0.15±0.28
Temirtau new town	0.27±0.33	0.12±0.30	0.14±0.19
Chkalovo	0.11±0.12	0.04±0.10	0.07±0.08
Gagarinskoye	0.15±0.21	0.11±0.20*	0.03±0.05
Samarkand	0.29±0.41	0.22±0.34*	0.05±0.10
Rostovka	0.27±0.33	0.21±0.34*	0.05±0.07
Mean (n= 226)	0.23±0.31	0.13±0.26	0.09±0.17

During the fieldtrip in 2005, many residents in Temirtau and Chkalovo reported that they were concerned about Hg contamination in the River Nura and did not eat river fish caught by themselves. It was observed that in local markets, some sellers placed the fish origin on their stalls and mentioned their fish caught from the Samarkand Reservoir or Karaganda, clean water systems. Discussions with the vendors indicated that they recognized the sensitive issue of Hg in fish from the River Nura. The consciousness of Hg pollution in local water system was likely to result in reduced river fish consumption. Burger and Gochfeld (2006) reported that the population of New Jersey had a willingness to change their dietary behaviour when they realised the possible health risks from fish consumption. Likewise, people living close to the polluted sources were likely to raise more environmental concerns than those living outside of the heavily polluted area and to select imported and uncontaminated food in their diets.

Economic status of individuals potentially affected the choices of dietary behaviour. The people living in the riverine villages were generally poor and survived as subsistence farming growing vegetables and feeding livestock. Fish, like other food items, were mainly produced/caught by themselves. Conversely, the residents in Temirtau and Chkalovo had paid jobs with stable salaries and they tended to purchase food from local markets.

The average fish portion per meal ranged from 1.21 to 94.14 g/meal (Table 32). Low meal weights of the fish species including crayfish, shellfish and perch were shown, because a few interviewees had consumed in the past year and the average meal weights of the population were calculated by dividing the reported fish meal sizes by the number of the interviewees. In other word, their distribution functions were skewed with a typical distribution shown in Figure 20. Hence, selecting the correct distribution function was essential to represent dietary intake patterns.

The reported average daily fish consumption portions varied widely. Some residents did not eat fish, whereas some reported eating large portion sizes of fish, up to 500g per day. The large variation is likely to be as a result of the diverse dietary behaviour of individuals or their ethnicities, since the interviewees are ethnic Kazakh, Russian, Ukrainian whose dietary intake patterns regarding fish and shellfish consumption were related to their background.

Table 32 Distribution of mean portion of fish consumption for various fish species (g/meal)

(n=228)	Fish portion (g/meal)			
	Mean	**SD**	**Min**	**Max**
Pike or zander	60.46	104.99	0.00	500.00
Bream or roach	69.58	118.60	0.00	500.00
Carp	94.14	134.64	0.00	500.00
Crayfish	14.86	62.53	0.00	500.00
Shellfish	1.21	15.68	0.00	250.00
Herring	77.60	94.47	0.00	500.00
Perch	9.14	44.19	0.00	333.33
Other fish species	10.74	54.69	0.00	500.00

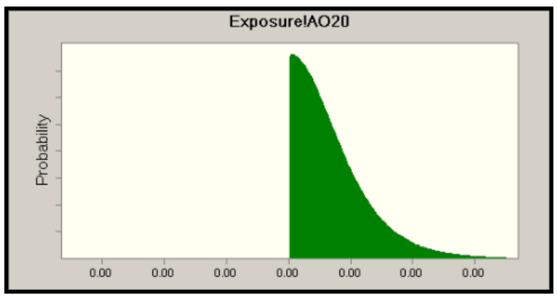


Figure 20 Typical distribution of parameters of meal weights including crayfish, shellfish and perch

Table 33 shows the distributions and the 95th percentile values of fish intake frequencies (meal/day) and intake rate (g/meal) for the whole population. The data for separate residential areas and the distribution curves are listed in Appendix G. The distribution data were site-specific and assumed based on the questionnaire survey.

Table 33 Distributions of frequency of fish consumption and fish consumption expressed as an average intake for the population as a whole

Media/Area	Distribution type (site-specific)	Distribution parameters (site-specific)	95th-tile
Fish intake frequency: river fish (unit: meals/day)			
Pike or zander	Gamma	Location= -0.001; scale= 0.040; shape= 0.332	0.07
Bream or roach	Logistic	Mean= 0.003; scale= 0.115	0.28
Carp	Gamma	Location= -0.001; scale= 0.155; shape= 0.331	0.28
Crayfish	Logistic	Mean= 0; scale= 0.010	0.01
Shellfish	Constant	0	0
Herring	Constant	0	0
Perch	Logistic	Mean= 0.003; scale= 0.023	0.02
Other species	Logistic	Mean= 0; scale= 0.010	0
Fish intake frequency: market fish (unit: meals/day)			
Pike or zander	Gamma	Location= -0.001; scale= 0.041; shape= 0.359	0.08
Bream or roach	Gamma	Location= -0.001; scale= 0.075; shape= 0.287	0.14
Carp	Weibull	Location= -0.028; scale= 0.180; shape= 0.282	0.08
Crayfish	Normal	Mean= 0.001; std. dev.= 0.006	0.01
Shellfish	Logistic	Mean= 0; scale= 0.001	0
Herring	Gamma	Location= -0.001; scale= 0.080; shape= 0.383	0.14
Perch	Minimum Extreme	Likeliest= 0.006; scale= 0.019	0
Other species	Logistic	Mean= 0; scale= 0.003	0.01
Fish meal portion: river fish (unit: g/meal)			
Pike or zander	Logistic	Mean= 28.590; scale= 62.339	298.33
Bream or roach	Gamma	Location= -0.833; scale= 126.234; shape= 0.451	366.25
Carp	Normal	Mean= 118.849; std. dev.= 164.322	400
Crayfish	Logistic	Mean= 2.046; scale= 17.486	86.00
Shellfish	Constant	0	0
Herring	Constant	0	0
Perch	Minimum Extreme	Likeliest= 4.770; scale= 99.086	135.00
Other species	Logistic	Mean= 0.525; scale= 9.600	0
Fish meal portion: market fish (unit: g/meal)			
Pike or zander	Beta	Min.= -10.305; Max.= 3240; alpha= 0.382; beta= 12.750	300.00
Bream or roach	Logistic	Mean= 26.715; scale= 50.078	285.00
Carp	Logistic	Mean= 7.137; scale= 26.624	333.33
Crayfish	Logistic	Mean= 4.179; scale= 33.728	142.50
Shellfish	Minimum Extreme	Likeliest= 19.622; scale= 61.632	0
Herring	Beta	Min.= -28.132; Max.= 889.680; alpha= 0.575; beta= 4.202	300.00
Perch	Constant	1	0
Other species	Minimum Extreme	Likeliest= 16.084; scale= 141.345	100.00

4.3.2 Dietary intake patterns of other foodstuffs

Consumption frequencies of beef and dairy sourced locally from households or markets are reported in Table 34. Similar intake rates were observed within the six residential areas. However, more than two third of beef meals in Temirtau were purchased from the markets, whereas the people in the villages particularly Rostovka mainly ate their own cattle. The residents in Rostovka only had one ninth of their beef meals purchased from the local market.

Table 34 Mean beef and dairy product consumption frequencies of from different sources

Food intake frequency (meal/day)	Beef / Mean±SD			Dairy / Mean±SD		
	Total	Home	Market	Total	Home	Market
Temirtau old town	0.35±0.38	0.09±0.24	0.22±0.32	0.75±0.37	0.31±0.45	0.30±0.41
Temirtau new town	0.35±0.27	0.02±0.14	0.26±0.35	0.66±0.45	0.11±0.28	0.47±0.50
Chkalovo	0.36±0.33	0.19±0.27	0.09±0.23	1.03±0.59	0.95±0.66	0.05±0.19
Gagarinskoye	0.43±0.50	0.20±0.41	0.14±0.31	1.23±0.62	0.98±0.77	0.17±0.47
Samarkand	0.35±0.36	0.16±0.26	0.15±0.31	0.78±0.57	0.48±0.65	0.25±0.38
Rostovka	0.28±0.27	0.23±0.28	0.03±0.06	0.83±0.62	0.72±0.67	0.06±0.19
Mean (n= 218)	0.36±0.38	0.14±0.29	0.16±0.30	0.88±0.57	0.56±0.67	0.24±0.42

The residents of Chkalovo and Gagarinskoye consumed dairy products more frequently than in the other areas, milk being mainly from their own cows. The people of Temirtau, Samarkand and Rostovka ate meat and dairy product with almost similar frequency but from difference sources. Town people particularly living in the new district of Temirtau mainly consumed commercial beef/dairy products, whereas in Samarkand and Rostovka the beef/dairy products were mostly home-sourced. Overall, most people at the study site consumed significantly more meat and dairy product from their self-owned cattle, being approximately twice as often as commercial dairy/beef products.

Mean consumption frequencies of tomatoes and cucumbers are shown in Table 35. Similar to beef and diary product, the people living in the riverine villages Chkalovo, Samarkand and Rostovka consumed significantly more vegetables from their own gardens than from the local market. Those living in Temirtau ate vegetables equally from market and from their own gardens.

The residential properties in the riverine villages provided large garden spaces and water sources for the residents to grow fruit and vegetables. Conversely, the residents in the Temirtau town, particularly in the new district, typically lived in apartments with less space for agriculture. Some residents of this region owned dacha on the hills, but these private gardens or farms were not commonly affordable for the general public. Even if they owned a dacha, these owners could only go to their dachas at weekends. In other periods, vegetables were bought from markets.

Table 35 Mean consumption frequencies of tomatoes and cucumbers from different sources at the study site

Food intake frequency (meal/day)	Tomatoes / Mean±SD			Cucumbers / Mean±SD		
	Total	Home	Market	Total	Home	Market
Temirtau old town	0.53±0.33	0.26±0.29	0.23±0.30	0.52±0.40	0.27±0.30	0.22±0.32
Temirtau new town	0.43±0.32	0.21±0.28	0.20±0.25	0.42±0.28	0.18±0.22	0.20±0.22
Chkalovo	0.60±0.46	0.53±0.44	0.07±0.09	0.45±0.35	0.39±0.32	0.06±0.07
Gagarinskoye	0.54±0.40	0.32±0.34	0.20±0.31	0.52±0.41	0.30±0.33	0.17±0.30
Samarkand	0.56±0.43	0.42±0.43	0.14±0.16	0.60±0.45	0.41±0.44	0.14±0.17
Rostovka	0.45±0.30	0.36±0.30	0.09±0.12	0.39±0.29	0.33±0.30	0.06±0.09
Mean (n= 218)	0.52±0.38	0.33±0.35	0.17±0.24	0.48±0.37	0.30±0.32	0.15±0.24

The sources of the food supply varied seasonally. In summer, when this fieldwork was finished, the residents could go fishing or gardening. However, in winter, the outdoor activities ceased due to the extreme cold weather. Hence, the consumption of fresh food decreased as it was purchased at the markets or shops.

Table 36 shows that the meal sizes of beef, dairy product, tomatoes and cucumbers were varied from 0 to 3 kg. In this survey, male participants, particularly outdoor male workers, responded that a large quantity of food was needed in order to maintain their physical energy.

Table 36 Distributions of meal portion (g/meal) for food items

	Daily fish portion (g/meal)			
	Mean	SD	Min	Max
Beef (n=223)	151.53	126.55	0	1000
Dairy (n=223)	316.08	373.43	0	3000
Tomatoes (n=224)	246.96	168.79	0	1000
Cucumbers (n=227)	199.87	150.61	0	1000

The distributions and the 95th percentile data of food mean intake frequencies (meal/day) and meal sizes (g/meal) for the whole population are given in Table 37. The data for separated residential areas and the distribution curves are listed in Appendix G. The distribution data were site-specific based on the questionnaire information.

Table 37 Distributions of food meal portions and frequencies of whole population

Media/Area	Distribution type (site-specific)	Distribution parameters (site-specific)	95th-tile
Other food intake frequency (unit: meals/day)			
Beef	Gamma	Location =0; scale = 0.64; shape = 0.56	1
Dairy	Beta	Min. = -0.16; Max. = 3.31; alpha = 2.03; beta = 4.72	2
Tomatoes	Gamma	Location =-0.03; scale = 0.31; shape = 1.73	1
Cucumbers	Gamma	Location =-0.02; scale = 0.31; shape = 1.73	1
Other food meal portion (unit: g/meal)			
Beef	Logistic	Mean = 139.71; scale = 64.52	333.00
Dairy	Gamma	Location = -3.75; scale = 332.76; shape = 0.96	1000.00
Tomatoes	Maximum Extreme	Likeliest = 182.49; scale = 116.15	473.97
Cucumbers	Maximum Extreme	Likeliest = 138.84; scale = 100.62	300.00

4.4 Exposure levels

The composition of the population that took part in the survey is shown in Table 38, and the distribution of Hg concentrations in the 289 hair samples is shown in Table 39 and Figure 21. There were two samples whose concentrations of Hg were below the detection limit (< 2 ng/g). The overall Hg concentration in the hair of the local people aged 2-83 years old was from 0.009 to 5.184 µg/g with the mean of 0.577 µg/g.

The hair samples were also collected from a non-Hg-polluted city of Almaty in Kazakhstan regarded as a control group. The Hg concentrations in the control group consisting of thirteen people ranged from 0.310 to 0.896 µg/g with the mean of 0.245 µg/g. The average Hg concentration in the hair collected from the study areas were twice as high as the control group. Nevertheless, Hg levels in the hair of the study population were lower than expected, since high Hg levels were observed in their environment. Hair Hg levels at the study site were generally moderate and similar to the school children exposed to Hg due to dental amalgam in Spain (geometric mean: 0.77 µg/g, range: 0.18-2.44 µg/g) and the subjects with dental amalgam exposure in Albania (mean: 0.705 µg/g, range: 0.195-1.955 µg/g) (Babi et al., 2000, Batista et al., 1996). On average the Hg levels in the study population's hair were lower than the residents in Seoul in South Korea, a city with relatively low Hg exposure and with a mean hair Hg concentration of 1.7 µg/g in males and 1.1 µg/g in females (Lee et al., 2000, Harada et al., 1998, Airey, 1983b). Furthermore, the hair Hg levels in this study were appreciably lower than some Hg-contaminated areas such as Minamata in Japan where Hg concentrations ranged from 2.46-705 µg/g (Harada, 1995). The frequent fish eaters of Cambodia had Hg concentrations in hair with a mean of 7.3 µg/g and a range 0.54-190 µg/g (Agusa et al., 2005) and the Brazilians living in Sai Cinza near a gold mining area with a mean of 16 µg/g and a range of 4.5-90.4 µg/g (Santos et al., 2002). The comparison may imply that frequent consumption of fish and shellfish in those areas lead to higher levels of Hg in people's hair than the Temirtau site. For example, in the Hg poising case of Minamata, the extremely high levels of Hg in hair was due to Japanese tradition of eating sushi i.e. raw fish, which had Hg levels over 1ppm and greatly increase Hg intake doses in human bodies (Wheeler, 1996), whereas in Kazakhstan fish is often cooked for appreciable length of time which is known to drive off much of the organic Hg (Figure 21).

Table 38 Composition of participants in questionnaire survey

Item	n	%	Item	n	%
1. Sex			**7. Where do you get your drinking water?**		
Male	114	39.66	Central water supply	198	68.62
Female	175	60.34	Well	32	11.03
			Borehole	50	17.24
2. Age (year-old)			Others	6	2.07
Under 16	34	11.72			
16-30	80	27.59	**8. Have you coloured or chemical treated your hair in the past month?**		
31-45	69	23.79			
45 above	106	36.90	No	206	71.38
			Yes	79	27.24
3. Ethnic origin			Missing	4	1.38
Russian	180	62.07			
Kazakh	38	13.45	**9. If you are a female, are you pregnant now?**		
Tartar	21	7.24	No	168	96.00
German	27	9.31	1-month	2	1.14
Ukrainian	11	3.79	2-month	1	0.57
Greek	6	2.07	3-month	1	0.57
Others	3	1.03	4-month	1	0.57
Missing	3	1.03	6-month	1	0.57
			8-month	1	0.57
4. Occupation					
Never worked in the acetaldehyde plant	261	90.34	**10. Did you eat fish in the past one year?**		
Worked/working in the acetaldehyde plant	23	7.93	No	20	6.90
Missing	5	1.72	Yes	266	92.07
			Missing	3	1.03
5. Body weight (kg)					
Under 40	17	5.86	**11. Fishermen?**		
40-60	88	30.34	No	264	91.38
60-80	121	42.07	Yes	25	8.62
80-100	57	19.66			
Over 100	6	2.10			
6. Residential area					
Temirtau Old Town	50	17.24			
Temirtau New Town	50	17.24			
Chkalova	46	15.86			
Gagarinskoye	68	23.79			
Samarkand	43	14.83			
Roskovka	32	11.03			
All	**289**	**100**	**All**	**289**	**100**

Although low Hg levels in hair were generally observed in this survey, there were still 12.9% of the hair samples that exceeded the safety standard 1 µg/g for hair Hg, corresponding to the reference of dose (RfD) 0.1 µg/kg body weight/day developed by US EPA (1997d). The population may be at risk for health impacts due to elevated Hg exposure, but the risk was likely to be low for most people, although a small proportion of the population had levels over four times higher.

Table 39 Distribution of Hg concentrations in hair at the study site (n= 287)

	Hair Hg (µg/g)
Mean	0.577
SD	0.843
Min.	0.009
Max.	5.184
25^{th}-tile	0.153
50^{th}-tile	0.273
75^{th}-tile	0.622
95^{th}-tile	2.449

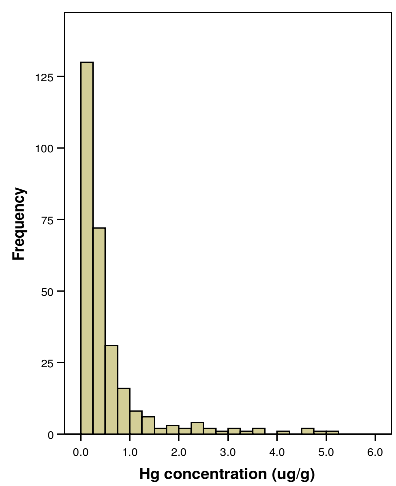


Figure 21 Frequency distribution of Hg concentrations in hair at the study site

5. RESULTS OF RISK ASSESSMENT FOR HUMANS EXPOSED TO MERCURY

Hg contamination was tested in the environmental media at the study site, based on the survey results. Hg levels in the soils and surface water located relatively close to the polluted sites were higher than the further downstream. The contaminant was also seen to be transferred to the local food chains and accumulated particularly in river fish. Due to the effective bioavailability of MeHg, the common form of Hg in fish and shellfish, the health impacts were likely to be more problematic on the riverside population at the study site. Furthermore, compared with MeHg, elemental Hg and inorganic Hg compounds are hardly absorbed by biological species. In an effort to estimate the levels of human exposure to Hg accurately, the methylation rates in the environmental compartments at the study site were considered in the simulations of Hg exposure risk for the potentially exposed population.

This chapter described the simulations of health risk of the study populations exposed to Hg via several potential pathways. The survey results presented in Chapter 4 were adopted as input values regarding site-specific exposure parameters in the simulations. Both probabilistic and deterministic approaches were used to simulate the exposure risks, which were presented as Hazard quotient (HQ) and Hazard index (HI) values. Hazard quotient or Hazard index greater than one indicated that the adverse health effects due to Hg were likely to occur. As regional differences of Hg contamination levels were observed in the survey data, the human exposure estimates were stratified into six residences. The estimates of human exposure aimed at answering these problems: (a) via which critical exposure media and routes that the local people were exposed to Hg, (b) the most exposed area where the residents had potential adverse health problems and their threatening exposure levels, and (c) the influential variables that accounted for the most variance in the simulations determined by sensitivity analysis.

5.1 Risk assessment using Monte Carlo with assumption of total Hg as MeHg

The more potential pathways of human exposure to Hg at the study site were identified as being accidental soil ingestion, dermal contact of soils, soil ingestion from vegetables, water ingestion, dermal contact of water during showering and swimming, accidental water ingestion while swimming and ingestion of foodstuffs including fish and shellfish, beef, dairy product, tomatoes and cucumbers. Hg intake via inhalation of outdoor air is estimated in Section 5.4, as the exposure to gaseous Hg is mainly elemental Hg which results in a different definition of the safety level from the previous exposure pathways. Figure 22 shows that the average daily intake doses of a Monte Carlo simulation for the study population follows a log-normal distribution (mean: 0.11±0.22 μg/kg body weight/day). The individual distributions in the six residential areas are presented in Appendix H. Table 40 gives the modeled average daily doses (ADDs) of Hg intake and the health risks via the modeled pathways in the six residential areas when total Hg measured in the media is considered as MeHg. The simulated ADD for the population as a whole was 0.097 and 0.24 μg/kg body weight/day at the median and the reasonable maximum exposure (RME, the 95^{th} percentile) levels, respectively. The estimated average Hg daily doses were the lowest in Chkalovo and the highest average Hg daily doses were simulated in Samarkand. Overall, the simulated ADDs of the six residential areas were comparable with the differences of less than one magnitude between the areas.

Fish and shellfish consumption was estimated to the most crucial pathway for Hg exposure in every residential area. The highest daily Hg intake dose through this route was in Samarkand. In addition to fish and shellfish, and contrary to what might be expected, consumption of cucumbers accounted for high levels of Hg exposure. Overall, the estimated Hg exposure doses of the study population were mainly via dietary intake and not via the environmental media.

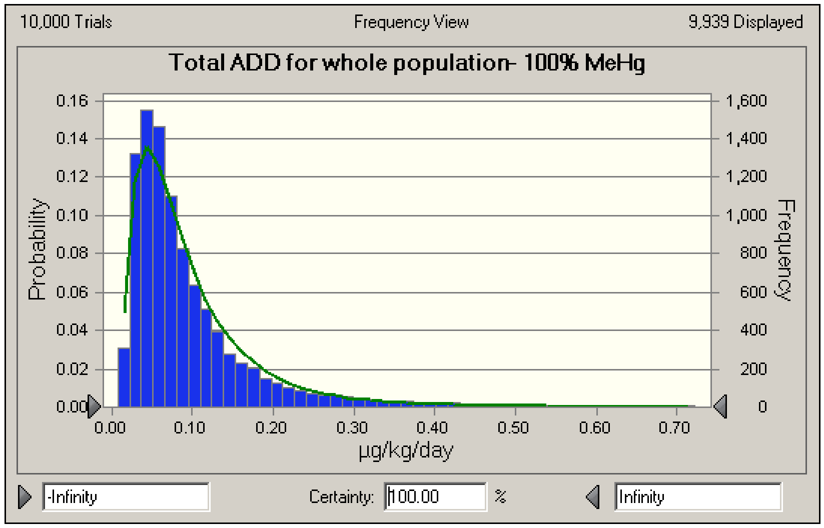


Figure 22 Probability distribution of the predicted ADDs for whole study population with assumption of total Hg as MeHg

Table 40 Monte Carlo simulation estimated Hg exposure and non-cancer hazard quotient for the population at the 50th and 95th percentile at the study site with assumption of 100% MeHg

Area/Pathway	ADD (µg/kg body weight/day)		Hazard quotient	
	50th-tile	95th-tile	50th-tile	95th-tile
Temirtau old town				
Soil ingestion	5.7E-04	5.2E-03	0.01	0.05
Soil dermal contact	1.6E-05	5.3E-04	0.00	0.03
Soil from vegetables	2.8E-06	1.9E-05	0.00	0.00
Water ingestion	6.7E-05	1.7E-03	0.00	0.02
Dermal contact in shower	1.5E-09	2.4E-08	0.00	0.00
Accidental ingestion while swimming	4.4E-07	1.2E-05	0.00	0.00
Dermal contact while swimming	3.2E-09	8.9E-08	0.00	0.00
Fish/shellfish ingestion	5.4E-02	1.6E-01	0.54	1.61
Beef ingestion	2.9E-04	3.1E-03	0.00	0.03
Dairy ingestion	2.1E-03	1.2E-02	0.02	0.12
Tomato ingestion	1.2E-02	3.2E-02	0.12	0.32
Cucumber ingestion	1.6E-02	8.2E-02	0.16	0.82
Total/Hazard index	9.7E-02	2.4E-01	0.84	2.99
Temirtau new town				
Soil ingestion	3.1E-05	2.8E-04	0.00	0.00
Soil dermal contact	7.1E-07	2.5E-05	0.00	0.00
Soil from vegetables	1.3E-06	5.4E-06	0.00	0.00
Water ingestion	1.5E-05	3.7E-05	0.00	0.00
Dermal contact in shower	8.8E-10	1.7E-09	0.00	0.00
Accidental ingestion while swimming	3.1E-07	2.0E-06	0.00	0.00
Dermal contact while swimming	2.3E-09	1.4E-08	0.00	0.00
Fish/shellfish ingestion	3.8E-02	1.6E-01	0.38	1.63
Beef ingestion	2.8E-04	3.2E-03	0.00	0.03
Dairy ingestion	2.3E-03	1.4E-02	0.02	0.14
Tomato ingestion	1.1E-02	3.0E-02	0.11	0.30
Cucumber ingestion	1.2E-02	5.6E-02	0.12	0.56
Total/Hazard index	7.6E-02	2.2E-01	0.65	2.66

(continued)

Area/Pathway	ADD (µg/kg body weight/day)		Hazard quotient	
	50th-tile	95th-tile	50th-tile	95th-tile
Chkalovo				
Soil ingestion	4.0E-04	6.9E-03	0.00	0.07
Soil dermal contact	1.1E-05	4.7E-04	0.00	0.02
Soil from vegetables	5.9E-06	4.6E-05	0.00	0.00
Water ingestion	2.9E-05	5.0E-04	0.00	0.00
Dermal contact in shower	1.5E-09	2.6E-08	0.00	0.00
Accidental ingestion while swimming	3.6E-05	2.4E-04	0.00	0.00
Dermal contact while swimming	2.7E-07	1.7E-06	0.00	0.00
Fish/shellfish ingestion	1.2E-02	6.7E-02	0.12	0.67
Beef ingestion	6.1E-04	2.9E-03	0.01	0.03
Dairy ingestion	2.5E-03	1.6E-02	0.03	0.16
Tomato ingestion	8.7E-03	2.8E-02	0.09	0.28
Cucumber ingestion	1.1E-02	5.2E-02	0.11	0.52
Total/Hazard index	4.5E-02	1.2E-01	0.36	1.76
Gagarinskoye				
Soil ingestion	4.0E-04	6.9E-03	0.00	0.07
Soil dermal contact	1.2E-06	3.4E-05	0.00	0.00
Soil from vegetables	1.6E-06	1.1E-05	0.00	0.00
Water ingestion	5.0E-05	7.9E-04	0.00	0.01
Dermal contact in shower	2.5E-09	4.0E-08	0.00	0.00
Accidental ingestion while swimming	3.1E-05	1.5E-04	0.00	0.00
Dermal contact while swimming	2.3E-07	1.1E-06	0.00	0.00
Fish/shellfish ingestion	2.7E-02	2.1E-01	0.27	2.08
Beef ingestion	2.8E-04	4.1E-03	0.00	0.04
Dairy ingestion	1.6E-03	8.2E-03	0.02	0.08
Tomato ingestion	8.3E-03	2.5E-02	0.08	0.25
Cucumber ingestion	1.2E-02	6.8E-02	0.12	0.68
Total/Hazard index	5.9E-02	2.7E-01	0.49	3.21
Samarkand				
Soil ingestion	3.7E-05	1.4E-04	0.00	0.00
Soil dermal contact	1.1E-06	1.8E-05	0.00	0.00
Soil from vegetables	6.8E-06	1.1E-05	0.00	0.00
Water ingestion	8.0E-04	2.0E-03	0.01	0.02
Dermal contact in shower	4.6E-08	9.3E-08	0.00	0.00
Accidental ingestion while swimming	2.3E-05	9.7E-05	0.00	0.00
Dermal contact while swimming	1.7E-07	7.2E-07	0.00	0.00
Fish/shellfish ingestion	9.2E-02	4.7E-01	0.92	4.66
Beef ingestion	5.2E-04	3.9E-03	0.01	0.04
Dairy ingestion	2.1E-03	1.9E-02	0.02	0.19
Tomato ingestion	9.0E-03	2.7E-02	0.09	0.27
Cucumber ingestion	1.5E-02	6.7E-02	0.15	0.67
Total/Hazard index	1.3E-01	4.9E-01	1.19	5.85
Rostovka				
Soil ingestion	3.2E-05	1.2E-04	0.00	0.00
Soil dermal contact	9.3E-07	1.6E-05	0.00	0.00
Soil from vegetables	6.8E-06	1.3E-05	0.00	0.00
Water ingestion	1.6E-05	4.5E-05	0.00	0.00
Dermal contact in shower	8.9E-10	2.1E-09	0.00	0.00
Accidental ingestion while swimming	1.5E-05	8.3E-05	0.00	0.00
Dermal contact while swimming	1.1E-07	6.0E-07	0.00	0.00
Fish/shellfish ingestion	5.4E-02	3.8E-01	0.54	3.79
Beef ingestion	3.9E-04	2.3E-03	0.00	0.02
Dairy ingestion	1.5E-03	9.9E-03	0.01	0.10
Tomato ingestion	9.3E-03	2.1E-02	0.09	0.21
Cucumber ingestion	1.2E-02	5.2E-02	0.12	0.52
Total/Hazard index	8.5E-02	4.0E-01	0.77	4.64

(continued)

Area/Pathway	ADD (µg/kg body weight/day)		Hazard quotient	
	50th-tile	95th-tile	50th-tile	95th-tile
Total population				
Soil ingestion	2.3E-04	6.3E-03	0.00	0.06
Soil dermal contact	6.2E-06	4.2E-04	0.00	0.02
Soil from vegetables	7.5E-06	2.3E-05	0.00	0.00
Water ingestion	3.6E-05	5.9E-04	0.00	0.01
Dermal contact in shower	1.9E-09	2.8E-08	0.00	0.00
Accidental ingestion while swimming	1.9E-05	1.7E-04	0.00	0.00
Dermal contact while swimming	1.3E-07	1.3E-06	0.00	0.00
Fish/shellfish ingestion	3.0E-02	2.2E-01	0.30	2.23
Beef ingestion	3.3E-04	3.5E-03	0.00	0.03
Dairy ingestion	2.0E-03	1.5E-02	0.02	0.15
Tomato ingestion	1.0E-02	2.8E-02	0.10	0.28
Cucumber ingestion	1.2E-02	6.8E-02	0.12	0.68
Total/Hazard index	7.0E-02	2.9E-01	0.40	3.47

A hazard quotients (HQ) of less than one in the various pathways showed that the non-carcinogenic risks due to Hg exposure were unlikely to occur at the median estimate level (the 50th percentile). However, a hazard index (HI) greater than one was found for the Samarkand population. It indicated that more than 50% of the population in Samarkand may be potentially at risk of harmful health under the synergy of Hg exposure via these routes. In terms of the RME (the 95th percentile), the hazard quotients exceeded one as a result of the fish and shellfish ingestion pathway. High levels of Hg intake doses through fish and shellfish ingestion resulted in the highest hazard index, up to 5.85 in Samarkand, followed by the villages of Rostovka and Gagarinskoye.

The exposure pathway of cucumber consumption accounted for the next highest risk intake route of Hg exposure. Nevertheless, the levels of the exposure risk via cucumber consumption maintained a safe level even at the 95th percentile level in all the six residences.

Low levels of Hg exposure were attributed to be from soils, as the hazard quotients representing the exposure of accidental ingestion and dermal contact of soils were less than 0.1. The RME levels in the regions of Temirtau old town and Chkalovo were appreciably higher. The increasing risk of the adverse health effects is most likely to be a result of the polluted chemical plant and its waste deposits, as the areas are adjacent to the heavily Hg-contaminated factory of AO Karbide, the Main drain and the outfall.

Approximately 22% of the variation in the ADD simulation for the study population was accounted for by the parameter of body weight of individuals (Figure 23). This is not surprising as heavier people need a higher average daily intake dose than small people to achieve the same concentrations of Hg in their tissues. The negative relationship between the ADD and the parameter of body weight confirmed this. Apart from body weight, the most influential parameter was average intake frequency of river carp. Dietary intake patterns of river fish species including bream, roach and perch also played an important role on daily intake doses of Hg. In addition to fish, ingestion of tomatoes and cucumbers, particularly the later, affected daily Hg intake doses. The vegetable-related parameters accounted for approximately 22% of the variation in the ADD simulation.

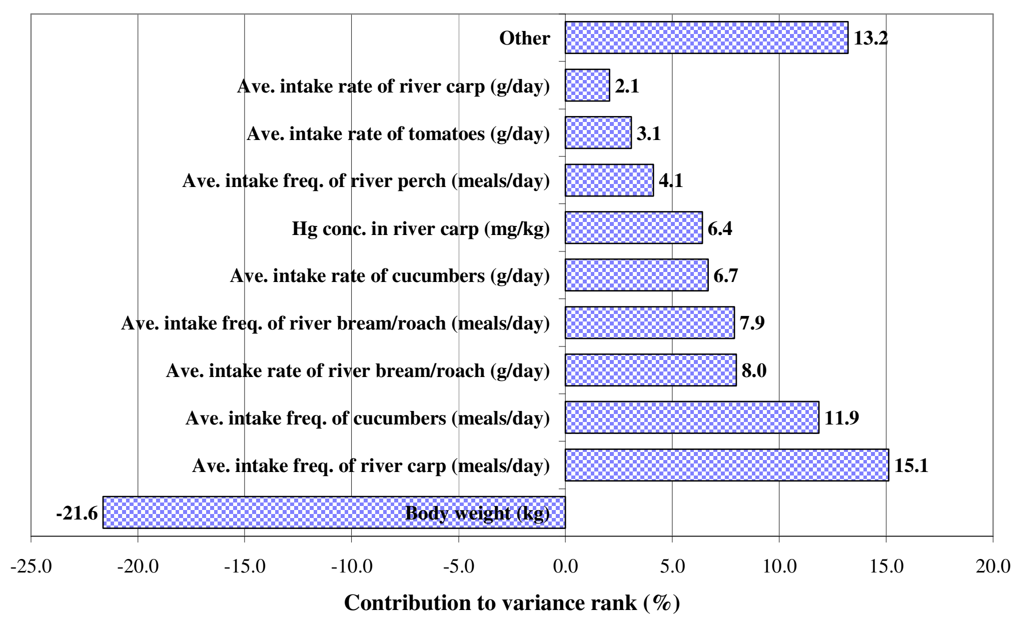


Figure 23 Sensitivity analysis of the ADD simulation with assumption of 100% MeHg

5.2 Risk assessment using Monte Carlo simulation with assumption of varied proportion of MeHg

In the second scenario, methylation rates of 1.5% in soils, 5% in water and 25% in foodstuffs were taken into account. Exposure pathways of ingestion and dermal contact of soils, and ingestion of fish and shellfish, beef, dairy product, tomatoes and cucumbers were selected in the assessment, as these routes were shown in the previous section to pose a measurable risk in the previous simulation.

Figure 24 shows that the average daily intake doses of Hg simulated for the study population as a whole follows a log-normal distribution (mean: 0.08 µg/kg body weight/day, standard deviation: 0.33). The distributions in the six residential areas are illustrated in Appendix H. For the whole population, the ADD was estimated to be 0.04 µg/kg body weight/day at the median estimate level and 0.24 µg/kg body weight/day at the upper 95% estimate level (Table 41). Similar to the previous simulation, the residents of Samarkand were shown to have the highest daily intake doses of Hg, whereas those living in Chkalovo had the lowest intake of Hg, being approximately one-fifth of the intake of those in Samarkand. Despite the comparable daily intake doses of Hg in Temirtau old town and Rostovka at the 50^{th} percentile, the intake dose in Rostovka was higher than in Temirtau old town in the RME. The high daily intake doses simulated at the 95^{th} percentile for the exposed population in Rostovka may be attributed to larger river fish meal portions and higher river fish intake frequencies by the local people in this riverine village than those in Temirtau old town.

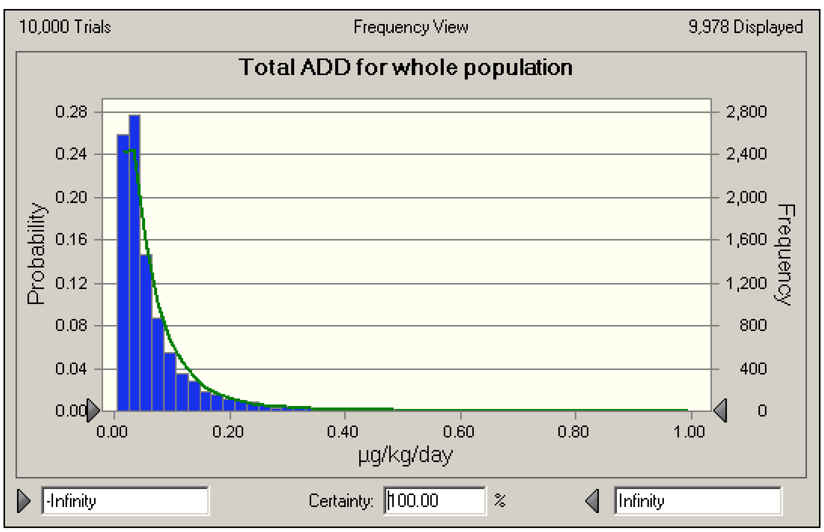


Figure 24 Probability distribution of the predicted ADDs for whole study population with assumption of proportional MeHg to total Hg in various exposure pathway

Table 41 Estimated Hg exposure and non-cancer risk for the population at the study site with assumption of varied methylation rates in the exposure media

Area/Pathway	ADD (µg/kg body weight/day)		Hazard quotient	
	50th-tile	95th-tile	50th-tile	95th-tile
Temirtau old town				
Soil ingestion	1.2E-04	1.0E-03	0.00	0.01
Soil dermal contact	1.8E-06	6.2E-05	0.00	0.00
Fish/shellfish ingestion	5.4E-02	1.6E-01	0.54	1.61
Beef ingestion	7.3E-05	8.2E-04	0.00	0.01
Dairy ingestion	5.1E-04	3.5E-03	0.01	0.03
Tomatoes ingestion	3.0E-03	7.3E-03	0.03	0.08
Cucumbers ingestion	3.8E-03	1.5E-02	0.04	0.21
Total/Hazard index	6.5E-02	1.7E-01	0.61	1.95
Temirtau new town				
Soils ingestion	6.2E-06	5.6E-05	0.00	0.00
Soil dermal contact	8.1E-08	2.9E-06	0.00	0.00
Fish/shellfish ingestion	3.8E-02	1.6E-01	0.38	1.63
Beef ingestion	6.7E-05	8.2E-04	0.00	0.01
Dairy ingestion	5.7E-04	3.5E-03	0.01	0.03
Tomatoes ingestion	2.9E-03	7.3E-03	0.03	0.07
Cucumbers ingestion	3.2E-03	1.5E-02	0.03	0.15
Total/Hazard index	4.8E-02	1.7E-01	0.45	1.89
Chkalovo				
Soils ingestion	8.0E-05	1.3E-03	0.00	0.01
Soil dermal contact	1.2E-06	5.7E-05	0.00	0.00
Fish/shellfish ingestion	1.2E-02	6.7E-02	0.12	0.67
Beef ingestion	1.5E-04	7.3E-04	0.00	0.01
Dairy ingestion	6.3E-04	4.0E-03	0.01	0.04
Tomatoes ingestion	2.2E-03	7.0E-03	0.02	0.07
Cucumbers ingestion	2.7E-03	1.3E-02	0.03	0.13
Total/Hazard index	2.1E-02	7.7E-02	0.18	0.94
Gagarinskoye				
Soils ingestion	8.5E-06	7.0E-05	0.00	0.00
Soil dermal contact	1.3E-07	3.6E-06	0.00	0.00
Fish/shellfish ingestion	2.7E-02	2.1E-01	0.27	2.08
Beef ingestion	7.2E-05	9.8E-04	0.00	0.01
Dairy ingestion	4.0E-04	2.0E-03	0.00	0.02
Tomatoes ingestion	2.1E-03	6.5E-03	0.02	0.06
Cucumbers ingestion	2.9E-03	1.7E-02	0.03	0.17
Total/Hazard index	3.5E-02	2.3E-01	0.32	2.34
Samarkand				
Soils ingestion	7.5E-06	2.7E-05	0.00	0.00
Soil dermal contact	1.2E-07	2.0E-06	0.00	0.00
Fish/shellfish ingestion	9.2E-02	4.7E-01	0.92	4.66
Beef ingestion	1.3E-04	9.9E-04	0.00	0.01
Dairy ingestion	5.2E-04	4.5E-03	0.01	0.05
Tomatoes ingestion	2.2E-03	7.0E-03	0.02	0.07
Cucumbers ingestion	3.8E-03	1.7E-02	0.04	0.17
Total/Hazard index	1.0E-01	4.4E-01	0.98	4.95
Rostovka				
Soils ingestion	6.4E-06	2.4E-05	0.00	0.02
Soil dermal contact	1.0E-07	1.8E-06	0.00	0.00
Fish/shellfish ingestion	5.4E-02	3.8E-01	0.54	3.79
Beef ingestion	9.7E-05	5.7E-04	0.00	0.01
Dairy ingestion	3.6E-04	2.5E-03	0.00	0.02
Tomatoes ingestion	2.3E-03	5.1E-03	0.02	0.05
Cucumbers ingestion	2.9E-03	1.3E-02	0.03	0.13
Total/Hazard index	6.0E-02	3.8E-01	0.59	4.00

(continue)

Area/Pathway	ADD (µg/kg body weight/day)		Hazard quotient	
	50^{th}-tile	95^{th}-tile	50^{th}-tile	95^{th}-tile
Total population				
Soils ingestion	4.8E-05	1.3E-03	0.00	0.01
Soil dermal contact	7.6E-07	4.9E-05	0.00	0.00
Fish/shellfish ingestion	3.0E-02	2.2E-01	0.30	2.23
Beef ingestion	8.3E-05	8.7E-04	0.00	0.01
Dairy ingestion	5.0E-04	3.9E-03	0.01	0.04
Tomatoes ingestion	2.5E-03	7.0E-03	0.03	0.07
Cucumbers ingestion	3.0E-03	1.7E-02	0.03	0.17
Total/Hazard index	4.0E-02	2.4E-01	0.36	2.53

As might be expected, fish and shellfish ingestion accounted for the largest proportion of Hg intake in all residential areas, with up to 70% of the total Hg intake being attributed to this exposure route.

Non-carcinogenic risks illustrated in Figure 25 represent the level of Hg exposure for the population as a whole, with the level of risk being below the safety level (hazard index <1) at the 50^{th} percentile. However, at the 95^{th} percentile, the hazard index for the population as a whole was greater than one, indicating the potential health risk on the high exposure group (Figure 26). The highest hazard index was predicted to be in Samarkand at 5, followed by Rostovka. The minimal level of Hg exposure risk was in Chkalovo where the hazard index was predicted less than one even in the RME.

The results show that fish and shellfish consumption pose the major pathway for exposure to Hg in every residential area. It confirmed the works of others that the aquatic food chain is the main route for MeHg exposure to humans (NRC, 2000, USEPA, 1997c, WHO, 1990, Airey, 1983a, Birke et al., 1972, Johnsson et al., 2004, Morrissette et al., 2004, Sherlock and Quinn, 1988, Shipp et al., 2000, Tran et al., 2004). It is also clear that the consumption of fish and shellfish in the study area appears to be related to elevated levels of Hg in the population.

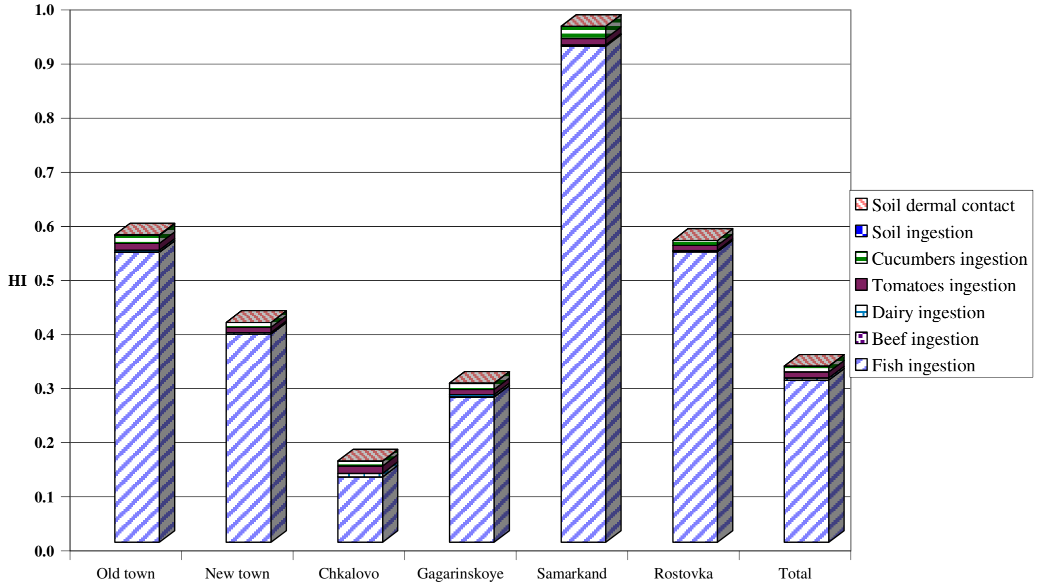


Figure 25 Hazard quotients and Hazard indices at 50th percentile in different residential areas

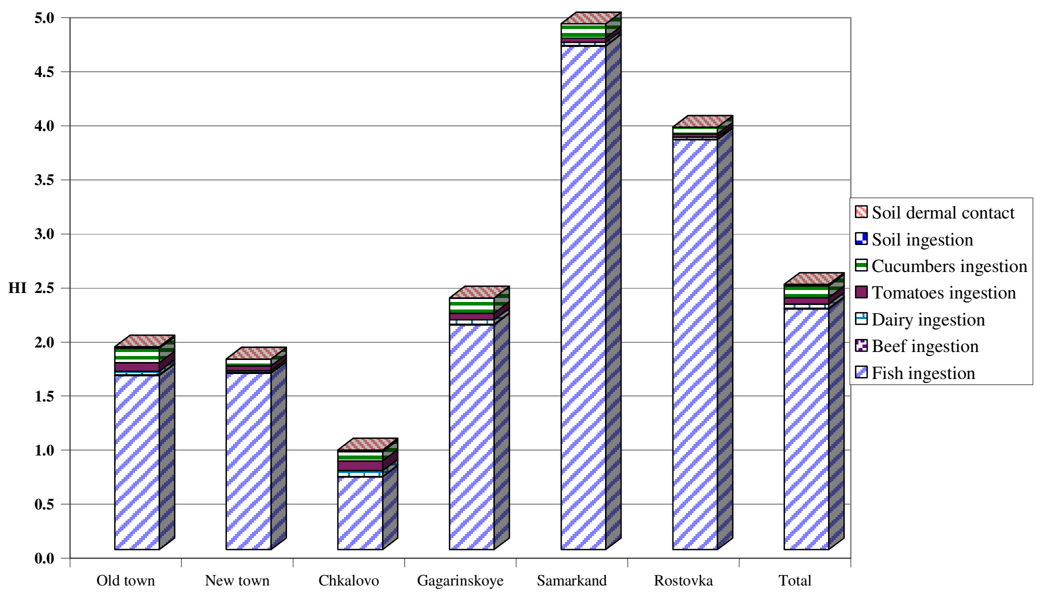


Figure 26 Hazard quotients and hazard indices at 95th percentile in different residential areas

Comparable and low levels of Hg exposure risk through cucumber consumption were observed in all six residential areas. Human exposure to Hg through this route is currently insignificant for the local people's health. This is a result of low proportions of MeHg in vegetables in general, and vegetables were unlikely to be the crucial exposure route to cause adverse health effects.

Figure 27 illustrates a sensitivity analysis of the Hg exposure dose simulation for the total population. The most influential parameter was the average intake frequency of river carp, which accounted for 23.5% of the output variance. The average intake frequency of river bream and roach was the next most significant variable. Of the ten most important parameters that affected the ADD simulation, eight were fish-related variables. Dietary intake patterns of fish, particularly meal frequency and intake rate of river fish, were the key parameters to influence the variance in the simulation result.

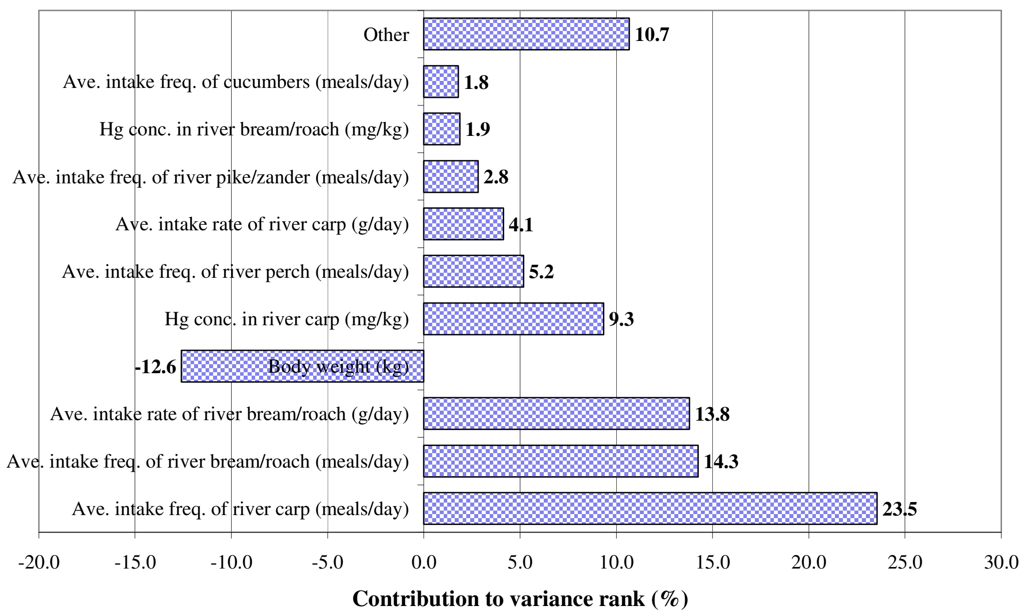


Figure 27 Sensitivity analysis of the ADD simulation with assumption of proportional MeHg in total Hg in various exposure pathways

5.3 Risk assessment using deterministic method of 95th percentile values

With the deterministic method and the assumption that all observed Hg concentrations in various media are MeHg, the evaluated mean ADD for Hg for the whole population was 0.88 µg/kg body weight/day (Table 42). The simulation showed that the highest ADD was in Samarkand (ADD: 1.6 µg/kg body weight/day), followed by Rostovka (ADD: 0.98 µg/kg body weight/day). Although food ingestion unsurprisingly accounted for the most significant proportion of Hg intake, the exposure pathways of ingestion and dermal contact of soils and ingestion of water were evaluated to contribute to larger amounts of Hg intake doses in Temirtau old town and Chkalovo than other residential areas.

Table 42 Estimated Hg exposure and non-cancer risk for the population at the study site using the deterministic method

Area/Pathway	100% MeHg		Varied MeHg proportions	
	ADD (µg/kg body weight/day)	Hazard Quotient	ADD (µg/kg body weight/day)	Hazard Quotient
Temirtau old town				
Soil ingestion	5.2E-03	0.05	1.0E-03	0.01
Soil dermal contact	4.5E-03	0.22	5.2E-04	0.02
Soil from vegetables	4.6E-05	0.00	-	-
Water ingestion	7.4E-03	0.07	-	-
Dermal contact in shower	1.5E-07	0.00	-	-
Accidental ingestion while swimming	9.5E-05	0.00	-	-
Dermal contact while swimming	7.8E-07	0.00	-	-
Fish/shellfish ingestion	3.3E-01	3.29	3.3E-01	3.29
Beef ingestion	2.8E-03	0.03	7.1E-04	0.01
Dairy ingestion	9.8E-03	0.10	2.4E-03	0.02
Tomato ingestion	3.5E-02	0.35	8.6E-03	0.09
Cucumber ingestion	8.9E-02	0.89	2.2E-02	0.22
Total/Hazard index	4.8E-01	5.00	3.6E-01	3.67
Temirtau new town				
Soil ingestion	1.9E-04	0.00	3.8E-05	0.00
Soil dermal contact	1.7E-04	0.01	1.9E-05	0.00
Soil from vegetables	4.1E-06	0.00	-	-
Water ingestion	2.0E-05	0.00	-	-
Dermal contact in shower	1.3E-09	0.00	-	-
Accidental ingestion while swimming	3.2E-06	0.00	-	-
Dermal contact while swimming	2.6E-08	0.00	-	-
Fish/shellfish ingestion	5.6E-01	5.58	5.6E-01	5.58
Beef ingestion	4.8E-03	0.05	1.2E-03	0.01
Dairy ingestion	9.9E-03	0.10	2.5E-03	0.02
Tomato ingestion	1.9E-02	0.19	4.7E-03	0.05
Cucumber ingestion	6.0E-02	0.60	1.5E-02	0.15
Total/Hazard index	6.5E-01	6.53	5.8E-01	5.82
Chkalovo				
Soil ingestion	4.6E-03	0.05	9.3E-04	0.01
Soil dermal contact	4.1E-03	0.19	4.6E-04	0.02
Soil from vegetables	4.3E-05	0.00	-	-
Water ingestion	2.1E-03	0.02	-	-
Dermal contact in shower	1.4E-07	0.00	-	-
Accidental ingestion while swimming	3.0E-04	0.00	-	-
Dermal contact while swimming	2.5E-06	0.00	-	-
Fish/shellfish ingestion	1.1E-01	1.09	1.1E-01	1.09
Beef ingestion	3.0E-03	0.03	7.6E-04	0.01
Dairy ingestion	1.7E-02	0.17	4.2E-03	0.04
Tomato ingestion	3.2E-02	0.32	8.1E-03	0.08
Cucumber ingestion	5.8E-02	0.58	1.4E-02	0.14
Total/Hazard index	2.3E-01	2.46	1.4E-01	1.40

(continue)

Area/Pathway	100% MeHg		Varied MeHg proportions	
	ADD (µg/kg body weight/day)	Hazard Quotient	ADD (µg/kg body weight/day)	Hazard Quotient
Gagarinskoye				
Soil ingestion	1.8E-04	0.00	3.6E-05	0.00
Soil dermal contact	1.6E-04	0.01	1.8E-05	0.00
Soil from vegetables	6.2E-06	0.00	-	-
Water ingestion	6.1E-04	0.01	-	-
Dermal contact in shower	4.1E-08	0.00	-	-
Accidental ingestion while swimming	2.1E-04	0.00	-	-
Dermal contact while swimming	1.7E-06	0.00	-	-
Fish/shellfish ingestion	4.6E-01	4.57	4.6E-01	4.57
Beef ingestion	3.4E-03	0.03	8.4E-04	0.01
Dairy ingestion	5.6E-03	0.06	1.4E-03	0.01
Tomato ingestion	1.1E-02	0.11	2.8E-03	0.03
Cucumber ingestion	5.4E-02	0.54	1.4E-02	0.14
Total/Hazard index	5.3E-01	5.33	4.8E-01	4.75
Samarkand				
Soil ingestion	4.4E-05	0.00	8.8E-06	0.00
Soil dermal contact	3.9E-05	0.00	4.4E-06	0.00
Soil from vegetables	5.1E-06	0.00	-	-
Water ingestion	1.0E-03	0.01	-	-
Dermal contact in shower	7.0E-08	0.00	-	-
Accidental ingestion while swimming	1.3E-04	0.00	-	-
Dermal contact while swimming	1.0E-06	0.00	-	-
Fish/shellfish ingestion	1.5E+00	14.66	1.5E+00	14.66
Beef ingestion	2.7E-03	0.03	6.7E-04	0.01
Dairy ingestion	2.0E-02	0.20	5.0E-03	0.05
Tomato ingestion	1.2E-02	0.12	3.0E-03	0.03
Cucumber ingestion	8.9E-02	0.89	2.2E-02	0.22
Total/Hazard index	1.6E+00	15.91	1.5E+00	14.97
Rostovka				
Soil ingestion	3.1E-05	0.00	6.3E-06	0.00
Soil dermal contact	2.7E-05	0.00	3.1E-06	0.00
Soil from vegetables	4.3E-06	0.00	-	-
Water ingestion	1.8E-05	0.00	-	-
Dermal contact in shower	1.2E-09	0.00	-	-
Accidental ingestion while swimming	9.3E-05	0.00	-	-
Dermal contact while swimming	7.6E-07	0.00	-	-
Fish/shellfish ingestion	9.2E-01	9.22	9.2E-01	9.22
Beef ingestion	3.0E-03	0.03	7.4E-04	0.01
Dairy ingestion	7.2E-03	0.07	1.8E-03	0.02
Tomato ingestion	7.8E-03	0.08	2.0E-03	0.02
Cucumber ingestion	3.6E-02	0.36	9.1E-03	0.09
Total/Hazard index	9.8E-01	9.77	9.4E-01	9.36
Total population				
Soil ingestion	4.8E-03	0.05	9.7E-04	0.01
Soil dermal contact	4.2E-03	0.20	4.8E-04	0.02
Soil from vegetables	4.3E-05	0.00	-	-
Water ingestion	9.0E-04	0.01	-	-
Dermal contact in shower	6.0E-08	0.00	-	-
Accidental ingestion while swimming	2.4E-04	0.00	-	-
Dermal contact while swimming	1.9E-06	0.00	-	-
Fish/shellfish ingestion	7.6E-01	7.62	7.6E-01	7.62
Beef ingestion	3.2E-03	0.03	8.1E-04	0.01
Dairy ingestion	1.9E-02	0.19	4.9E-03	0.05
Tomato ingestion	1.8E-02	0.18	4.4E-03	0.04
Cucumber ingestion	6.6E-02	0.66	1.7E-02	0.17
Total/Hazard index	8.8E-01	8.95	7.9E-01	7.92

When hazard quotients and hazard indices were calculated, a similar pattern of high Hg exposure risks via food consumption was observed in the six different residential areas. The hazard quotients of fish and shellfish ingestion were greater than one, indicating that the exposure route was highly likely to pose high levels of Hg exposure risk to the residents. Among the study population, the residents in Samarkand had the highest level of Hg exposure risk via fish and shellfish ingestion, with the hazard quotient higher than 15. The other exposure route was ingestion of cucumbers, which could result in high Hg exposure risk particularly in Temirtau old town and Samarkand. Based on the single-point evaluation, the exposure risk through this route was estimated to exceed the safety level for all the population (hazard index > 1).

The exposure route of dermal contact of soils posed higher health risk for the local people living in Temirtau old town district and Chaklovo than those living in the other areas. With the increasing distance from the primary sources of the contamination like the acetaldehyde factory and Zhaur Swamp, the risk posed by soil was reduced.

In the second scenario where Hg methylation rates in soils, water and foodstuffs were considered as 1.5%, 5% and 25% of total Hg, respectively, the simulated ADDs ranged from the lowest of 0.14 µg/kg body weight/day in Chkalovo to the highest of 1.5 µg/kg body weight/day in Samarkand. Compared with the evaluation with the first scenario, the total amount of daily Hg intake rate in Chkalovo decreased steeply by about 40%, whereas the decreases of the ADDs between the two scenario-based evaluations in the other five areas were less than 25%. This was because the environmental medium of soil accounted for a large proportion of the total intake dose in Chkalovo. As a result, when the likely percentage of MeHg form of Hg in soils was taken into account, the estimated ADD decreased. For the other areas, the evaluated ADDs with the second scenario remained nearly consistent with the first scenario.

In the probabilistic and deterministic simulations, the main pathway of Hg exposure at the study site was consistently demonstrated to be fish and shellfish consumption, but the evaluated ADDs using the deterministic method were close to or higher than the results at the upper percentile (the 95th percentile) in the probabilistic simulations. Some single-point ADD estimates were even three times larger than that at the upper level in probabilistic simulations. In other words, the single-point estimates were relatively conservative. The discrepancies were likely to result from the single-point inputs selected from a wide range of exposure data. Since there is a wide range of variation in the data sets, the deterministic approach with the characteristic of the inflexibility on data selection led to the worse-case estimation, hence it gives a caused a conservative and upper-bound evaluation.

5.4 Risk assessment of air exposure

Figure 28 illustrates the simulated distribution of Hg exposure dose for the study population as a whole, being a maximum extreme distribution (likeliest = 0.03, scale = 0.01). The probabilistic simulation results for the six residential areas are given in Appendix G. Table 43 shows the results of a Monte Carlo simulation of the estimated average daily Hg exposure dose distribution from the outdoor inhalation of air. Not surprisingly, higher Hg exposure doses were estimated in the town of Temirtau than other residential areas. The daily Hg intake doses via outdoor respiration in Temirtau were approximately five-fold higher than the riverine villages. The result implicated that Hg exposure to the population via the atmosphere decreased dramatically with the increasing distance from the main contaminated site.

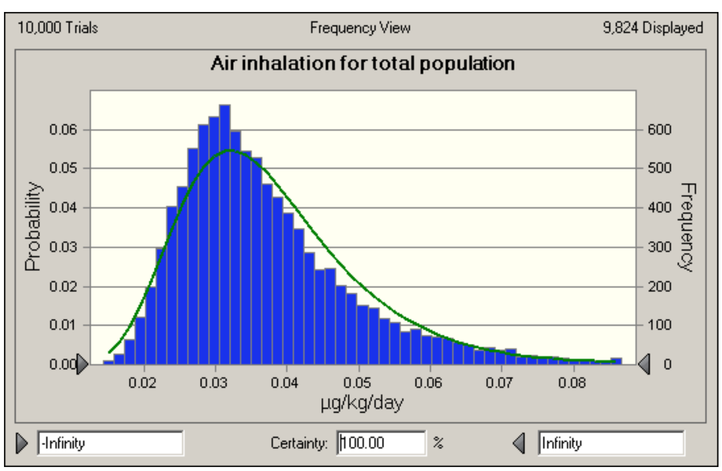


Figure 28 Distribution of Hg exposure dose simulation for the study population as a whole

Table 43 Estimated Hg exposure via outdoor air inhalation for the population at the study site

Area	Monte Carlo simulation for ADD (µg/kg body weight/day)	
	50^{th}-tile	95^{th}-tile
Temirtau old town	3.3E-02	5.7E-02
Temirtau new town	3.4E-02	5.7E-02
Chkalovo	7.1E-03	1.3E-02
Gagarinskoye	8.4E-03	1.7E-02
Samarkand	7.6E-03	1.2E-02
Rostovka	7.7E-03	1.5E-02
Total population	3.4E-02	6.8E-02

The USEPA (1997e) recommends the reference concentration (RfC) of 0.3 µg/m^3 as the threshold level for Hg concentrations in the atmosphere to prevent human health problems. The reason why this safety guideline was developed for elemental Hg is that most Hg in the air is elemental Hg (Clarkson, 1998, USEPA, 1997c). Moreover, the fact that the current data on organic Hg exposure via inhalation are limited and inconclusive (The RAIS website: http://rais.ornl.gov/tox/profiles/methyl_mercury_ragsa.shtml) makes the estimates of the health impacts due to inhalation of organic Hg unavailable. Since Hg concentrations in the atmosphere at the study site were below the RfC level (see section 4.1.4), the adverse health effects were unlikely to occur in the local resident population. Outdoor air inhalation was presently an insignificant pathway for the population exposed to Hg.

5.5 Discussion

Despite the high levels of Hg contamination found in the environmental media at the study site, the predominant exposure pathway for Hg intake was simulated to be via the indirect exposure pathway of dietary intake, particularly fish and shellfish consumption. This result is consistent with several previous studies associated with human exposure to Hg (Birke et al., 1972, Li et al., 2006, Horvat et al., 2003, Goldblum et al., 2006, Constantinou et al., 1995). The result also implies that Hg may have been transported and accumulated significantly in the aquatic food chain at the study site.

The finding indicated high levels of Hg exposure in the villages further downstream. This seems different from what was expected, as Hg levels in the immediate environment close to the primary sources of pollution were found to be higher than the downstream areas. However, Hg is distributed by the river into downstream river silts in the study areas (Figure 15 in Section 3.1.2). It has been observed that although the levels of Hg in the silts decreased downstream, the levels of organic matter increased with distance from the dam. This could have resulted in conduction with higher methylation rates (Ullrich et al., 2007c). Lifestyle and dietary behaviour of the local people were also important in explaining the elevated Hg levels in the population living downstream. Based on the survey data, the residents of Samarkand and Rostovka tended to catch fish from the river or the lakes in the neighbourhood instead of buying it from the local market. Compared with Chkalovo with river fish consumption as low as 0.04 meals/day, Samarkand residents ate 0.22 meals of river fish per day in average. Because of more Hg contents in river fish than market fish, frequent consumption of river fish potentially results in higher levels of exposure risk to the people living the downstream residences.

Temirtau old town and Chkalovo were found to have relatively higher ADDs through accidental soil ingestion. This was due to more severe Hg pollution in the soils and river sediments near the primary Hg-pollution sources than the more distant areas from those hotspots (Yanin, 1997, Stratienko, 2004, Posch & Partners Consulting Engineers, 2004, Heaven et al., 2000b, Heaven et al., 2000a). Hg exposure via accidental soil ingestion may potentially be more elevated in children, particularly those in the period of hand-mouthing behaviour (Department for the Environment Food and Rural Affairs (DEFRA), 2002c). Nevertheless, the current exposure simulation indicated that Hg exposure via accidental ingestion of soils was below the safety level for the study population.

Furthermore, sandstorms coming down from the northern areas occur frequently in summer raising the potential level of Hg exposure. During the season, fine soils are carried by winds and blown into residential areas and communities. To prevent soils and dust entering their houses the local people generally put mats in front of the entrances, take off shoes before getting into the houses, and tidy furniture and rugs regularly. Nevertheless, house dust prevails at the study site. The case that mud and dust carried into the houses by pets is also commonly observed in those residential areas. Such instances may increase the Hg exposure risk via accidental soil ingestion but the significance is waiting for a further study.

The simulation indicated that cucumbers contributed higher proportions of Hg intake doses than the other local foodstuffs, except fish. Although Hg intake doses via cucumber consumption were still below the acceptable limit, this study suggests that Hg exposure through cucumber ingestion is notable, particularly during the harvest season when the local people eat more cucumbers grown in their own gardens. In addition to tomatoes and cucumbers, potatoes were observed to be one of the important staple crops in the local people's diet during the fieldtrip in 2005. Unfortunately, the harvest season of the potatoes is in the autumn, which resulted in the absence of this foodstuff in the food frequency questionnaire. Since three out of eleven samples of potatoes examined contained Hg concentrations exceeding 0.01 mg/kg, the acceptable level recommended by the Kazakh authority, dietary intake of potatoes together with Hg contamination in the potatoes grown locally at the study site should be monitoring in the future.

The local people consumed fresh vegetables in summer, whereas the portions of fresh vegetables were reduced in winter and homemade pickles were consumed instead. Seasonal difference on diet may affect Hg exposure estimates for the study population. As a result, food frequency questionnaire surveys are advised to be carried out in different seasons.

The simulations of human health risks were for adults and youngsters aged over 15, because the simulations need the exposure information associated with dietary behaviour, which may be unreliably provided by young children. Moreover, the sample size of children under 15 years old was too small to obtain significant exposure simulation results in this survey. For the exposure assessment of the children at the contaminated site, a further survey to collect data from this population is needed.

5.6 Summary

This study conducted Hg exposure assessments using the probabilistic approach and obtained simulated ADDs that ranged from 0.07 to 0.29 μg/kg body weight/day when Hg was assumed to be MeHg and from 0.004 to 0.24 μg/kg body weight/day when more realistic levels of MeHg fraction were assumed for different media. Both simulations followed log-normal distribution. The local population were less likely to encounter adverse health effects resulting from Hg exposure, as the hazard indices in the six residential areas were generally less than one at the 50^{th} level. The Samarkand residents were estimated to have the highest exposure risk as indicated by the estimated hazard index shown to be as high as 1.19 at the 50^{th} level. Despite low exposure risk in general, high-end exposed population at the 95^{th} percentile were simulated to have potential risk of Hg exposure via fish and shellfish consumption. People having very large amounts of fish and shellfish consumption were likely to have harmful health effects due to Hg exposure.

The main route of Hg exposure for the study population was fish and shellfish consumption. Within the six residential areas, the residents in Samarkand were predicted to have the highest daily Hg intake dose through this exposure route. Based on the questionnaire data, the residents of the villages frequently consumed self-caught fish from the contaminated River Nura or the local lakes. As river fish have been shown to contain more Hg than imported fish in the market, people consuming more river fish were likely to have elevated Hg levels in their bodies.

The deterministic method showed an agreement with the probabilistic method in that fish and shellfish consumption was considered to be the significant pathway of Hg exposure risk for the local people. However, the outcomes of the former method were similar to or larger than the values at the 95^{th} percentile in the Monte Carlo simulations. It was due to the fact that the worse cases were used in the deterministic method.

6. VERIFICATION OF MODELLED MERCURY EXPOSURE ESTIMATES AGAINST OBSERVED DATA

The verification of the simulations carried out in Chapter 5 for Hg exposure levels is given in this chapter. As the exposure levels via the measurement of the chemical's concentrations in the biomarkers were thought to reflect more accurate burdens of chemicals in individuals than estimation of intake and uptake (Ponce et al., 1998), the average daily Hg intake doses of the study population simulated using both deterministic and probabilistic methods were compared with Hg intake estimates that converted from the observed Hg concentrations in the hair of the same population (See Section 4.4). In this chapter, verification procedures are used to examine (a) the applicability of the deterministic and probabilistic approaches on exposure assessments for Hg, (b) the deviations between questionnaire-based and biomarker-based exposure levels conducted in different residential populations and (c) the validity of the dose-response relationship. Uncertainty in the estimates may result from several sources. The exposure data (i.e. due to diverse daily activities of the potential exposed individuals), errors originating from the use of default assumptions for certain factors whose actual data are difficult to be collected from the population in the practical application, metabolic variation between individuals and accuracy in the conversion factors such as blood:hair conversion ratio of Hg in human bodies. The sources of variability and uncertainty in the two exposure estimation methods are discussed in the final section of this chapter.

6.1 Verification of modelled simulation of Hg exposure against survey data of Hg exposure

Table 44 compares the distribution of the simulated average daily intake of Hg using the Monte Carlo approach and the assumption that the proportion of MeHg was different in each medium class included in the simulation in Chapter 5 with the corresponding distribution of Hg average daily intake doses derived from Hg concentrations in the hair samples of the population in the study areas. The simulated ADDs ranged from 0.003 to 12.233 µg/kg body weight/day and had a similar distribution to the ADDs of Hg derived from the Hg levels in hair using the one-compartment model. The variability of the mean value of the simulated Hg intake doses (Equation 14 in Section 3.10) was approximately 23%.

Table 44 Comparison of the simulated cumulative distribution of Hg average daily intake doses using the Monte Carlo against Hg average daily intake doses derived from measured Hg concentrations in hair

		Hg intake dose (µg/kg body weight/day)	
		Corresponding to hair Hg (n=289)	**Monte Carlo (10,000 trials)**
Cumulative percentile (%)	**0**	0	0.003
	10	0.010	0.015
	20	0.010	0.020
	30	0.020	0.026
	40	0.020	0.032
	50	0.030	0.040
	60	0.040	0.051
	70	0.050	0.067
	80	0.080	0.095
	90	0.133	0.154
	95	0.253	0.239
	99.5	0.507	0.760
	100	0.550	12.233
Mean		0.061	0.075
Variability		0	22.64%

The Monte Carlo simulation produced a very high dose of daily Hg intake (12.233 µg/kg body weight/day) at the 100th percentile and was almost certainly the result of the simulation of an extreme event. At the 99.5th percentile the simulated intake dose was 0.76 µg/kg body weight/day. The maximum value was shown to have a probability of one in ten thousand. The extremely high value at the high end of the output distributions was caused from the calculation randomly picking several values at the upper-bound (i.e., above the 95th percentile) from the distributions of a number of exposure parameters (such as Hg concentration in soil and fish ingestion rate). As a result, an overly large and excessive value was generated at the tails in the probabilistic simulation. Such high Hg exposure levels were unlikely to occur to the study population. This study tended to ignore the maximum values in the simulation outputs, as the information at the tails of input distributions often is not as reliable as the central values, according to the USPEA (1997b). Figure 29 gives the simulated percentage distribution of the population that had different levels of Hg intake (***Simulated ADD***) and compares them with the values calculated from measured Hg concentrations in the hair of the population (***Observed ADD***). The probabilistic simulation underestimated the proportion of the population (37%) showing very low exposure levels (ADDs < 0.03 µg/kg body weight/day), compared with the ADDs calculated from Hg concentrations in hair (59%). The difference was made by the predicted exposed population being consistently and slightly higher in the simulation in the 0.03 to 0.24 µg/kg body weight/day range. At high levels (ADDs > 0.12 µg/kg body weight/day), the two approaches gave very comparable estimates.

When the RfD of 0.1 µg/kg body weight/day was adopted as the acceptable intake dose, approximately 19% of the population were indicated to have potential Hg exposure risks based on the Monte Carlo simulation which compared very favourably with the 17% determined from the hair samples. A value of 0.3 µg/kg body weight/day is recognized as a safety limit daily intake by the ATSDR and 0.47 µg/kg body weight/day by the Joint FAO/WHO Committee on Food Additives (JECFA). When these two safety levels were applied to the probabilistic simulation for the study population as a whole, there were respectively 3.22% and 1.31% of the population exceeding the levels. Compared with the data of average Hg intake doses derived from the study population's hair Hg (3.5% and 1.4%, respective), the differences in probability were less than 2%. The Monte Carlo simulation appeared to produce a stable and reliable prediction for the high-end exposed population.

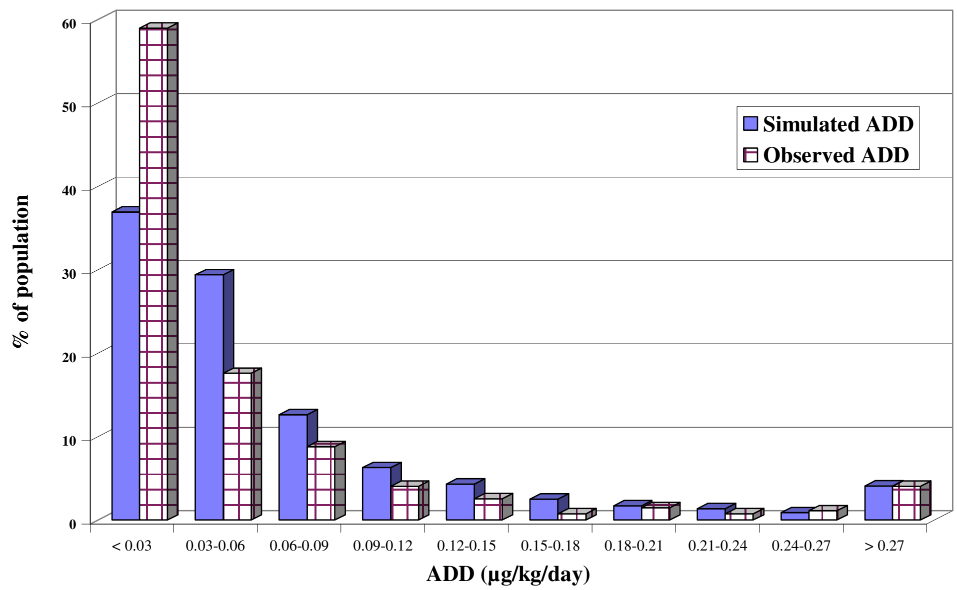
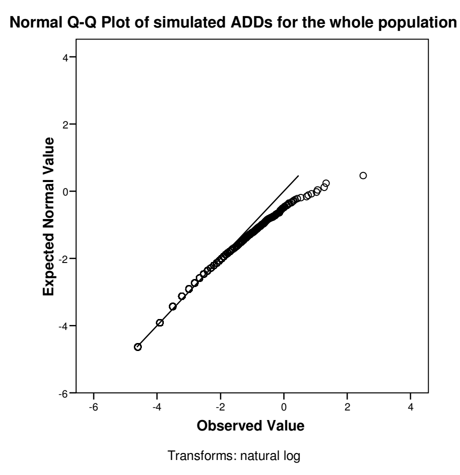
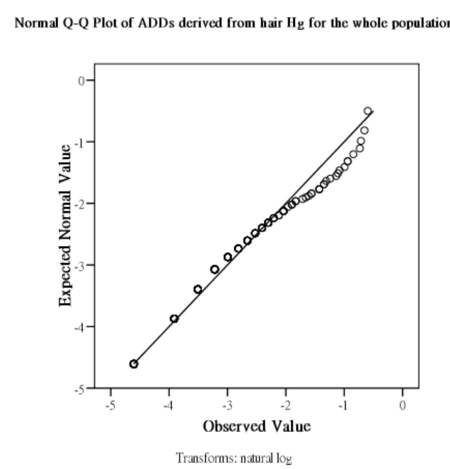


Figure 29 Frequency distributions of simulated ADDs and ADDs corresponding to measured Hg in hair of the whole population

Figure 30 show the visual inspection results using Q-Q plots for the simulated ADDs and the ADDs derived from Hg concentrations in the hair of the study population. After natural logarithm transformation, the two datasets tended to be normally distributed. However, the simulated upper-bound data were overestimated, as the observed values on the X-axis were larger than the expected normal values on the Y-axis.



(a) Simulated ADDs



(b) ADDs corresponding to measured Hg in hair

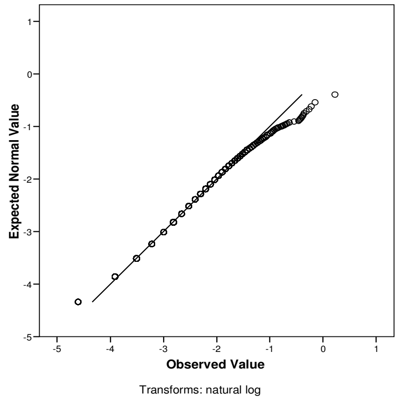
Figure 30 Transformed Q-Q of simulated ADDs and ADDs corresponding to measured Hg in hair of the whole population

The result of the simulation of Hg exposure showed a good fit result when the proportions of MeHg in the environmental media were taken into account. The result may imply that the assumption of the fractions of MeHg in total Hg in various exposure media was appropriate in the simulation for human exposure levels.

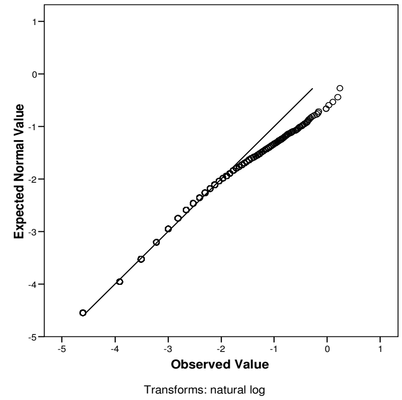
In addition to the probabilistic approach, the deterministic method was applied to evaluate the ADD of the study population. When the exposure data at the 95^{th} level were used as the input values, the single-point ADD was estimated to be 0.79 µg/kg body weight/day (not shown in Table 44). This value of the ADD was likely to be an overestimation because it exceeded the maximum intake dose back-calculated from the highest measured Hg concentration in hair. In comparison with the ADDs predicted from the Monte Carlo simulation, the single-value estimate was approximately three times greater than the probabilistic estimate at the 95^{th} percentile and even slightly greater than the probabilistic estimate at the 99.5^{th} percentile (0.96 µg/kg body weight/day). The variability of the single-value evaluation was as high as 1164%.

6.2 Verification of simulation methods using regional data

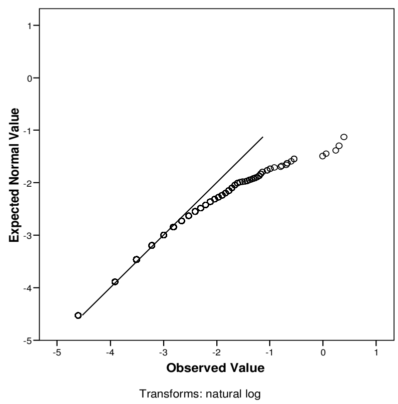
The Q-Q plots of the ADDs estimated from both the Monte Carlo simulation and stratified by the six locations of the study areas are shown in Figure 31. The simulated data appeared to be normally distributed after natural logarithm transformation. The values of the probabilities at the 0, 10^{th}, 20^{th}, 30^{th}, 40^{th}, 50^{th}, 60^{th}, 70^{th}, 80^{th}, 90^{th} and 95^{th} percentiles and their values are shown in Table 45. The simulations of Hg exposure intake doses were overestimated in the areas of Temirtau, Gagarinskoye and Samarkand and underestimated in Chkalovo and Rostovka. Of the six locations, the best prediction of the daily Hg intake was in Rostovka, where the mean of the simulated ADDs was nearly consistent with that corresponding to the hair Hg data.



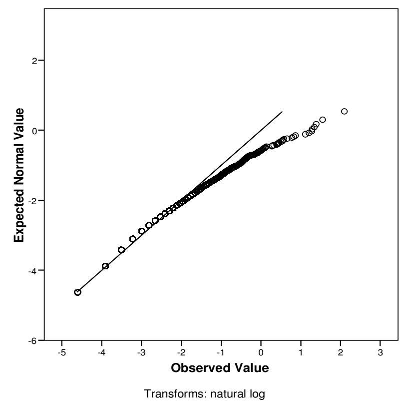
a) Temirtau old town



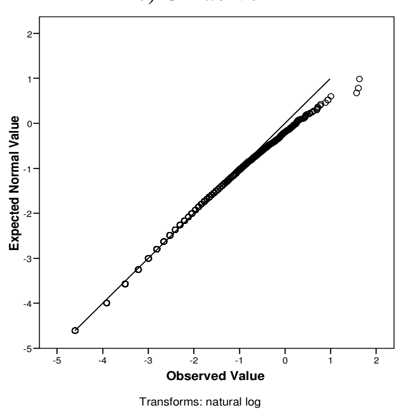
b) Temirtau new town



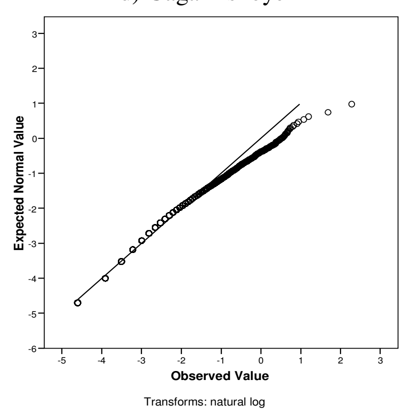
c) Chkaolvo



d) Gagarinskoye



e) Samarkand



f) Rostovka

Figure 31 Q-Q plots of simulated ADDs of the six residential areas using the Monte Carlo technique

The probabilistic simulation for the population in Chkalovo had lower exposure levels than the verification data, particularly at the upper 10^{th} percentile levels. The fact that this residential area is close to the heavily polluted sites may lead to increased Hg contamination in this area. The probabilistic simulations could have underestimated the proportions of MeHg in total Hg in the various environmental media, which would result in an underestimation in the probabilistic simulations. Hence, whether there are higher methylation rates than the study's assumptions in the surroundings of Chkalovo should be further investigated.

Table 45 Comparison for the six residential areas between the probability distributions of ADDs for 12 different percentiles of simulated and observed data

Percentile (%)	Observed ADDs corresponding to actual hair Hg (µg/kg body weight/day)					
	Temirtau old town (n=50)	Temirtau new town (n=52)	Chkalovo (n=45)	Gagarinskoye (n=67)	Samarkand (n=43)	Rostovka (n=32)
0	0.000	0.010	0.010	0.000	0.000	0.020
10	0.010	0.020	0.010	0.000	0.010	0.030
20	0.018	0.020	0.010	0.010	0.010	0.040
30	0.020	0.020	0.020	0.010	0.020	0.040
40	0.026	0.024	0.020	0.010	0.030	0.050
50	0.030	0.030	0.030	0.020	0.040	0.085
60	0.034	0.030	0.030	0.026	0.050	0.122
70	0.040	0.040	0.040	0.032	0.080	0.144
80	0.060	0.060	0.060	0.058	0.100	0.240
90	0.091	0.080	0.110	0.094	0.252	0.315
95	0.111	0.119	0.206	0.144	0.387	0.354
100	0.290	0.520	0.480	0.430	0.490	0.550
Mean	0.043	0.049	0.054	0.041	0.085	0.133
Percentile (%)	**Simulated ADDs using Monte Carlo (µg/kg body weight/day) / 10,000 trials**					
	Temirtau old town	**Temirtau new town**	**Chkalovo**	**Gagarinskoye**	**Samarkand**	**Rostovka**
0	0.005	0.005	0.003	0.003	0.007	0.004
10	0.030	0.022	0.010	0.012	0.038	0.022
20	0.039	0.029	0.013	0.017	0.052	0.030
30	0.047	0.035	0.015	0.022	0.066	0.039
40	0.056	0.041	0.018	0.028	0.082	0.048
50	0.065	0.048	0.021	0.035	0.101	0.060
60	0.075	0.057	0.025	0.045	0.126	0.075
70	0.087	0.068	0.030	0.060	0.158	0.100
80	0.106	0.087	0.039	0.088	0.212	0.144
90	0.137	0.124	0.055	0.151	0.321	0.243
95	0.173	0.174	0.077	0.234	0.443	0.382
100	1.246	1.273	1.481	8.154	5.114	9.774
Mean	0.078	0.067	0.030	0.073	0.156	0.115
Variability	81.18%	37.52%	-44.11%	78.14%	82.97%	-13.31%

In spite of discrepancies between the ADDs simulated by the Monte Carlo approach and the observed ADDs calculated from Hg exposure concentrations in hair, the variability of the mean values in the probabilistic simulations for the six residences was less than 100%, indicating that the outputs of the Monte Carlo simulations were not excessively overestimated. Even at the upper-percentile levels (e.g. the 95th percentile), the estimated predictability was still acceptable.

The use of the single-value method gave extremely high estimates of ADDs, compared with the Hg intake doses evaluated from the Hg concentrations in hair

(Table 46 and Figure 32). In five of the six residential areas, the single point simulations exceeded the Hg intake doses calculated from the upper 5% levels of the Hg concentrations in the hair samples, whereas the ADD single-value simulation in Chkalovo had the low and negative variability of -33%. Not surprisingly, the levels of the variability in the single-value simulations were higher than the mean values and the 95th percentile estimates of the probabilistic simulations.

Table 46 Verification of simulated single-value ADDs using the deterministic method for different residential areas

Area	Item	Observed ADDs	Simulated single-value ADDs
Temirtau old town	**95th-tile (µg/kg body weight/day)**	0.111	0.365
	Variability (%)		229
Temirtau new town	**95th-tile (µg/kg body weight/day)**	0.119	0.582
	Variability (%)		389
Chkalovo	**95th-tile (µg/kg body weight/day)**	0.206	0.138
	Variability (%)		-33
Gagarinskoye	**95th-tile (µg/kg body weight/day)**	0.144	0.475
	Variability (%)		230
Samarkand	**95th-tile (µg/kg body weight/day)**	0.387	1.480
	Variability (%)		282
Rostovka	**95th-tile (µg/kg body weight/day)**	0.334	0.886
	Variability (%)		151

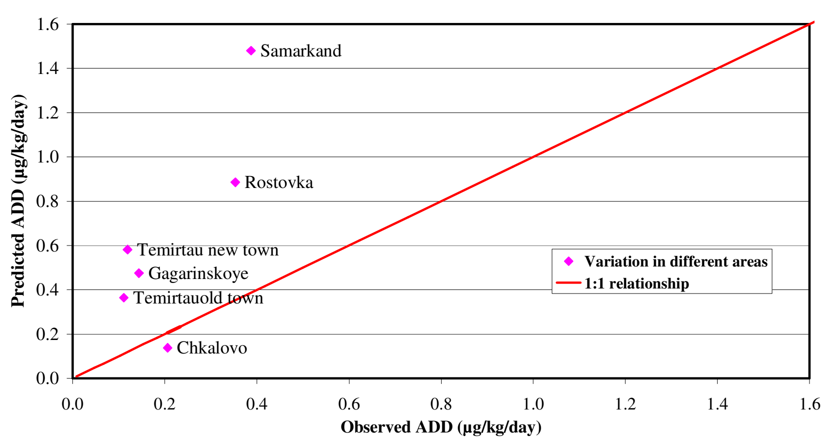


Figure 32 Relationship between estimates of ADDs using the deterministic method for six residential areas against the observed highest values

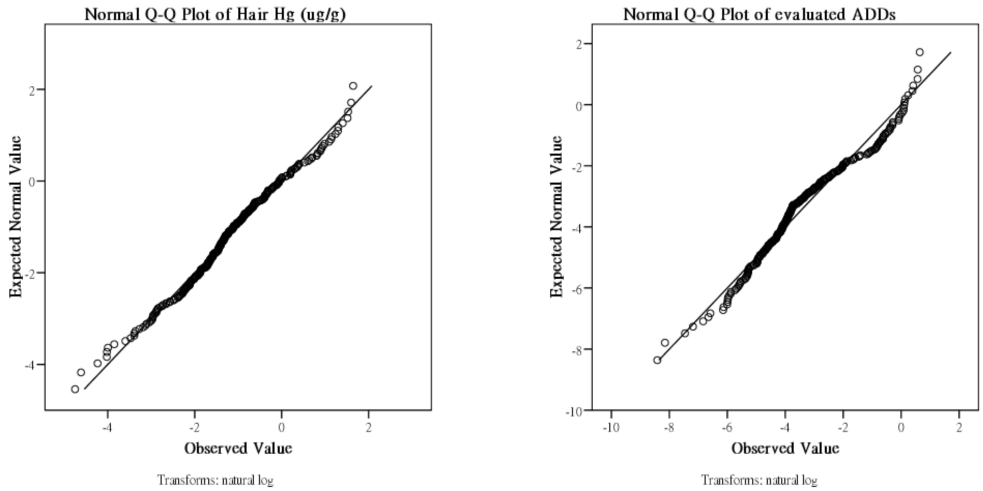
6.3 Verification of dose-response relationship using questionnaire data and paired hair Hg concentrations

The average daily Hg intake doses of the study population established from dietary questionnaire data ranged from 0 to 1.150 μg/kg body weight/day, with a mean of 0.107 μg/kg body weight/day (Table 47). However, the Hg intake dose of 0.061 μg/kg body weight/day for a 70 kg-adult was estimated from a measured mean Hg concentration in hair of 0.572 μg/g. This was approximately half that of the estimated value. In other words, the established dose-response relationship based on the questionnaire data was poor. Furthermore, the ADDs evaluated from the questionnaire data showed that approximately 21% of the population were potentially likely to have adverse health effects due to Hg exposure, as their average daily Hg intake doses were higher than the RfD of 0.1 μg/kg body weight/day. Compared with 17% of the population at risk determined based on observed Hg levels in hair, the exposure estimates based on questionnaire data were more conservative.

Figure 33 show the visual inspection results using Q-Q plots for the Hg concentrations in the hair of the study population and the evaluated ADDs based on questionnaire data. The raw data were not formed naturally distributed, but after nature logarithm transformation, the two datasets tended to be normally distributed. Hence, the data were logarithm-transformed to test the correlationship and relationship between the evaluated ADDs based on questionnaire data and the Hg concentrations in the hair of the residents at the study site.

Table 47 Distribution of average daily Hg intake doses established from questionnaires of the study population (n= 203)

		ADDs from questionnaires (μg/kg body weight/day)
Mean		0.107
SD		0.216
Min.		0
Max.		1.150
	25	0.010
	50	0.022
Percentiles (%)	**75**	0.074
	90	0.361
	95	0.604
Variability		75.41%



(a) Hg concentrations in the hair (b) Evaluated ADDs based on questionnaires

Figure 33 Q-Q plots of the Hg concentrations in the hair of the study population (µg/g) and the evaluated ADDs (µg/kg body weight/day) based on questionnaire data.

A correlation analysis had a weak ($r = 0.444$) yet statistically significant correlation ($p. < 0.001$) between the amount of Hg in hair and estimated Hg intake through food consumption. This model implies that, after logarithm transforming, on average, every unit of daily Hg intake dose is associated with an increase of 0.444 µg/g of Hg in hair. Total individual daily doses were calculated using the one-compartment model given in Section 3.10. The logarithm transformed data indicated that daily Hg intake dose was plotted against Hg concentrations in paired participant's hair to give a linear regression model. This was a significant relationship between these parameters (0.197, $p. < 0.001$). It indicates that approximately 20% of the variation of Hg concentrations in hair was considered to be related to Hg intake via food consumption.

6.4 Discussion

Overestimation of Hg exposure simulations using probabilistic and deterministic methods

The probabilistic simulations using the Monte Carlo gave conservative values for the amount of Hg that the study population were exposed to, generally giving higher exposure levels of Hg for the study population in comparison with the corresponding actual Hg concentrations in the hair samples. However, the slightly conservative simulations were thought to be an effective risk estimate for human exposure to slightly high levels of Hg at the study site. The advantage of probabilistic simulation is that risk managers can identify the risk of the adverse health effects before on-site Hg levels become critical.

The discrepancy between the probabilistic simulation and the validation data of the population as a whole was approximately 27% and the largest discrepancy of 83% was in Samarkand village. The simulation results are better than that found in other studies. For example, Gosselin et al. (2006) who predicted average Hg intake doses that were seven-fold higher than that reconstructured from the measured MeHg concentrations in the hair of the same population. Canuel et al. (2006) and Loranger et al. (2002) predicted Hg levels that were at least 1.5-fold higher than the actual Hg exposure concentrations in hair. These previous verification studies all used the deterministic method to simulate human exposure to Hg. Therefore, the overestimated results in those studies may imply that a different methodology of the Monte Carlo approach produces a more accurate Hg exposure assessment.

In this study the Monte Carlo simulation using a wide range of exposure data to develop probabilistic outputs provided a fairly valid estimate of human exposure to Hg. The finding that the probabilistic approach to assessing exposure risk was more accurate than the single point approach was also in accordance with the finding of Boon et al. (2003) who assessed the dietary exposure to pesticide based on questionnaire-based data and obtained a more accurate probabilistic simulation result than using the single-point method.

The single-point method overestimated Hg intake doses of the study population, being higher than the hair Hg exposure levels at the 95th percentile. This approach gave a particularly high estimate of Hg absorption for the population of Samarkand. In this village, there were significantly more fishermen who self-estimated both frequent fish consumption and large portion sizes. Some of the estimates of food portion sizes appeared particularly large (*it may be the result of fishermen overestimating the weight of fish they caught*). Those overestimated self-reported data could explain the discrepancy between the single-point Hg intake estimates and the

Hg intake doses derived from the Hg concentrations in hair.

Low level of Hg exposure of the study population

Hg exposure doses established from concentrations of Hg in hair were lower than the modelled Hg intake. In fact, the Hg exposure levels in the local people's hair were lower than expected, since the River Nura has been polluted for more than 50 years and has been shown to be highly contaminated (Heaven et al., 2000b, Heaven et al., 2000a). The low level of Hg exposure may be due to the fact that Hg has been shown to be bound tightly to the fly ash. Several studies (Gupta and Ali, 2004, Rio and Delebarre, 2003, Sen and De, 1987) have concluded that fly ash, the by-product of power stations, could be an effective adsorbent to remove Hg in wastewater and alleviate the mobility of Hg in silts and soils. Rio and Delebarre (2003) reported that the powerful Hg adsorption ability was seen in sulfo-calcic fly ash. The equilibrium of adsorption reaction of the studied fly ash reached a stable condition after 72 hours with the maximum adsorption capacity of 4.9 mg/g. The millions of tonnes of fly ash at the study site had a high affinity for Hg dumped in the river above the wastewater discharge point (Stratienko, 2004, Yanin, 1997). It is likely that the bioavailability of Hg has been reduced as a result.

Uncertainty and variability inherent in Hg exposure estimates

There were sources of uncertainty and potential variability originating in the mathematical exposure assessments in the verification study. A measure of this uncertainty and variability is desirable if the validity of the Hg exposure simulation and verification studies are to be accepted.

One of the sources of uncertainty is from laboratory analysis. The quality of laboratory analysis is likely to contribute to the uncertainty and variability in both hair Hg concentrations and exposure dose estimation. Barr et al. (2006) indicated that the uncertainty inherent in chemical analyses potentially occurred in several fields including the initial extraction from a solid matrix (e.g., dust, wipes, soil), cleanup of the sample extract, analytical detection methodologies and techniques, method performance, quality assurance and quality control in a laboratory and even from the beginning of sampling. Hill and von Holst (2001) considered that the undesirable uncertainties generating from an analytical method may contribute to random errors, systematic errors and spurious errors (mistakes). However, those errors were nearly impossible to remove, as analyst performance, equipment, reagents, test conditions and so on were unlikely to be completely consistent during a period of time or between different laboratories (Hill and von Holst, 2001). In the present study, laboratory analytical uncertainty may be inherent in Hg quantification in samples of hair, foodstuffs and various environmental media. Nevertheless, regular interlaboratory exchange of samples and the use of standard reference material were both conducted by the National Institute for Minamata Disease (National Institute for Minamata Disease (NIMD), 2006) and the chemical laboratories in Almaty (Tanton et al., 1999, Ullrich et al., 2007b) to control analytical errors. An average recovery of 98.8+/-0.65% was reported by Ullrich et al. (2007b). As a result, the analytical variability or laboratory imprecision should be insignificant in this study.

To assess the extent and likelihood of actual Hg exposure of a population, the mathematical exposure models require input values including the characterisations of the exposure chemical, contamination levels of the chemical in the potential pathways, the duration/periods of exposure activities conducted by the exposed subjects, the fractions of Hg forms and their bioavailability and so on. When complete information on exposure is lacking and will not be available in the foreseeable future, surrogates or statistical data are used. This study adopted the proxy values for a number of input parameters such as daily ingestion rates of soil or water and duration of exposure activities that may be attributed to uncertainties. Although these data were collected from the recommendations of the USEPA (1989, 1997a) and other related literature (see Table 13 in Section 3.9), inconsistency inevitably existed between the surrogates or the assumption and the real situations. For example, the methylation rates in the on-site soil and water may be variable spatially or with time series instead of steady-state. Such steady-state assumptions may result in the over- or underestimated outputs of the exposure simulations.

The food frequency questionnaires were designed to collect the varied dietary behaviour among individuals. The non-measured replies in response to average food portion size or food intake frequency provided by the interviewees were likely to account for much of the uncertainties in the estimates of Hg intake doses. As mentioned previously, statistically large daily meal portions were observed in a significant number of the questionnaires. Although the cases of overly large food weights beyond the upper 5th percentile of the data distributions were ignored, the wide range of dietary intake data still likely to cause an overestimation of ADDs with the single-value method (see Table 46 and Figure 32 in Section 6.2). The questionnaire-based data have shown the influence on the validity of the exposure simulations using the Monte Carlo simulation. The sensitivity analyses indicated that the major proportion of the variation in the exposure simulations were the variables associated with pattern of river fish consumption Figure 27 in Section 5.2). In addition, the interviewees' poor estimates of fish consumption are likely to have resulted in an overestimation of Hg exposure levels in the dose-response relationship. Therefore, there was only a relatively weak correlation between Hg levels in the hair samples of the individuals and their average Hg intake doses (r = 0.444, in Section 6.3).

Uncertainties inherent in survey data have commonly been reported in previous studies. Self-reported quantity of fish intake rates were collected from nearly 3000 children and women in the United States and found that they overestimated two- and three-day daily averages of consumption, compared with food records and 24-hour recalls (Tran et al., 2004). Yokoo et al. (2001) also advised that the calibration equation for the surveyed fish weights may necessarily be stratified by sex and adjusted by age and season variables. Recall difficulty also caused invalid estimates of human exposure to Hg in other studies (Loranger et al., 2002, Canuel et al., 2006, Gosselin et al., 2006). To conduct the verification of the modelled hair Hg levels, Canuel et al. (2006) collected data on fish intake patterns and Hg concentrations in hair from Innuit communities in three locations. The simulated hair Hg levels of the study population were demonstrated to be 1.5- to 14-times higher than the measured concentrations. Gosselin et al. (2006) compared the MeHg intake doses established from the measured hair Hg concentrations in the indigenous populations in Canada to the ones evaluated from food consumption questionnaires for the same population. The average of the reconstructed MeHg intake doses was seven times lower than the questionnaire-based evaluation, due to incorrect data of fish and shellfish consumption reported by the study population. Furthermore, Loranger et al. (2002) concluded in their study that their modelled MeHg concentrations in hair were six-fold higher than the real MeHg concentrations in hair. The higher MeHg levels in hair being simulated were the result of unrealistic estimates of fish and the fish intake rates provided by 94 recreational anglers. Clearly people consistently tend to overestimate meal portion size and frequency.

Another type of uncertainty related to survey data in this study was sample size. Over thirty samples of soil, water and fish were collected from the study areas, but the sample numbers of other foodstuffs including beef, dairy product, tomatoes and cucumbers were limited. Those small number of samples potentially caused the uncertainties in Hg contamination levels in the environmental media and Hg exposure estimates, as the quality of the data in the exposure assessments was associated with the number of collected samples (Nieuwenhuijsen et al., 2006). Therefore, the unrepresentative numbers of food samples may be attributed as giving rise to the uncertainties in the estimation of human Hg exposure.

Uncertainty and variability generating in the use of Hg levels in hair

Although blood and hair samples are thought to represent body burdens of a chemical, the exposure data measured via biomarkers tend to provide the human exposure at a single point in time (Paustenbach and Galbraith, 2006, Bartell et al., 2004). For example, the Hg concentration measured in the scalp hair of 1 cm closest to the root only represents the Hg exposure of the past month. The temporal impact on the studies associated with human exposure to Hg may be significant because Hg can be readily detected in human blood after consuming Hg-contaminated food. Therefore, Valberg et al. (1996) indicated that the timing of biomarker collection for the intention of exposure measurement was important during the process of health assessment. Gosselin et al. (2006) further suggested that, in the sampling procedure, the study population should be asked to avoid any MeHg contaminated food for at least two days prior to the blood test.

Seasonality of behaviour and food consumption can result in distinct seasonal body burdens of Hg. The factor of time may affect the accuracy of Hg exposure levels measured via the hair samples of the study population. Hair collection was carried out in early summer, when the melting waters of the River Nura and the lakes were used as the recreation places for fishing and swimming by the local people. The elevated exposure to Hg at this time may be the result of increased fish consumption or outdoor activities which resulted in higher Hg concentrations in hair than other periods of the year, for example, in winter. On the other hand, in winter, potatoes are the local staple food and their irrigation water source is very high in Hg. Hence, such seasonal foodstuffs may vary Hg loads on the population. A seasonal trend on Hg exposure levels due to temporal complexity of dietary behaviour was observed and reported by a review associated with the biomarker ratios of blood:hair, blood:intake and hair:intake in maternal bodies (Bartell et al., 2000). Therefore, there were uncertainties in this study as hair samples were collected only once from the potentially exposed individuals. In this case, the hair Hg data should be thought of as the levels of Hg exposure during the past month rather than reflecting the average exposure doses throughout the exposure period.

This study suggests that there were potentially wide variability in seasonal dietary behaviour in the study areas. It would clearly be advantageous to sample two to four times a year but the cost would be considerable.

Uncertainty and variability may occur in the estimation of Hg intake doses that were restructured from Hg concentrations in hair, as published data were used as the inputs in the one-compartment model. The frequently used ratio of Hg content in hair (μg/g) to that in blood (μg/mL) of 250 was adopted in the pharmacokinetic model in this study to back-calculate the average individual Hg intake dose from the Hg concentrations measured in their hair. This was based on the strong corelation between the amount of Hg in human blood and hair and the dose-response relationship of the Hg exposure (WHO, 1990, NRC, 2000). Although steady state was assumed, there would be some variability existing in the biological parameters, particularly as the range of the ratio has been found to be from 140 to 370, which could cause a variation of -45% to 48% (WHO, 1990). In addition, there was the consideration of the fact that hair Hg levels may vary in different segments of a strand of hair or be differed by pregnancy, making the conversion ratio of Hg between the biomarkers hair and blood variable (Lipfert, 1997, Stern and Smith, 2003).

A mean value of 50 days reported by Sherlock et al. (1984) was adopted by the USEPA (2001a) to determine the elimination constant of 0.014 for estimation of MeHg from the blood and was used in this study. Although the half life of 50-day has been assumed in recent studies in human exposure models (Stern, 1997, Swartout and Rice, 2000), the variable of half lives of Hg in human blood has been found to range from 32 to 189 days (Al-Shahristani and Shihab, 1974, Aberg et al., 1969). As a result of such a wide range of data, the variable could account for 30-40% of the uncertainty in the risk assessment of the harmful health effects and the Hg intake estimates (Lipfert, 1997).

The variables relating to personal kinetic conditions in the one-compartment model were assumed to be constant in this study, but inhomogeneity among the population potentially contributed to the variability in the Hg exposure estimates derived from Hg concentrations in hair. In some studies (Ponce et al., 1998, Sanga et al., 2001), those unknown heterogeneity within the population were demonstrated to significantly result in either bias or variance in Hg exposure estimates established from biomarker data. Therefore, the study suggests that Hg concentrations in hair would be a better indicator for average Hg intake rates of individuals, with the available data distributions of personal physio-kinetic parameters including the ratio of total Hg concentration in hair to that in blood, half life and bioavailability of Hg in the blood and hair growth speed. Nevertheless, the data collection would require a long period of time and involve high research costs.

6.5 Summary

The probabilistic estimates of Hg exposure gave a better and more reliable estimate of population risk than the deterministic evaluation due to probability outputs for the risk assessment of human exposure to Hg were slightly conservative. Nevertheless, when the simulations were verified for the six residential areas, discrepancies were observed, possibly resulting from survey errors. The survey errors, particularly unconfirmed dietary behaviour data collected via the food frequency questionnaires, were likely to result in the overestimated Hg exposure levels for most of the locations. Incorrect survey data may also lead to a weak relationship between the estimated daily Hg intake doses of individuals and their Hg concentrations in hair.

The validity of the estimate of risk is valid for early summer season as seasonal dietary patterns could change the level of fish in other seasons. While the verified Hg intake doses were derived from the observed Hg concentrations in hair using the one-compartment model, interindividual variation was likely to account for variability. Since varied excretion and absorption rates of Hg in human bodies have been reported by several preliminary studies, the data of individual biological conditions could be collected for resulting actual Hg exposure levels of the individual.

7. EXPOSURE MODEL FOR ESTIMATING MERCURY LEVELS IN HAIR

Hg concentrations in people's scalp hair have been commonly used for the indicators of human exposure. In addition to the benefits of easy collection, transport, and storage, hair samples have been considered to reflect historical records of environmental exposure through quite accurate measurement techniques. The finding that Hg levels in hair are highly associated with those in blood supports scalp hair as the strong biomarker. For the above reasons, Hg concentrations in the residents' hair samples were determined to provide the actual levels of Hg exposure of the study population.

As discussed in Section 2.2.5, the relationship between frequent fish and shellfish consumption and elevated Hg concentrations in hair has been widely studied as a result of significant Hg bioaccumulation in the aquatic food chains and human bodies. In addition to fish and shellfish consumption, the probabilities of Hg exposure levels of individuals have been conducted to be varied between different genders, ages and residential locations (Barbosa et al., 2001, Batista et al., 1996, De Oliveira et al., 2004, De Oliveira Santos et al., 2000, Foo et al., 1988, Holsbeek et al., 1996, Lee et al., 2000, Santos et al., 2002). Geographic and demographic characteristics contributed the effects in a dose-response relationship. In this chapter, Hg exposure levels of the study population were determined by Hg concentrations in their hair and examined the effects of the variables including genders, ages, occupations, fish eaters/non-fish eaters, residential areas and chemical treatment on hair. A model was eventually established to describe the relationship between the involved variables and Hg concentrations in hair. This model was expected to define the group potentially sensitive to Hg exposure.

7.1 Relationships between demographic factors and Hg concentrations in hair

Table 48 shows that the average Hg concentrations in hair samples from the town and the villages varied from 0.381 μg/g in Gagarinskoye to 1.244 μg/g in Rostovka and statistically significant differences were found in the Hg concentrations in the hair between the populations in the residential areas. The Hg levels in Rostovka and Samarkand were higher than other residential areas, with the Hg concentrations in the hair samples in the local people living in Rostovka being significantly higher than those in Temirtau town, Chkolova and Gagarinskoye. As discussed in Chapter 5, many residents in Rostovka and Samarkand tended to consume self-caught fish from the river or the neighbouring oxbow lakes. As fish and shellfish consumption is known to be the main pathway for Hg accumulation in men (WHO, 1990), it is likely to provide a plausible explanation for the difference.

Table 48 Hg concentrations (μg/g) in hair samples from residential areas

Hg concentrations (μg/g)	**n**	**Mean**	**SD**	**Min.**	**Max.**
Temirtau old town	50	0.396	0.440	0.021	2.690
Temirtau new town	52	0.458	0.694	0.052	4.947
Chkalovo	45	0.512	0.789	0.057	4.558
Gagarinskoye	67	0.388	0.622	0.009	4.083
Samarkand	42	0.806	1.100	0.014	4.620
Rostovka	32	**1.244***	1.205	0.165	5.184
Mean	288	0.577	0.841	0.009	5.184

*: higher Hg level in Rostovka with significant level < 0.05 than in other areas except Samarkand

There were no significant differences in Hg concentrations in hair when the population was stratified by age (less than age 16, age 16-30, age 31-45, age 45 above). However, when the data were split into below and above 45 years old, the population over 45 years old had significantly higher levels of Hg in hair than those under 45 years old in all areas but Samarkand and Rostovka villages (Table 49), with the older population as a whole having statistically higher Hg concentrations in their hair than younger people.

Males had approximately twice as much Hg in their hair than females at the study site (Table 50). Men ate significantly more fish and shellfish than women (75g/day and 40g/day, respectively). Hence, the consumption of fish and shellfish intake appeared to play an important role on the difference of Hg concentrations in hair between the two subgroups. However, there were also other factors that would have contributed to this finding. In pregnant women, a significant amount of Hg burdens in their bodies could have been taken up by fetuses during pregnancy, as observed by Barbosa et al. (1998b) and Barbosa and Dorea (1998a). Unfortunately, there were only eight pregnant women in this survey. Hence, it was impossible to identify the significance of the potentially lower Hg concentrations in women's hair during pregnancy.

Table 49 Relationship between Hg concentration in hair between age classes and locations

Areas	Hair Hg (µg/g) (n.)	
	Under 45 years old	**Over 45 years old**
Temirtau old town	0.288 (30)	0.557 (20) **
Temirtau new town	0.319 (31)	0.714 (19) *
Chkolova	0.267 (25)	0.833 (19) **
Gagarinskoye	0.263 (46)	0.617 (23) **
Samarkand	0.698 (32)	1.156 (10)
Rostovka	1.360 (18)	1.096 (14)
Mean	0.462 (182)	0.777 (105) **

*: significant level < 0.05 **: significant level < 0.01

Table 50 Relationships between a number of variables and the Hg concentrations in hair samples

Factor	n	Mean of Hg in hair (µg/g)
Gender		
Male	113	0.825**
Female	174	0.416
Had chemical treatment on hair in the past month		
No	205	0.660**
Yes	78	0.371
Occupation		
Never worked in the acetaldehyde factory	259	0.582
Worked/working in the acetaldehyde factory	23	0.570
Fishermen		
No	262	0.501
Yes	25	1.376**
Fish eating in the past year		
No	20	0.101
Yes	264	0.610**
Ethic origin		
Russian	179	0.548
Kazakh	38	0.362
Tartar	21	0.589
German	26	0.914
Ukrainian	11	0.763
Greek	6	1.179
Others	3	0.247
Drinking water		
Central water supply	198	0.623
Well	32	0.240
Borehole	48	0.531
Others	6	0.889

**: significant level < 0.01

People who chemically treated their hair had significantly less Hg in their hair than those who did not treat their hair, the levels being almost twice as high. However, the difference between genders was confounded with hair treatment as people who did not dye their hair were mainly men. The distribution of these subgroups indicated that 97.5% of the people with hair colouring treatment were female. When only female participants were tested, Hg concentrations in hair between women with and without chemical hair treatment were 0.451±0.588 and 0.375±0.566 μg/g, respectively, but the differences were not significant (*p.* = 0.276, Table 51 and Figure 34). Therefore, the covariate effect as a result of gender is attributable to the statistical difference on Hg concentrations in hair between hair colouring treatment and normal hair subpopulations.

Table 51 Hg concentrations in hair between gender and chemical treatment on hair

Hair Hg (μg/g)	Chemical treatment on hair in the past month		
	No	Yes	Total
Male	0.846	0.303	0.825
Female	0.451	0.375	0.416

People who responded in this survey that they were fishermen and consumed a lot of fish had significantly more Hg in their hair than those who had ate less fish and shellfish (Table 52). As there were only twenty-five fishermen (out of 287) in this survey, a poor reliability in the statistical analysis may be caused by the bias. Therefore, the difference was further investigated between the Hg levels in the residents of Samarkand village by separating the population into those with fish related occupations/leisure activities and those with other occupations. The residents of this village were selected as approximately 50% of the fishermen interviewed were from Samarkand and Hg exposure via the environmental media could be neglected due to them all living in the same residential area. The result confirmed that the hair Hg level in the non-fisherman subgroup was significantly lower than in fishermen.

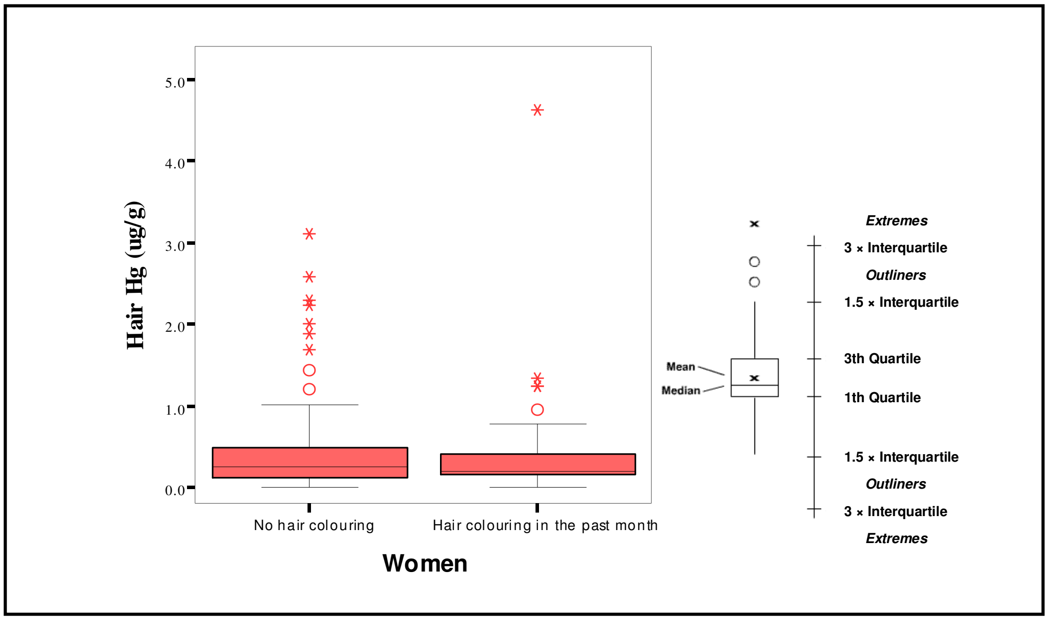


Figure 34 Comparison of Hg concentrations in women's hair between those chemically treating their hair in the past month and whose who did not

The Hg levels in the hair of the non-fish-eaters of the population as a whole ranged from 0.009 to 0.358 µg/g and were approximately six times lower than those fish eaters. In Sweden, Lindberg et al. (2004) also reported the comparable Hg concentrations in the hair of non-fish-eaters (range: 0.04-0.32 µg/g).

Table 52 Difference in Hg concentrations in the hair of fishermen and non-fishermen in the village of Samarkand

Hair Hg (µg/g)	n	Mean	SD	Min.	Max.
Non-fishermen	29	0.440	0.683	0.014	3.676
Fishermen	13	1.623**	1.415	0.221	4.620
Mean	42	0.806	1.100	0.014	4.520

**: significant level < 0.01

7.2 Relationship between Hg concentrations in hair and dietary intake patterns

Table 53 shows the correlations between Hg concentrations in the hair of the study population and their dietary intake frequencies of different food types. Frequencies of consuming river fish species including bream or roach, carp and perch had positive correlations with Hg levels in hair, indicating that consuming these fish species caught from the River Nura elevated Hg exposure levels in hair. People consuming dairy product frequently tended to have low levels of Hg in hair, as a negative correlation is seen between Hg concentrations in hair and intake frequencies of dairy product.

Other correlations showed that people consuming river pike/zander frequently tended to have other fish species from the river, like bream or roach, carp and perch. The correlation coefficient was high between intake frequency of river pike/zander and intake frequency of river carp and intake frequency of river pike/zander and intake frequency of river perch, 0.559 and 0.623, respectively. Similarly, positive correlation coefficient are seen between intake frequencies of market pike or zander, bream or roach, carp and crayfish. It indicates that dietary intake behaviour was consistent between consumers who chose to eat market fish, regardless of fish species. High correlation coefficients were also found between intake frequency of river crayfish and intake frequency of market shellfish ($r = 0.945$) and intake frequency of tomatoes and intake frequency of cucumbers ($r = 0.799$). However, there were few people responding to consume shellfish in the past year. The high correlation coefficient might be a bias.

Table 53 Correlation coefficients between Hg concentrations in hair and food intake frequencies

	Hair Hg	River pike/zander	Market pike/zander	River bream/roach	Market bream/roach	River carp	Market carp	River crayfish	Market crayfish	Market shellfish	Market herring	River perch	Market perch	Other river fish	Other market fish
Hair Hg	1	0.040	-0.095	**0.147***	-0.026	**0.307*****	-0.075	-0.045	-0.025	0.010	0.030	**0.237*****	-0.030	0.054	-0.059
River pike/zander		1	0.110	**0.192****	**0.224****	**0.191****	**0.222*****	-0.007	-0.066	-0.026	-0.008	**0.623*****	**0.144***	0.080	-0.037
Market pike/zander			1	-0.063	**0.236****	-0.115	**0.369*****	-0.017	**0.296*****	0.002	0.088	-0.050	**0.226****	**-0.029*****	0.002
River bream/roach				1	0.045	**0.559*****	-0.023	-0.028	-0.065	-0.035	**-0.140***	**0.178***	-0.037	0.320	-0.070
Market bream/roach					1	-0.007	**0.360*****	-0.036	**0.392*****	-0.032	**0.194****	**0.195****	0.016	**-0.001****	-0.038
River carp						1	-0.059	-0.031	-0.076	-0.038	0.016	**0.185****	-0.060	0.215	-0.070
Market carp							1	-0.045	**0.319*****	-0.003	0.021	**0.249****	0.100	-0.057	-0.007
River crayfish								1	-0.026	**0.945*****	-0.005	-0.015	-0.018	-0.014	**0.448*****
Market crayfish									1	0.025	0.059	-0.035	-0.041	-0.032	-0.049
Market shellfish										1	0.035	-0.017	-0.016	-0.013	**0.431*****
Market herring											1	-0.076	0.126	-0.067	-0.029
River perch												1	-0.029	-0.022	-0.034
Market perch													1	-0.022	-0.033
Other river fish														1	-0.026
Other market fish															1

	Hair Hg	Beef	Dairy	Tomatoes	Cucumbers
Hair Hg	1	-0.096	**-0.186****	-0.068	-0.003
Beef		1	0.121	0.097	0.060
Dairy			1	0.053	0.099
Tomatoes				1	**0.799*****
Cucumbers					1

*: significant level < 0.05 **: significant level < 0.01 ***: significant level < 0.001

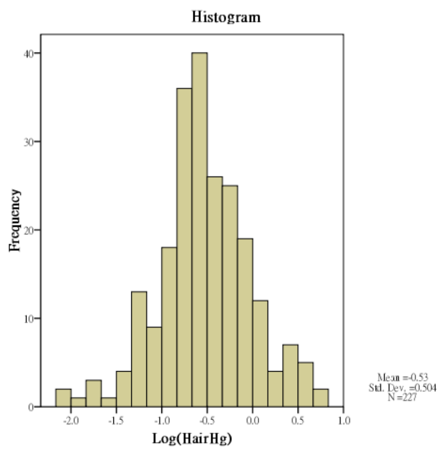
7.3 Development of an exposure model from observed variables

A regression model of Hg exposure in hair was described using calculated daily intake doses of Hg via food consumption as mentioned in Section 6.3 together with demographic factors including gender, residential area, fishery occupation and age. When the normality of the distribution of Hg concentrations in hair and the daily intake of Hg via food of the study population were studied, the variables were not normally distributed. Hence, logarithm transformation was conducted. The results of log-transformed Hg concentrations in hair and daily intake doses of Hg that are shown in Table 54 and Figure 35, confirming the assumption of being normally distributed, as the skewness and kurtosis were near zero and the histograms and the Q-Q plots were visually normal.

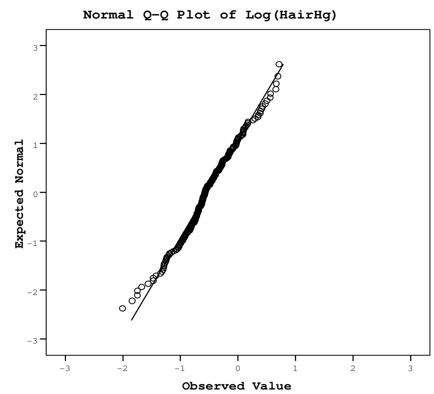
Table 54 Results of normality tests for log-transformed Hg concentrations in hair and daily intake doses of Hg

		Log(Hair Hg)		Log(ADDs)	
		Statistic	**Std. Error**	**Statistic**	**Std. Error**
Mean		-0.531	0.033	-1.532	0.049
95% Confidence	**Lower Bound**	-0.597	-	-1.629	-
Interval for Mean	**Upper Bound**	-0.466	-	-1.435	-
Median		-0.570	-	-1.637	-
Std. Deviation		0.504	-	0.720	-
Minimum		-2.060	-	-3.630	-
Maximum		0.710	-	0.060	-
Range		2.780	-	3.690	-
Skewness		-0.074	0.162	0.157	0.166
Kurtosis		0.495	0.322	-0.027	0.330

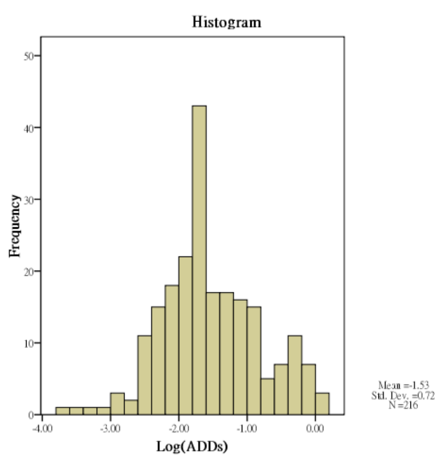
Log-transformed estimated average daily doses of Hg accounted for approximately 20% of the variance. When the most significant variables are added stepwise into the model, as shown on Model 5 in Table 55, the value of R^2 increased significantly to 0.40, indicating that 40% of variance of Hg concentrations in the study population's hair was explained by this model. When the demographic variables of 'location' and 'fisherman' were added to the regression models, the magnitudes of the variance on Hg concentrations in hair were enhanced significantly by 12.9% and 4.3%, respectively, showing that these variables were important factors in the exposure model. When the variable of 'gender' was added into the models, however, the effect of variation explanation contributed by this variable was insignificant.



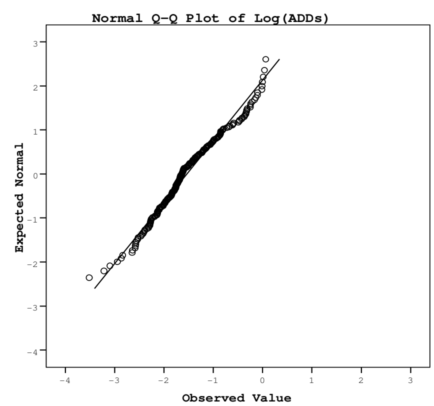
a. Histogram of Log(HairHg)



b. Q-Q plot of Log(HairHg)



c. Histogram of Log(ADDs)



d. Q-Q plot of Log(ADDs)

Figure 35 Histograms and Q-Q plots of log-transformed Hg concentrations in hair (Log(HairHg)) and daily intake doses of Hg (Log(ADDs))

The interactions of dietary intake rates of Hg with 'age' were demonstrated to contribute to the significant differences in predictability of the exposure model. Compared with Model 4 where there were only main effects selected, Model 5 consisting of main effects and interactive effects significantly explained the variance by 3.2%.

Table 55 Elevation of models using different number of input variables

	R^2	F	Change statistics	
			ΔR^2	F change
Model 1: Log-transformed estimated ADDs from questionnaires	0.194	50.190***	0.194	50.190***
Model 2: Log-transformed estimated ADDs from questionnaires, location	0.250	16.170***	0.129	7.740***
Model 3: Log-transformed estimated ADDs from questionnaires, location, fisherman	0.366	16.668***	0.043	13.622***
Model 4: Log-transformed estimated ADDs from questionnaires, location, fisherman, gender	0.372	14.883***	0.006	1.881
Model 5 (Established model): Log-transformed estimated ADDs from questionnaires, location, fisherman, gender, log-transformed estimated ADDs from questionnaires × age	0.404	15.061***	0.032	10.727**

***: significant level < 0.001

The residual versus fitted plot as shown in Figure 36 was used to test heteroscedasticity of the model. If the model was well-fitted, there should be no pattern to the residuals plotted against the fitted values. The pattern was unclear with randomly the distributing data points, an indication of mild heteroscedasticity.

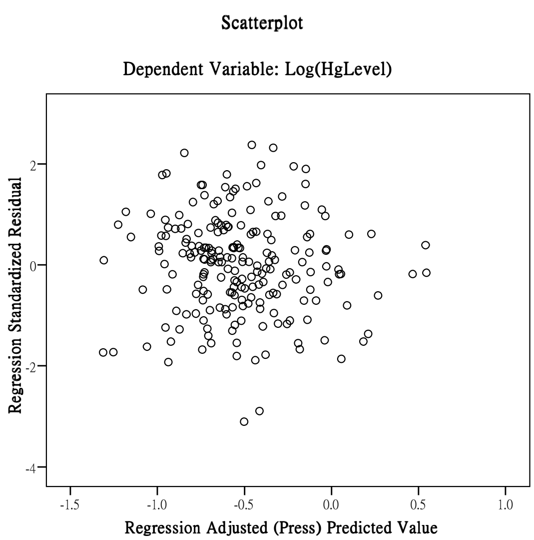


Figure 36 Scatter plot of residual versus fitted data of the developed model

The model given below (Equation 15) shows the fully expanded Model 5 and shows the significance of the different factors in adding to the body Hg burdens.

$$\begin{aligned}\text{Hg concentration in hair} = {} & \beta_0 + \beta_1 \cdot \text{Log(estimated Hg intake doses)} + \\ & \beta_{21} \cdot \text{(Temirtau new town)} + \beta_{22} \cdot \text{(Chkalovo)} + \\ & \beta_{23} \cdot \text{(Gagarinskoye)} + \beta_{24} \cdot \text{(Samarkand)} + \beta_{25} \cdot \text{(Rostovka)} + \\ & \beta_3 \cdot \text{fisherman} + \beta_4 \cdot \text{male} + \beta_5 \cdot 〔\text{Log(estimated Hg intake} \\ & \text{doses)} \times \text{age}〕\end{aligned}$$

Equation 15

The equation is simplified as Equation 16.

$$\begin{aligned}\text{Hg concentration in hair} = {} & \beta_0 + \beta_1 \cdot \text{Log(estimated Hg intake doses)} + \beta_{2i} \cdot \text{location}_i \\ & + \beta_3 \cdot \text{fisherman} + \beta_4 \cdot \text{male} + \beta_5 \cdot 〔\text{Log(estimated Hg intake} \\ & \text{doses)} \times \text{age}〕\end{aligned}$$

Equation 16

where i = 1-5; dummy variable ‘location’

The significant coefficients together with multicollinearity of the independent variables in the model are listed in Table 56. The values of ‘VIF’ (variance inflation factor) were all less than 20, indicating that the regression model estimates of the coefficients were stable and the standard errors for the coefficients were rarely inflated. Furthermore, the number of variables selected in this model was representative and appropriate.

In addition to log-transformed average daily Hg intake doses, the variable of ‘Rostovka’ was assessed as having a large effect (t = 3.956, *p.* <0.001) on the relationship. It implied that the Rostovka population had higher Hg concentrations in hair, compared with the population with the same fish and shellfish consumption rates in Temirtau old town. As higher Hg concentrations were found in the river fish in the downstream section of the River Nura than in the upstream (see Section 4.2.1), the result was not surprising.

Table 56 Model coefficient and multicollinearity associated with Hg levels in hair (dependent variable) and personal characteristics and dietary intake patterns (independent variables)

Variable		Symbol	Unstandardised coefficient	Standardised coefficient (β)	t.	Collinearity Statistics	
						Tolerance	VIF
(Constant)		β_0	-0.256		-2.511*		
Main effects							
Log(estimated Hg intake doses)		β_1	0.379	0.521	6.107***	0.410	2.440
Location[a]	Temirtau new town	β_{21}	0.062	0.051	0.698	0.568	1.759
	Chkalovo	β_{22}	0.160	0.104	1.552	0.663	1.507
	Gagarinskoye	β_{23}	-0.173	-0.134	-1.886	0.586	1.705
	Samarkand	β_{24}	-0.046	-0.029	-0.434	0.663	1.508
	Rostovka	β_{25}	0.396	0.269	3.956***	0.644	1.552
Fisherman[b]		β_3	0.333	0.185	3.037***	0.801	1.248
Male[c]		β_4	0.070	0.067	1.119	0.836	1.196
Interactive effect							
Log(estimated Hg intake doses) × age		β_5	-0.003	-0.265	-3.275***	0.456	2.192

a: Temirtau old town as the reference group.
b: Non-fisherman as the reference group.
c: Female as the reference group.
*: significant level < 0.05
***: significant level < 0.001

The variable of 'fisherman/or not' showed a significant effect on Hg concentrations in hair, indicating that fishermen had significantly higher levels of Hg in hair than the general population. According to the survey data, larger average daily portions of fish consumption for fishermen were demonstrated than for the general public, 103 g/day and 49 g/day, respectively, indicating that fish caught from the Hg-contaminated River Nura clearly being a major intake pathway for fishermen.

There was a significantly positive interaction between age and average daily intake doses of Hg. During the fieldtrip, fishing in the river or lakes was reported as a normal recreation for the local people, particularly for elderly or retired people. Fishing may be their major social activity in summer and river fish and is likely to be their primary fish source. They are also the poorest in society and river fish provided a cheap source of food. Hence, these wild fish with high Hg contents are likely to be the major source of the higher levels of Hg in the older generation.

7.4 Discussion

Correlation analysis in this chapter identified that Hg concentrations in hair were associated with daily Hg intake estimated from local food consumption (see Table 53), showing that Hg concentrations in hair were positively correlated with consumption rates of river fish and shellfish self-caught from the contaminated Nura River and the neighbouring lakes. In contrast, the contribution from the other foodstuffs to the elevated Hg concentrations in hair was limited, with a negative correlation between frequency of dairy consumption and hair Hg levels. The finding was in accordance with those of Passos et al. (2003) who conducted a study in the Amazon and reported that beef and chicken consumption had a negative correlation with Hg concentrations in the hair of pregnant women. Meat appeared to be a substitution for fish meals in the Amazonian Hg-polluted villages, in order to have a source of animal protein and diminish the risk of Hg exposure. The USEPA (1997f) and Lindberg et al. (2004) also reported that, in general, the terrestrial food chains can make only a slight contribution to human exposure to Hg.

An exposure model was established with variables relating to estimated Hg daily intake rates and demographic characteristics of the subjects including gender, residential location, age and frequent fishing activity. The model accounted for approximately 40% of the variation in Hg loads in the hair of the population. This is appreciably higher compared with other Hg exposure studies which found that they could only explain limited variation in their models with respect to Hg loads in humans' scalp hair, less than 20% (Santos et al., 2002, Dakeishi et al., 2005, IP et al., 2004, Legrand et al., 2005, Budtz-Jorgensen et al., 2007, Bjornberg et al., 2003).

Hg exposure levels in the study population were particularly high in the fishermen subgroup due to river fish intake. In the questionnaire survey, the nineteen fishermen at the study site reported eating river fish caught from the local areas approximately twice per week, whilst the general population ate river fish once per week. Such fish ingestion rates were lower than Japanese fishermen who consumed on average 333.6g of fish per day (Harada, 1995). A similar finding can be seen in the study of Lebel et al. (1997) who indicated that the local fishermen in the Amazon Basin consumed fish more frequently than the general male population, 68.8% ± 28.8 and 43.9% ± 33.6 on average. As a result, those Amazon fishermen had extremely high levels of hair Hg up to 142.4 μg/g.

The male population were shown to have significantly higher Hg concentrations in their hair than females (see Table 50 in Section 7.1). Burger (2000) suggested that it may be due to the fact that males carried out more fishing activities and hence generally had more chance than women to consume wild-caught fish. Likewise, several studies stated that the larger amounts of food consumption of males than females attributed to more Hg accumulation in males' hair (Foo et al., 1988, Johnsson et al., 2004, Knobeloch et al., 2007, Lee et al., 2000, Shimomura et al., 1980, Yasutake et al., 2003). Lipfert (1997) suggested that the test bias generating in studies of Hg dose-response relationships was likely to occur due to a short half-life of Hg. Therefore, if women eat fish less frequently than men, this could be a source of errors.

The positive coefficient for the interaction between average daily Hg intake doses and gender was insignificant (see Table 56 in Section 7.3). It may indicate that women having as much Hg intake via food chains as men were likely to accumulate similar amount of Hg in hair to men. Several studies have shown distinctive Hg absorption of females. For example, Nielsen and Andersen (1991) carried out an experimental study and found higher MeHg levels in the blood and brain of female than male mice due to the difference of toxicokinetics between male and female mice. An observation confirmed by Vahter et al. (1994) was that, with respect to chronic exposure, MeHg bonded to the fat in the blood and brain of female non-human primates. Kadar et al. (2005) suggested that the high lipid solubility of MeHg may favour the uptake of the Hg by women than men as they generally have higher lipid levels than men. However, there is still a need for further studies which employ comprehensive animal experimental studies to clarify the varied physio-characteristics between genders during Hg exposure. It is suggested that women of fertility age at the sites of such this study select imported fish and shellfish instead of that caught from the River Nura, in order to avoid potential Hg exposure risks on themselves and their offspring.

In this study, 'location' was an important factor resulting in statistically significant regional differences on Hg concentrations in the hair of the local people, accounting for approximately 13% of the variance in Hg exposure levels of the general population. These results appeared to reflect the differences between their dietary behaviour, particularly on the amounts and the sources of fish and shellfish ingested within the six residential areas. Based on the questionnaire data, the majority of the fish consumed by the local people living in the riverine villages of Samarkand and Rostovka were sourced from the river, whereas more than one-half of the fish consumed by the residents of Temirtau were mainly purchased from the local market (Table 31 in Section 4.3). Due to high Hg content in river fish, the population living in the villages had higher Hg concentrations in hair. In other words, the main reason for the observed regional differences in hair Hg levels was the different lifestyles in the residential areas. Feng (1998) reported similar results that dietary intake patterns caused regional difference on total Hg in hair. In his study, the residents in the coastal district had higher amounts of total Hg in their hair, compared with those living in the mountainous district and the middle district of the same Prefecture of Tokushima in Japan. More fish intake in the daily diet of the residents in the coastal district than the residents of the other areas was the main reason. As a result of a significant association between the residential area and Hg concentration in hair, 'location' could be acting as a proxy for one or more underlying variables. The real factors to represent regional characteristics should be considered for defining the effect of 'location' on Hg exposure levels in hair.

In addition to 'location', whether river fish or commercial fish was consumed potentially depended on the residents' social and economic conditions. For example, the love-to-eat-fish population may have a paid job and frequently purchase uncommon and imported fish and shellfish, whereas market fish and shellfish were less likely to be consumed regularly by the elder population in the study areas, as they were mostly pensioners or out of work with less financial support. Instead, many of the elder population fished for their fish supply. This practice may have resulted in the positive interaction of '$F_5 \times$ age' to Hg concentrations in hair. Since the dietary behaviour of the study population was observed to be connected to economic conditions, social economic factors (e.g. education level, employment status and job salary) should be investigated in future studies to help identify the population at most risk from Hg exposure.

After increasing several variables regarding individuals' demographic data, the relationship between Hg concentrations in hair and Hg exposure doses through dietary intake became more evident. The exposure model implicated that different Hg exposure risk was carried by different groups. As an exposure model consisting of demographic characteristics, it is beneficial to specify the potentially high-exposed population. In a long-term aspect, the model can be used to assess and give suggestions to the population particularly at high risk.

7.5 Summary

This study analysed the levels of human Hg exposure at the highly Hg polluted site at Temirtau. Subgroups of males, people aged over 45 and fishermen or anglers consuming river fish frequently were found to have elevated Hg exposure levels in hair. Although the population without chemical treatment on hair had statistically higher levels of Hg in hair than those with chemical treatment on hair, that result was demonstrated to confound with the factor of gender. As frequently consuming river fish was found to be positively related to increasing Hg concentrations in hair, the people exposed to high levels of Hg were likely to have large portions of fish and shellfish consumption.

This study established an exposure model to assess body burdens of Hg in hair with the variables of both dietary intake patterns and personal data. The variables accounted for approximately 40% of the variation in the predictive model for Hg concentrations in hair. It indicated that in addition to food consumption giving the effects on elevated Hg exposure levels in hair, the included demographic characteristics of gender, residential location, age and fishery occupation enhanced the explanation ability of the Hg exposure model. In spite of Hg levels in the hair of the study population related to individuals' demographic characteristics, the exposure model based on questionnaire survey data may not imply interindividual variability in a dose-response relationship, because interindividual variability needs further laboratory experiments to verify. This model rather implied that different lifestyles may result in varied Hg exposure risk. Lifestyle together with dietary behaviour should be taken into account in the studies associated with the relationship between true exposure and Hg intake, in order to clearly define the group potentially sensitive to Hg exposure.

8. CONCLUSIONS AND RECOMMENDATIONS

8.1 Conclusions

Because exposure estimate equations/models provide a quick, convenient and cost-efficient method to evaluate the potential risk of adverse health effects on humans, they are used to estimate potential exposure levels of common environmental toxicants in many chemically polluted areas. As risk managers often rely on results of exposure assessments to identify remediation, reliable validation of exposure dose assessments is highly desirable. The traditional exposure evaluation is conducted using the deterministic method that typically uses the worse cases (the 95^{th} percentile) as the single-value inputs to ensure the safe and conservative level produced for management, legislation and remediation. The deterministic method generally produces overestimated results, as overestimated Hg exposure doses have been seen in several studies (Gosselin et al., 2006, Canuel et al., 2006, Loranger et al., 2002). This study applied the deterministic method coupled with the exposure equations and questionnaire data to estimate Hg daily intake dose of the potentially exposed population in Central Kazakhstan. The result was approximately 1.5-time greater than the Hg intake dose derived from the highest Hg concentration in the hair of the study population. Hence, the deterministic Hg exposure estimate overestimated the body loads of Hg in hair, which have been reported to be one of the most reliable biomarkers for establishing actual levels of human exposure to Hg (WHO, 1990, Phelps et al., 1980, Lipfert, 1997).

To stochastically estimate the levels of Hg exposure, this study adopted the Monte Carlo simulation approach to establish the estimated average daily Hg intake doses from environmental variables contributing to the body loads of the population. The simulation gave a similar distribution to that established from the measured Hg concentrations in hair. Although compared with the data established from Hg levels in hair, this probabilistic simulation slightly overestimated the average daily Hg intake of the population by approximately 23%, less than one order of magnitude. The valid probabilistic exposure simulation was attributed to the use of a wide range of exposure data (e.g. Hg concentrations in the environmental media, average daily food consumption rates) covering the variance of the exposure activities among individuals. It was particularly essential that the comprehensive data about fish and shellfish consumption were identified individually via dietary questionnaires, as fish and shellfish consumption has been considered as the major route of Hg poising for humans (WHO, 1990, USEPA, 1997e). However, such self-reported data were attributed to uncertainties in the probabilistic simulations due to poor reliability of questionnaire data provided by interviewees. Similar findings were reported by several previous studies (Yokoo et al., 2001, Loranger et al., 2002, Tran et al., 2004, Canuel et al., 2006, Gosselin et al., 2006). Uncertainties may also originate from the limited sample numbers of drinking water, beef and vegetable when stratified by the six locations and the surrogates (e.g. ingestion rates of soil and water per day) inevitably used in the simulations. The uncertainties were thought to result in the difference between the probabilistic simulation data and the hair Hg exposure data. Despite all the uncertainties, the probabilistic method appears to be robust and effective in assessing the true levels of human exposure to Hg. This method is concluded to be very reliable for assessing human exposure risk.

The study has shown that the Hg concentrations in the hair of the study population ranged from 0.009 to 5.184 μg/g (mean = 0.577 μg/g), with approximately 17% of the hair samples exceeding the acceptable level of 1 μg/g of Hg in hair, corresponding to the RfD of 0.1 μg/kg body weight/day recommended by the USEPA (1997d). The exposure levels of the local population were not very severe, compared with the best-known Hg poisoning case of Minamata Disease. Lower levels of Hg exposure in hair may be due to that Hg being tightly bound to the fly ash at the study site, since Rio and Delebarre (2003) has demonstrated the good Hg adsorption ability of the sulfo-calcic fly ash, the type of fly ash at the study site. Furthermore, this study indicated that high levels of Hg accumulation in the hair of the local population were associated with frequently consuming river fish. Fishermen and male anglers were the populations most at risk, mainly due to the lifestyle of consuming self-caught fish from the polluted River Nura or the lakes derived from the river.

8.2 Recommendations

8.2.1 Recommendations for future research

1. The work identified fish consumption as the major source of Hg intake but

the intake model only accounted for approximately 40% of the variance in Hg concentrations in the hair of individuals. This sort of explanation level is higher than the findings of many other researches (Santos et al., 2002, Dakeishi et al., 2005, IP et al., 2004, Legrand et al., 2005, Budtz-Jorgensen et al., 2007, Bjornberg et al., 2003). Nearly all studies rely on only sampled dietary intake data which just accounted for small proportions of variation in the modelled Hg concentrations in hair. This study indicates that demographic characteristics of individuals and their interactive effects with dietary intake data should be considered in the establishment of the Hg exposure model.

2. During the work, a number of weaknesses become apparent in holding a single sampling campaign, namely people's response. The questionnaire survey clearly provided indicative data on the type of diet but it was relatively unreliable because of the recall difficulty. This was particularly important in Kazakhstan where the local people eat little fish in winter due to the thick ice on the lakes and eat more fish in summer when the weather is more suitable for fishing. It would be interesting to see if estimates of Hg intake varied at different times of year, as their diets changed dramatically between seasons. The accuracy of Hg exposure estimates might be improved by carrying out questionnaire surveys of households maybe four times a year. It would also be interesting to have a subsample data collection on food samples monthly to get an accurate estimate of the Hg intake level.
3. A single hair sampling period of the 29^{th}, June and the 9^{th}, July in 2005 could be problematic, as the hair samples collected from the root of the scalp only reflect the exposure over the previous one to two months. It is therefore difficult to say how representing the exposure levels in the hair samples are to the annual mean level of exposure. It would be helpful to look at Hg levels throughout a year to establish seasonal variation. A study of seasonal changes in Hg contents in hair would be useful in referring the estimates of annual hg exposure.
4. The safety levels of the daily Hg intake developed by the health authorities such as the USEPA (1997e) and the Agency for Toxic Substances and Disease Registry (1999a) are the indices for general public to assess the risk

of adverse health effects. Although Hg concentrations in hair can be adopted to restructure personal intake doses of Hg, interindividual differences on biological parameters such as rates of hair growth, blood:hair ratio and half life of Hg may contribute to variability in the conversion. As a result, the information of physio-kinetics of individuals or sophisticated toxicokinetic models could be used to estimate Hg intake doses from Hg concentrations in the biomarker of hair.

5. The factor 'location' was demonstrated to have an essential influence on humans exposed to Hg in the Nura villages. However, the significance behind this factor is correlated to the population's lifestyle. Since lifestyle has been observed to be connected to social economic status in this study, adding factors like family income or education level should improve the predictability of the exposure model.
6. The validity of the developed Hg exposure model needs to be confirmed. Future studies regarding the application of this exposure model to assess other Hg-exposed populations or larger scale surveys at Hg-exposed sites are desirable but beyond the scope of this study.

8.2.2 Recommendations for the local government

1. Local government needs to have an information campaign to raise awareness of the dangers of consuming fish caught in the River Nura and its oxbow lakes below Temirtau, as nearly 17% of the population could be considered at risk, all be it not critically. On the other hand, fish and shellfish are food sources rich in protein and low in saturated fat, direct dietary sources of beneficial omega-3 polyunsatuated fatty acids and docosahexaenoic acid which are important for brain and retinal development and contain antioxidants such as selenium and vitamin E (Harris, 1989, Connor, 2001). Fish consumption has been reported to reduce risk of coronary artery disease and sudden cardiac death (Kromhout et al., 1985, Albert et al., 1998). The n-3 fatty acids from fish oil have powerful antithrombotic actions to lower very-low density lipoprotein cholesterol and triglyceride levels, inhibit platelet aggregation and reduce blood pressure (Goodnight et al., 1981, Connor, 2001). Therefore, if the River Nura fish are consumed against the

advice not to consume it, the consumption rate of no more than once a week is recommended. Pregnant women should not eat river caught fish. This is based on the mean concentration of Hg in the river fish being 0.43 µg/g (see Table 24 and Table 25 in Section 4.2.1) and on average body weight of 67 kg of the local people. The result is that at this concentration, people need to eat less than on average of 33 g of fish per day if they are not to exceed the reference dose of 0.1 µg/kg body weight/day (assuming 95% of the Hg is as MeHg). This assumes a conservative meal size of 8 oz (equal to 225g), referring to the survey of the USEPA (1997a), and a safety factor of two.

2. There should be regular monitoring of Hg exposure levels in the population most at risk, like fishery occupational people and anglers. Hg concentration measurement in their hair should be carried out seasonally, in order to diminish the exposure impacts on human health.

APPENDIX A: Questionnaire for households and dust, soils and water sample collection

Sample List For Households

Area / Code	___________________________________

Part A. Dust

Sample Code & Place	Surface type
HD Floor (2 samples)	
☐ Playroom (Size? Clean frequency? How long they stay?) ________________________________	☐ Smooth floor ☐ Carpeted floor ☐ Others __________
☐ Living room (Size? Clean frequency? How long they stay?) ________________________________	☐ Smooth floor ☐ Carpeted floor ☐ Others __________
☐ Bedroom 1(Size? Clean frequency? How long they stay?) ________________________________	☐ Smooth floor ☐ Carpeted floor ☐ Others __________
☐ Bedroom 2 (Size? Clean frequency? How long they stay?) ________________________________	☐ Smooth floor ☐ Carpeted floor ☐ Others __________
☐ Other (Where? Clean frequency? How long they stay?) ________________________________	☐ Smooth floor ☐ Carpeted floor ☐ Others __________
HW Window (2-4 samples)	
Kitchen (Size? Clean frequency? How long they stay?) ____________________	☐ Interior window sill ☐ window trough
Total number of samples: __________	

Part B. Soils

Sample Code. & Area (record name of area used by the owner or residents; larger than 1 yr^2)	Bare or Covered
S Outdoor soils	
☐ Outdoor play area (Where? Size? Frequency? How long they stay?) ________________________________	☐ Bare ☐ Covered
☐ Garden (Where? Size? Frequency? How long they stay?) ________________________________	☐ Bare ☐ Covered
☐ Other (Where? Size? Frequency? How long they stay?) ________________________________	☐ Bare ☐ Covered
Total number of samples: __________	

Part C. Water

Sample Code. & Area	**Purpose**		
W Water			
☐ Tap water in the kitchen ______	☐ Drinking	☐ Cooking	☐ Swimming
	☐ Bathing	☐ Washing	☐ Irrigating
☐ Well ______	☐ Drinking	☐ Cooking	☐ Swimming
	☐ Bathing	☐ Washing	☐ Irrigating
☐ Standpipe ______	☐ Drinking	☐ Cooking	☐ Swimming
	☐ Bathing	☐ Washing	☐ Irrigating
☐ Other ______	☐ Drinking	☐ Cooking	☐ Swimming
	☐ Bathing	☐ Washing	☐ Irrigating
Total number of samples: ______			

Part D. General Data

Property address (Area)	______
Name of property owner (Surname)	______
The age of the house	______ years
Historical engineering of the house in past 2 years?	______ ______
Members of this family? Pets?	______
How many kids in the family? Age?	1/2/3/4/5/6, Age ______
Is there any door mat?	☐ No ☐ Yes, where: ______
Is there any shoe-removal place?	☐ No ☐ Yes, where: ______

APPENDIX B: Questionnaire of loose dust collecting from communal areas

Sample List For Public Area

Area / Code	______________________________

Part A. Loose Dust- Main Street

Sample Code & Place	Surface type
MS Main Street	
☐ Street 1 (Where? Name? GPS position? Surface Material?) ______________________________	
☐ Street 2 (Where? Name? GPS position? Surface Material?) ______________________________	
☐ Street 3 (Where? Name? GPS position? Surface Material?) ______________________________	
Total number of samples: ___________	

Part B. Loose Dust- The Front Of The Building

Sample Code & Place
FB The front of a communal building's entrance
☐ Building 1 (Where? Size? Name? GPS position? Surface Material?) ______________________________
☐ Building 2 (Where? Size? Name? GPS position? Surface Material?) ______________________________
☐ Building 3 (Where? Size? Name? GPS position? Surface Material?) ______________________________
☐ Building 4 (Where? Size? Name? GPS position? Surface Material?) ______________________________
☐ Building 5 (Where? Size? Name? GPS position? Surface Material?) ______________________________
Total number of samples: ___________

Part C. Loose Dust- The Communal Entrance

Sample Code & Place
IE Inside that communal entrance
☐ Building 1 (Where? Size? Name? GPS position? Surface Material?) ____________________
☐ Building 2 (Where? Size? Name? GPS position? Surface Material?) ____________________
☐ Building 3 (Where? Size? Name? GPS position? Surface Material?) ____________________
☐ Building 4 (Where? Size? Name? GPS position? Surface Material?) ____________________
☐ Building 5 (Where? Size? Name? GPS position? Surface Material?) ____________________
Total number of samples: _____________

APPENDIX C: Food frequency questionnaire

(English version)

Food Intake Questionnaire

The data in this questionnaire is used for academic study. Your personal information will not be exposed for any reasons. The questionnaire is divided into five sections (Part A to E): Fish Consumption, What Kind Of Fish Do You Eat, What Kind Of Food Do You Eat, General Data, and Fish Photos (for your reference).

Part A. Fish Consumption

Please check the frequency of your fish intake behaviour in the past one year. Listed below are statements about fish intake behaviour. Please check one box to indicate which you agree or disagree with the statement.

1. Did you eat fish in the past one year?

☐ No **(Jump to Part C.)** ☐ Yes **(Continue answering following questions)**

Part B. What Kind Of Fish Do You Eat?

There are five kinds of classification about the waterbody's food: ***a. pike or zander; b. bream or roach; c. carp; d. crayfish; e. shellfish.*** Please check the sources and the frequency you ate in the past one year. You can have more than one source for getting the food, but please check only one answer for each frequency and average serve. Please add more if you intake other fish by yourself.

B.1 Pike or Zander

1. Do you eat **Pike or Zander**?

☐ No **(Jump to next fish species)** ☐ Yes **(Continue answering following questions)**

2. How often do you eat these kinds of fish?

☐ 1~6 times per year	☐ 1 time per month	☐ 2~3 times per month
☐ 1 time per week	☐ 2 times per week	☐ 3~4 times per week
☐ 5~6 time per week	☐ 1 time per day	☐ 2 or more times per day

3. Where do you buy the fish?

Source (Choose more than one answer)	How Often? (One answer)					Average Serve/Per Time (One answer)				
	Seldom	Occasionally	Sometimes	Frequently	Always	1 serve	2 services	3 services	4 services	5 services
☐ Self-fishing	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Fresh market (Bazzar), from _______	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Canned market food	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Frozen market food, from_____	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐

B.2 Bream or Roach

1. Do you eat **Bream or Roach**?

☐ No **(Jump to next fish species)** ☐ Yes **(Continue answering following questions)**

2. How often do you eat these kinds of fish?

☐ 1~6 times per year ☐ 1 time per month ☐ 2~3 times per month
☐ 1 time per week ☐ 2 times per week ☐ 3~4 times per week
☐ 5~6 time per week ☐ 1 time per day ☐ 2 or more times per day

3. Where do you buy the fish?

Source (Choose more than one answer)	How Often? (One answer)					Average Serve/Per Time (One answer)				
	Seldom	Occasionally	Sometimes	Frequently	Always	1 serve	2 services	3 services	4 services	5 services
☐ Self-fishing	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Fresh market (Bazzar), from ______	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Canned market food	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Frozen market food, from_____	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐

B.3 Carp

1. Do you eat **Carp**?

☐ No **(Jump to next fish species)** ☐ Yes **(Continue answering following questions)**

2. How often do you eat this kind of fish?

☐ 1~6 times per year ☐ 1 time per month ☐ 2~3 times per month
☐ 1 time per week ☐ 2 times per week ☐ 3~4 times per week
☐ 5~6 time per week ☐ 1 time per day ☐ 2 or more times per day

3. Where do you buy the fish?

Source (Choose more than one answer)	How Often? (One answer)					Average Serve/Per Time (One answer)				
	Seldom	Occasionally	Sometimes	Frequently	Always	1 serve	2 services	3 services	4 services	5 services
☐ Self-fishing	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Fresh market (Bazzar), from ______	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Canned market food	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Frozen market food, from_____	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐

B.4 Crayfish

1. Do you eat **Crayfish**?

☐ No **(Jump to next fish species)** ☐ Yes **(Continue answering following questions)**

2. How often do you eat this kind of fish?

☐ 1~6 times per year ☐ 1 time per month ☐ 2~3 times per month
☐ 1 time per week ☐ 2 times per week ☐ 3~4 times per week
☐ 5~6 time per week ☐ 1 time per day ☐ 2 or more times per day

3. Where do you buy the fish?

Source (Choose more than one answer)	How Often? (One answer)					Average Serve/Per Time (One answer)				
	Seldom	Occasionally	Sometimes	Frequently	Always	1 serve	2 services	3 services	4 services	5 services
☐ Self-fishing	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Fresh market (Bazzar), from ______	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Canned market food	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Frozen market food, from______	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐

B.5 Shellfish

1. Do you eat **Shellfish**?

☐ No **(Jump to next fish species)** ☐ Yes **(Continue answering following questions)**

2. How often do you eat this kind of fish?

☐ 1~6 times per year ☐ 1 time per month ☐ 2~3 times per month
☐ 1 time per week ☐ 2 times per week ☐ 3~4 times per week
☐ 5~6 time per week ☐ 1 time per day ☐ 2 or more times per day

3. Where do you buy the fish?

Source (Choose more than one answer)	How Often? (One answer)					Average Serve/Per Time (One answer)				
	Seldom	Occasionally	Sometimes	Frequently	Always	1 serve	2 services	3 services	4 services	5 services
☐ Self-fishing	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Fresh market (Bazzar), from ______	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Canned market food	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Frozen market food, from______	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐

B.6 Other fish _________

1. Do you eat **Other fish**?

☐ No **(Jump to next fish species)** ☐ Yes **(Continue answering following questions)**

2. How often do you eat this kind of fish?

☐ 1~6 times per year ☐ 1 time per month ☐ 2~3 times per month
☐ 1 time per week ☐ 2 times per week ☐ 3~4 times per week
☐ 5~6 time per week ☐ 1 time per day ☐ 2 or more times per day

3. Where do you buy the fish?

Source (Choose more than one answer)	**How Often? (One answer)**					**Average Serve/Per Time (One answer)**				
	Seldom	Occasionally	Sometimes	Frequently	Always	1 serve	2 services	3 services	4 services	5 services
☐ Self-fishing	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Fresh market (Bazzar), from _______	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Canned market food	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Frozen market food, from_____	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐

B.7 Other fish _________

1. Do you eat **Other fish**?

☐ No **(Jump to next fish species)** ☐ Yes **(Continue answering following questions)**

2. How often do you eat this kind of fish?

☐ 1~6 times per year ☐ 1 time per month ☐ 2~3 times per month
☐ 1 time per week ☐ 2 times per week ☐ 3~4 times per week
☐ 5~6 time per week ☐ 1 time per day ☐ 2 or more times per day

3. Where do you buy the fish?

Source (Choose more than one answer)	**How Often? (One answer)**					**Average Serve/Per Time (One answer)**				
	Seldom	Occasionally	Sometimes	Frequently	Always	1 serve	2 services	3 services	4 services	5 services
☐ Self-fishing	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Fresh market (Bazzar), from _______	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Canned market food	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Frozen market food, from_____	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐

Part C. What Kind Of Food Do You Eat?

There are three kinds of food: ***a. Beef; b. Dairy; c. Tomato; d. Cucumber.*** Please check the sources and the frequency you ate in the past one year. You can have more than one source for getting the food, but please check only one answer for each frequency and average serve.

C.1 Beef

1. Do you eat **Beef**?

☐ No **(Jump to next food kind)** ☐ Yes **(Continue answering following questions)**

2. How often do you eat this kind of food?

☐ 1~6 times per year	☐ 1 time per month	☐ 2~3 times per month
☐ 1 time per week	☐ 2 times per week	☐ 3~4 times per week
☐ 5~6 time per week	☐ 1 time per day	☐ 2 or more times per day

3. Where do you buy the food?

Source (Choose more than one answer)	**How Often? (One answer)**					**Average Serve/Per Time (One answer)**				
	Seldom	Occasionally	Sometimes	Frequently	Always	1 serve	2 services	3 services	4 services	5 services
☐ Self-fed	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Fresh market (Bazzar), from _______	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Canned market food	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Frozen market food, from_____	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐

C.2 Dairy

1. Do you eat **Dairy**?

☐ No **(Jump to next food kind)** ☐ Yes **(Continue answering following questions)**

2. How often do you eat this kind of food?

☐ 1~6 times per year	☐ 1 time per month	☐ 2~3 times per month
☐ 1 time per week	☐ 2 times per week	☐ 3~4 times per week
☐ 5~6 time per week	☐ 1 time per day	☐ 2 or more times per day

3. Where do you buy the food?

Source (Choose more than one answer)	How Often? (One answer)					Average Serve/Per Time (One answer)				
	Seldom	Occasionally	Sometimes	Frequently	Always	1 serve	2 services	3 services	4 services	5 services
☐ Self-fed	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Fresh market (Bazzar), from ______	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Canned market food	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Frozen market food, from______	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐

C.3 Tomato

1. Do you eat?

☐ No **(Jump to next food kind)** ☐ Yes **(Continue answering following questions)**

2. How often do you eat this kind of food?

☐ 1~6 times per year	☐ 1 time per month	☐ 2~3 times per month
☐ 1 time per week	☐ 2 times per week	☐ 3~4 times per week
☐ 5~6 time per week	☐ 1 time per day	☐ 2 or more times per day

3. Where do you buy the food?

Source (Choose more than one answer)	How Often? (One answer)					Average Serve/Per Time (One answer)				
	Seldom	Occasionally	Sometimes	Frequently	Always	1 serve	2 services	3 services	4 services	5 services
☐ Self-grown	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Fresh market (Bazzar), from ______	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Canned market food	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Frozen market food, from______	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐

C.4 Cucumber

1. Do you eat?

☐ No **(Jump to next food kind)** ☐ Yes **(Continue answering following questions)**

2. How often do you eat this kind of food?

☐ 1~6 times per year	☐ 1 time per month	☐ 2~3 times per month
☐ 1 time per week	☐ 2 times per week	☐ 3~4 times per week
☐ 5~6 time per week	☐ 1 time per day	☐ 2 or more times per day

3. Where do you buy the food?

Source (Choose more than one answer)	**How Often? (One answer)**					**Average Serve/Per Time (One answer)**				
	Seldom	Occasionally	Sometimes	Frequently	Always	1 serve	2 services	3 services	4 services	5 services
☐ Self-grown	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Fresh market (Bazzar), from ________	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Canned market food	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
☐ Frozen market food, from______	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐

Part D. General Data

The purpose of the part is to keep traceable only if this questionnaire is uncompleted. Your personal data will be confidential. Only for this research.

1. Name: (Surname) ________________________	2. Sex: ☐ Male ☐ Female
3. Nationality: ______________	4. Occupation: ____________________

5. Age

☐ Under 16 ☐ 16-30 ☐ 31-45 ☐ 45 above

6. Residential area

☐ Temirtau ☐ Chkalovo ☐ Gagarinskoye
☐ Kalininskoe ☐ Samarkand ☐ Tegiz-Zhol

7. How weight do you have? (Kg)

☐ Less than 40 ☐ 40.01~60.00 ☐ 60.01~80.00 ☐ 80.01~100.00
☐ More than 100

8. Where do you get your drinking water?

____

9. Do you have siblings?

☐ No ☐ Yes, I'm the 1st/2nd/3rd/4th/5th/6th/____ child

10. Did you do artificial waving & colouring of hair in recent one month?

☐ No ☐ Yes

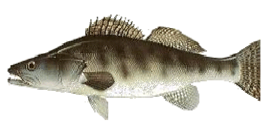
11. If you are a female, are you pregnant now?

☐ No ☐ Yes,________________-month

*** Need to know the result of the survey? ☐ No ☐ Yes_**

Part E. Fish Photos

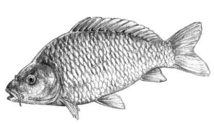
Pike



Zander

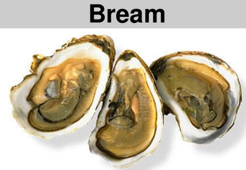
Bream

Roach



Carp

Crayfish



Shellfish

APPENDIX D: GIS coordinates of samples and questionnaire participants

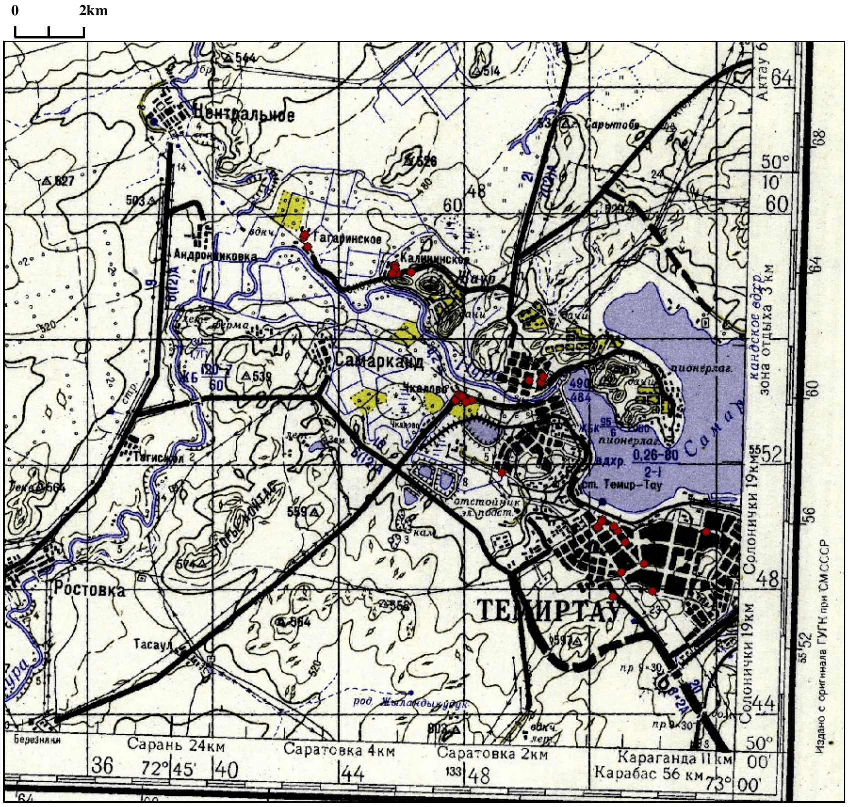


Figure D- 1 GIS coordinates of households participated in the survey

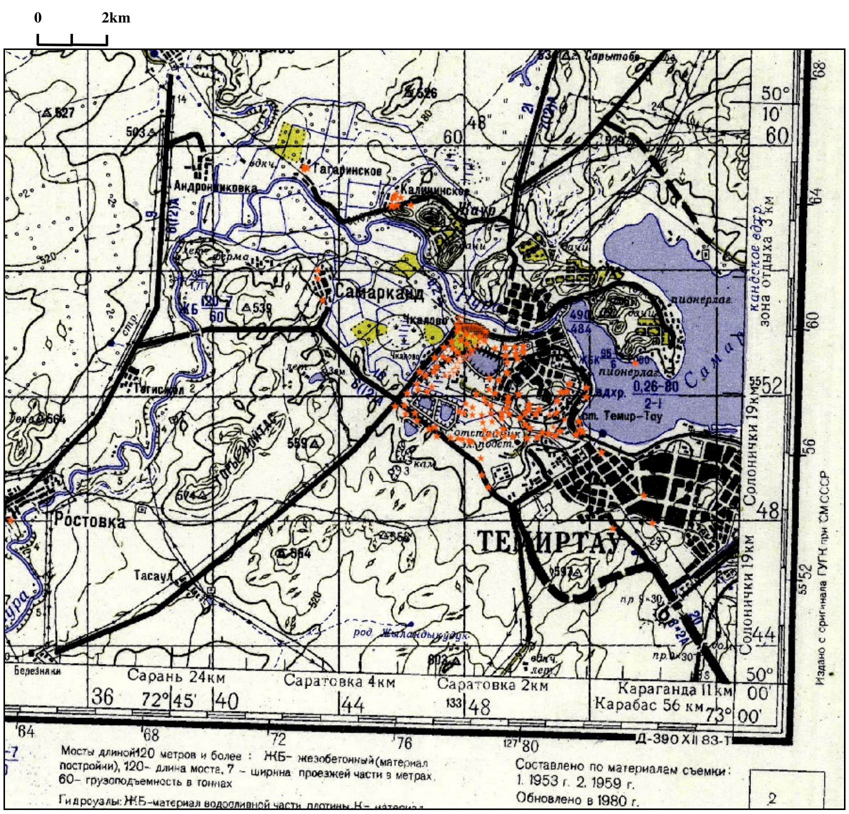


Figure D- 2 GIS coordinates of soil samples in Hg exposure simulations

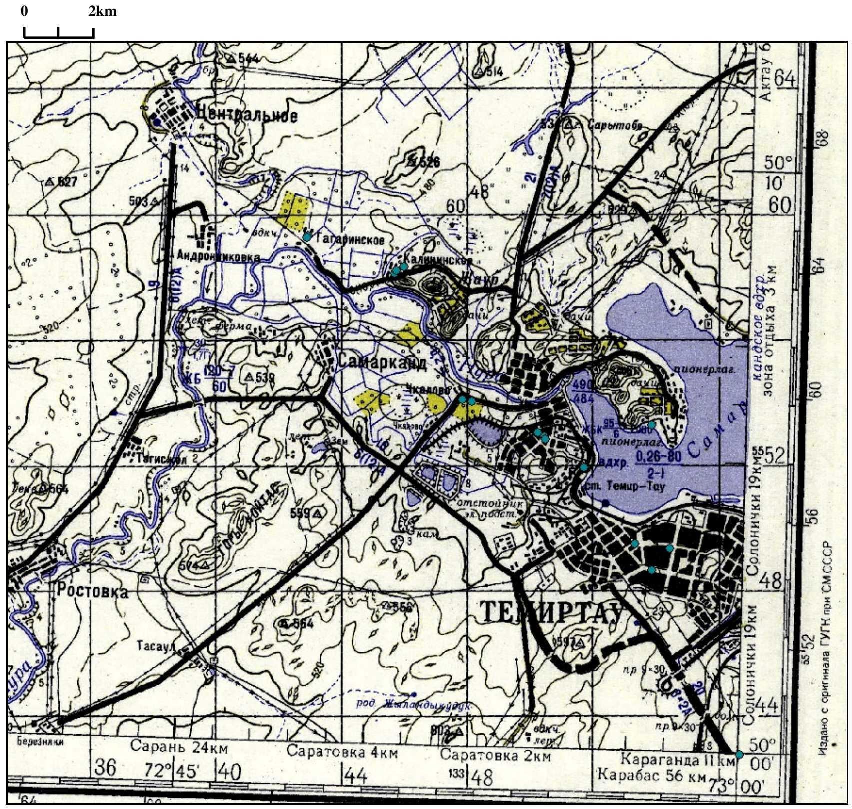


Figure D- 3 GIS coordinates of loose dust samples in Hg exposure simulations

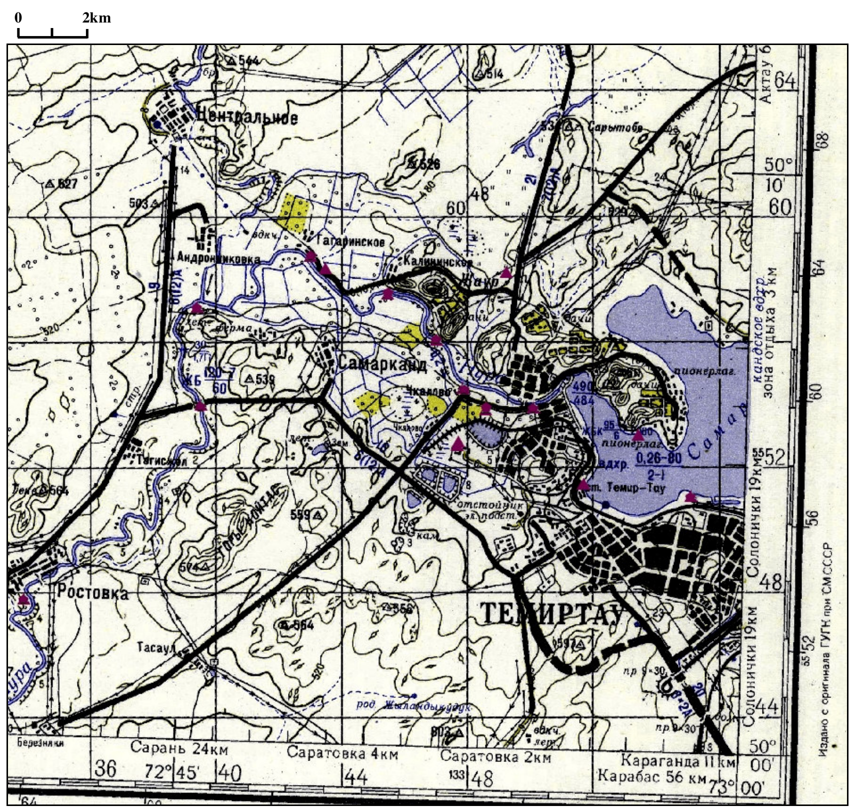


Figure D- 4 GIS coordinates of surface water samples in Hg exposure simulations

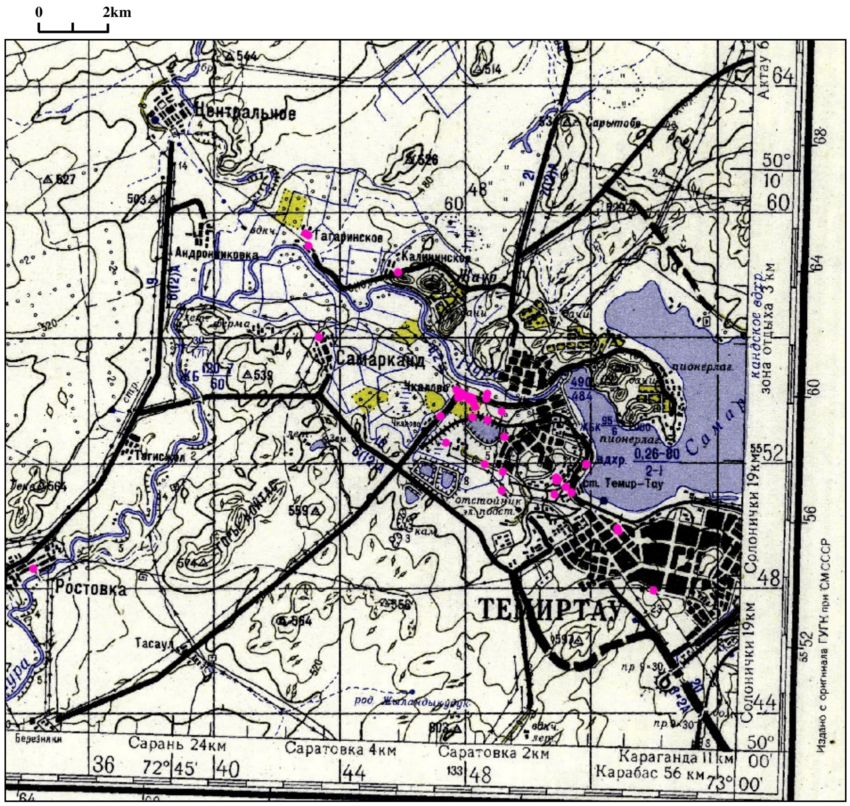


Figure D- 5 GIS coordinates of groundwater samples in Hg exposure simulations

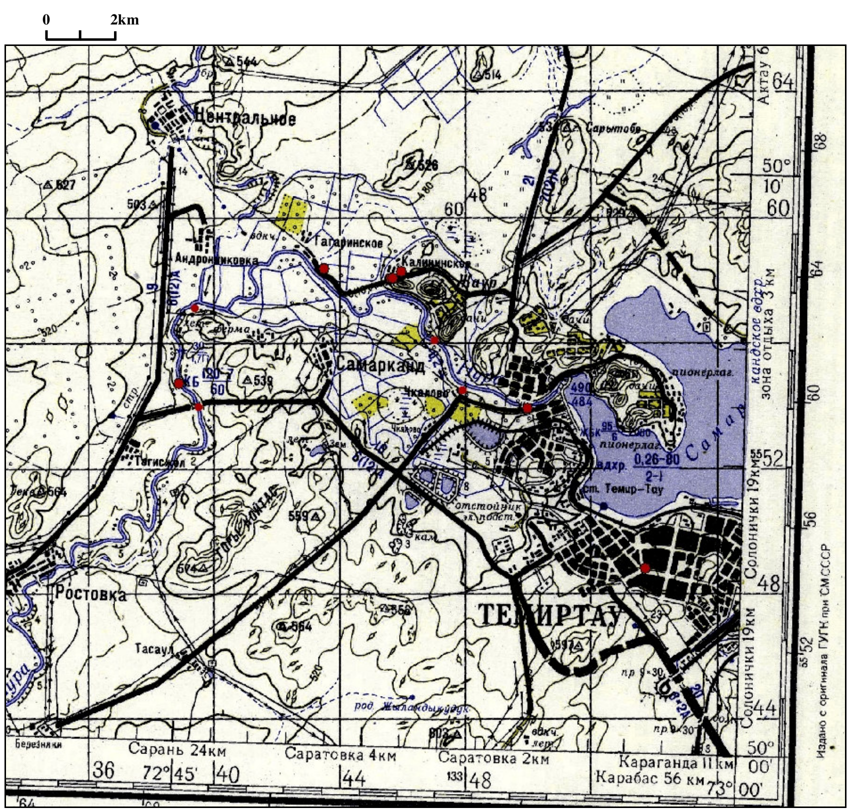


Figure D- 6 GIS coordinates of fish samples in Hg exposure simulations

0 2km

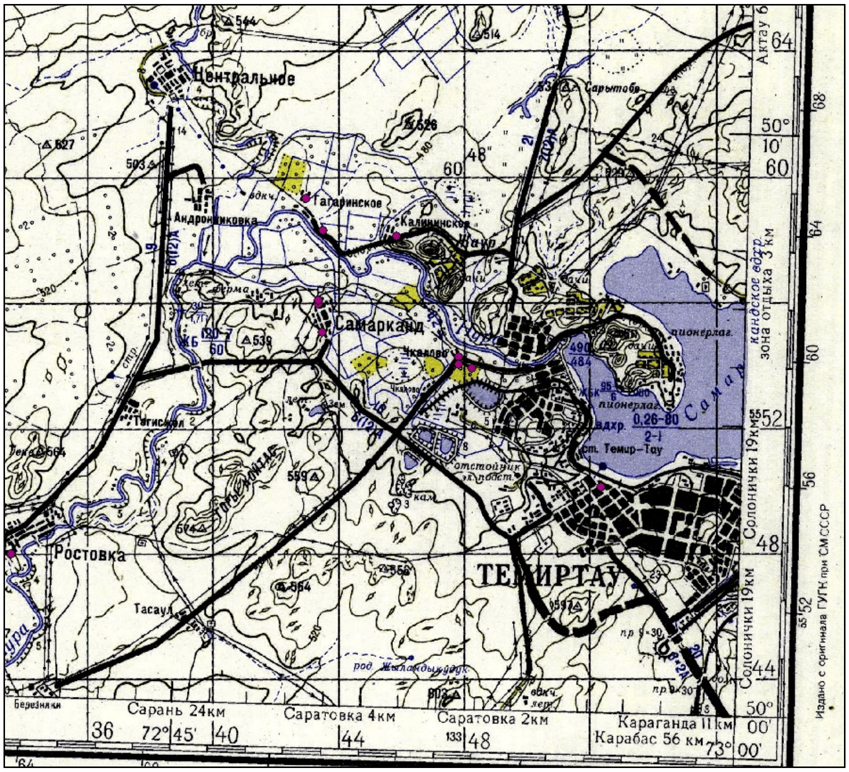


Figure D- 7 GIS coordinates of samples of beef, dairy, tomatoes or cucumbers in Hg exposure simulations

0 2km

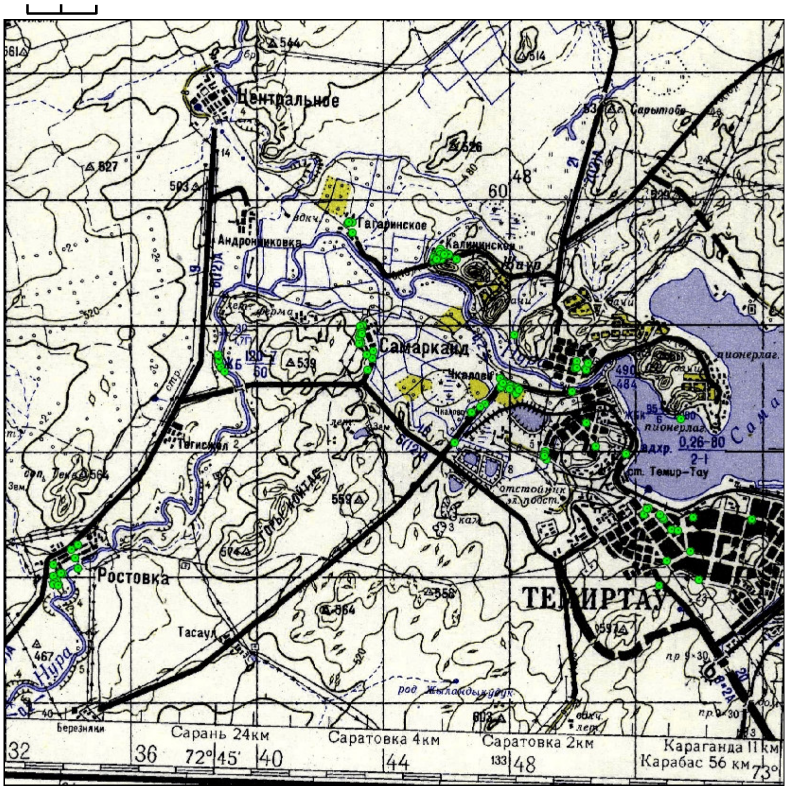


Figure D- 8 GIS coordinates of samples of hair together with food frequency questionnaire participants

APPENDIX E: Distributions and curves of general exposure parameters used in the Monte Carlo simulations

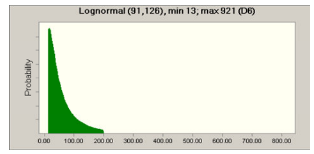
Table E- 1 Distribution and curves of the parameters for soil exposure simulations

Assumption: Ingestion rates of soils (mg/day)

Lognormal distribution with parameters:

Mean	65.00
Std. Dev.	82.00

Selected range is from 10.00 to 200.00

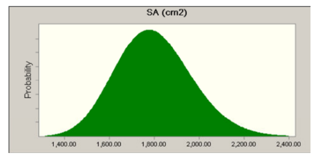


Assumption: Surface area of skin exposure (cm2)

Lognormal distribution with parameters:

Mean	1,800.00
Std. Dev.	170.00

Selected range is from 0.00 to 2,400.00

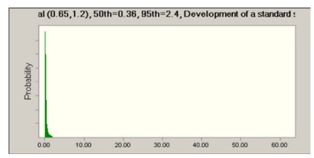


Assumption: Assumption: Adhesion factor (mg/cm2)

Lognormal distribution with parameters:

Mean	0.81
Std. Dev.	8.30

Selected range is from 0.00 to 2.10



Assumption: Fraction of soil ingestion from vegetables

Triangular distribution with parameters:

Minimum	0.00
Likeliest	0.13
Maximum	0.30

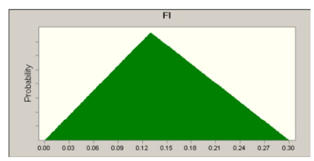


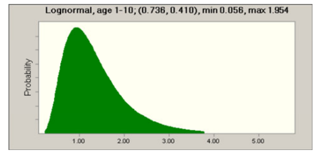
Table E- 2 Distribution and curves of the parameters for water exposure simulations

Assumption: Ingestion rate of drinking water

Lognormal distribution with parameters:

Mean	1.37
Std. Dev.	0.73

Selected range is from 0.15 to 3.78

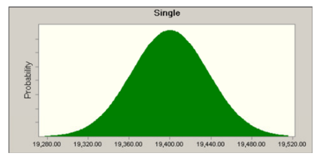


Assumption: Total skin surface area

Lognormal distribution with parameters:

Mean	19,400.00
Std. Dev.	37.40

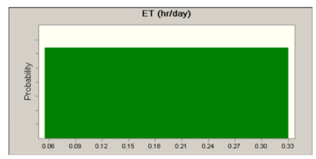
Selected range is from 16,600.00 to 22,800.00



Assumption: Exposure time during showering

Uniform distribution with parameters:

Minimum	0.06
Maximum	0.33



Assumption: Assumption: Exposure frequency of swimming

Triangular distribution with parameters:

Minimum	0.00
Likeliest	7.00
Maximum	60.00

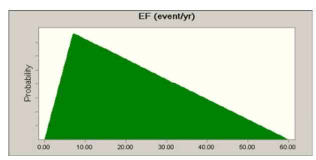


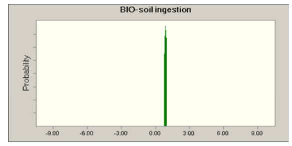
Table E- 3 Distribution and curves of general parameters for exposure simulations

Assumption: Bioavailability of MeHg that is absorbed via ingestion

Triangular distribution with parameters:

Minimum	-10.00
Likeliest	0.00
Maximum	10.00

Selected range is from 0.80 to 1.00

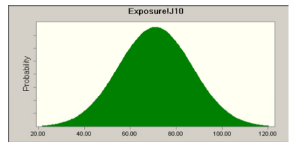


Assumption: Body weight (kg) in Temritau old town

Normal distribution with parameters:

Mean	70.90
Std. Dev.	15.98

Selected range is from 0.00 to 150.00

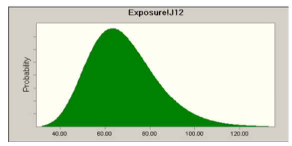


Assumption: Body weight (kg) in Temritau new town

Lognormal distribution with parameters:

Mean	68.19
Std. Dev.	15.51

Selected range is from 0.00 to 150.00

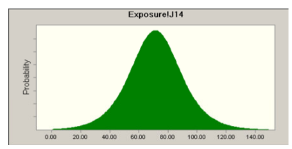


Assumption: Body weight (kg) in Chkalovo

Logistic distribution with parameters:

Mean	71.30
Scale	11.30

Selected range is from 0.00 to 150.00

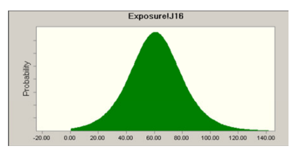


Assumption: Body weight (kg) in Gagarinskoye

Logistic distribution with parameters:

Mean	60.75
Scale	11.73

Selected range is from 0.00 to 150.00

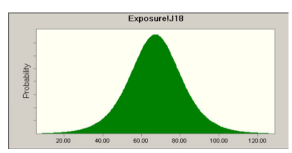


Assumption: Body weight (kg) in Samarkand

Logistic distribution with parameters:

Mean	67.16
Scale	8.48

Selected range is from 0.00 to 150.00

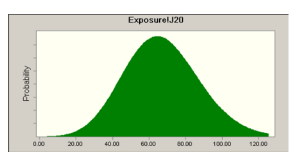


Assumption: Rostovka

Gamma distribution with parameters:

Location	-92.10
Scale	2.69
Shape	59.12855773

Selected range is from 0.00 to 150.00

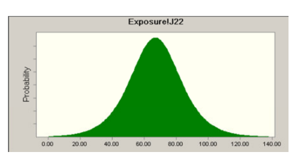


Assumption: Body weight (kg) in whole study areas

Logistic distribution with parameters:

Mean	67.01
Scale	10.26

Selected range is from 0.00 to 150.00



APPENDIX F: Distributions and curves of Hg concentrations in various environmental media for Monte Carlo simulations

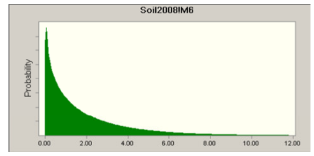
Table F- 1 Distributions and curves of Hg concentrations (mg/kg) in soils in different residential areas

Assumption: Hg concentrations in soils in Temirtau old town

Gamma distribution with parameters:

Location	-0.02
Scale	2.54
Shape	0.75

Selected range is from 0.00 to 1,000.00

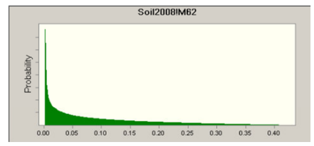


Assumption: Hg concentrations in soils in Temirtau new town

Beta distribution with parameters:

Minimum	0.00
Maximum	0.43
Alpha	0.53
Beta	1.92

Selected range is from 0.00 to 1,000.00

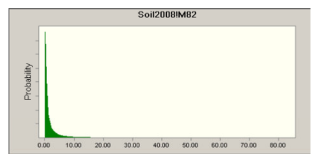


Assumption: Hg concentrations in soils in Chkalovo

Lognormal distribution with parameters:

Mean	2.40
Std. Dev.	7.29

Selected range is from 0.00 to 1,000.00

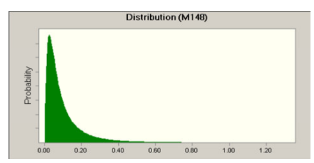


Assumption: Hg concentrations in soils in Gagarinskoye

Lognormal distribution with parameters:

Mean	0.11
Std. Dev.	0.13

Selected range is from 0.00 to 1,000.00



Assumption: Hg concentrations in soils in whole study areas

Lognormal distribution with parameters:

Mean	2.27
Std. Dev.	11.85

Selected range is from 0.00 to 1,000.00

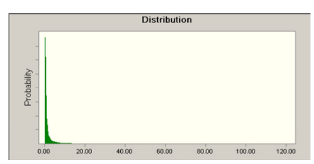


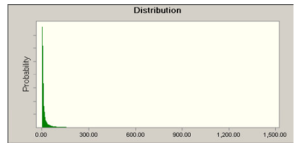
Table F- 2 Distributions and curves of Hg concentrations in surface water (ng/L) in different residential areas

Assumption: Hg concentration in surface water in Temirtau old town

Lognormal distribution with parameters:

Mean	26.52
Std. Dev.	148.63

Selected range is from 0.00 to 4,000.00

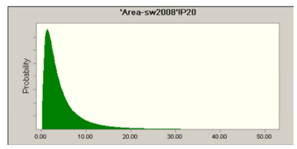


Assumption: Hg concentration in surface water in Temirtau new town

Lognormal distribution with parameters:

Mean	4.62
Std. Dev.	5.32

Selected range is from 0.00 to 4,000.00

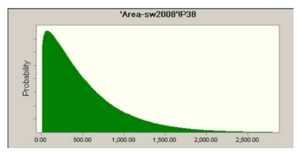


Assumption: Hg concentration in surface water in Chkalovo

Weibull distribution with parameters:

Location	10.31
Scale	530.86
Shape	1.10

Selected range is from 0.00 to 4,000.00

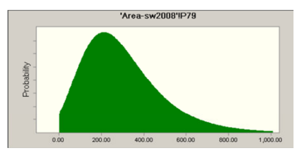


Assumption: Hg concentration in surface water in Gagarinskoye

Maximum Extreme distribution with parameters:

Likeliest	211.40
Scale	150.81

Selected range is from 0.00 to 4,000.00

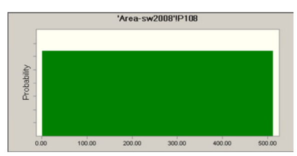


Assumption: Hg concentration in surface water in Samarkand

Uniform distribution with parameters:

Minimum	0.00
Maximum	512.00

Selected range is from 0.00 to 4,000.00

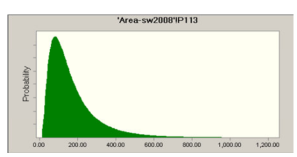


Assumption: Hg concentration in surface water in Rostovka

Lognormal distribution with parameters:

Mean	173.83
Std. Dev.	141.82

Selected range is from 0.00 to 4,000.00



Assumption: Hg concentration in surface water in whole study areas

Gamma distribution with parameters:

Location	-1.80
Scale	481.67
Shape	0.66

Selected range is from 0.00 to 4,000.00

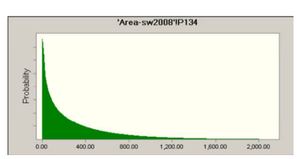


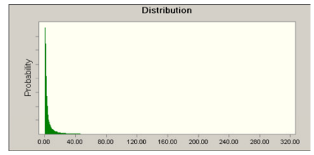
Table F- 3 Distributions and curves of Hg concentrations (ng/L) in groundwater in different residential areas

Assumption: Hg concentration in groundwater in Temirtau old town and Chkalovo

Lognormal distribution with parameters:

Mean	7.49
Std. Dev.	28.61

Selected range is from 0.00 to 500.00

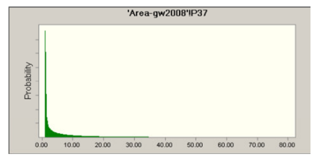


Assumption: Hg concentration in groundwater in Gagrinskoye

Gamma distribution with parameters:

Location	0.94
Scale	25.36
Shape	0.28

Selected range is from 0.00 to 500.00



Assumption: Hg concentration in groundwater in whole study areas

Lognormal distribution with parameters:

Mean	7.69
Std. Dev.	25.31

Selected range is from 0.00 to 500.00

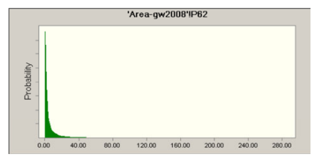


Table F- 4 Distributions and curves of Hg concentrations (ng/m^3) in the air in Temirtau and the whole study areas

Assumption: Hg concentration in air in Temirtau old town and the whole study areas

Lognormal distribution with parameters:

Mean	45.00
Std. Dev.	20.00

Selected range is from 75.00 to 300.00

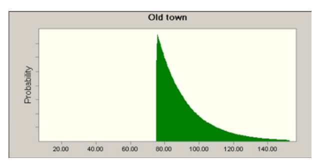


Table F- 5 Distributions and curves of Hg concentrations (mg/kg) in market fish in different residential areas

Assumption: Hg concentration in market fish

Uniform distribution with parameters:

Minimum	0.01
Maximum	0.07

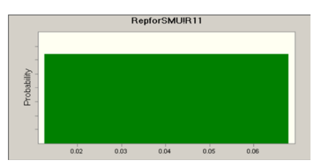


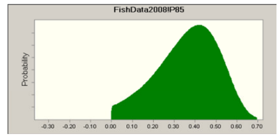
Table F- 6 Distributions and curves of Hg concentrations (mg/kg) in river fish in different residential areas

Assumption: Hg concentration in river pike, zander, crayfish, shellfish and other fish species as a w

Minimum Extreme distribution with parameters:

Likeliest	0.42
Scale	0.14

Selected range is from 0.00 to 2.00

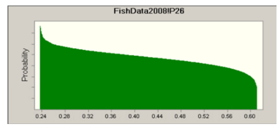


Assumption: Hg concentration in river bream/roach in Temirtau old town, new town and Chkalovo

Beta distribution with parameters:

Minimum	0.24
Maximum	0.61
Alpha	0.94
Beta	1.15

Selected range is from 0.00 to 2.00

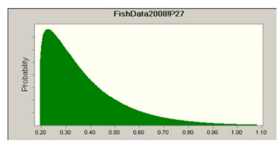


Assumption: Hg concentration in river bream/roach in Gagarinskoye

Gamma distribution with parameters:

Location	0.19
Scale	0.15
Shape	1.21

Selected range is from 0.00 to 2.00

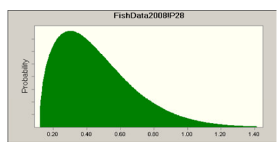


Assumption: Hg concentration in river bream/roach in Samarkand and Rostovka

Weibull distribution with parameters:

Location	0.12
Scale	0.38
Shape	1.49

Selected range is from 0.00 to 2.00

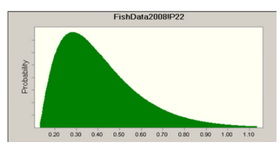


Assumption: Hg concentration in river bream/roach as a whole

Gamma distribution with parameters:

Location	0.13
Scale	0.13
Shape	2.20

Selected range is from 0.00 to 2.00

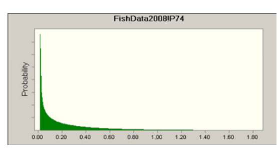


Assumption: Hg concentration in river carp as a whole

Gamma distribution with parameters:

Location	0.02
Scale	0.46
Shape	0.51

Selected range is from 0.00 to 2.00

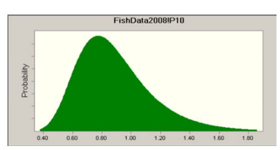


Assumption: Hg concentration in river perch in Samarkand and Rostovka

Maximum Extreme distribution with parameters:

Likeliest	0.77
Scale	0.21

Selected range is from 0.00 to 2.00

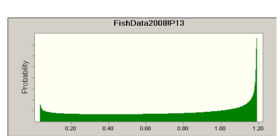


Assumption: Hg concentration in river perch in whole study areas

Beta distribution with parameters:

Minimum	0.03
Maximum	1.19
Alpha	0.81
Beta	0.58

Selected range is from 0.00 to 2.00



APPENDIX G: Distributions and curves of intake rate and intake frequency of food for Monte Carlo simulations

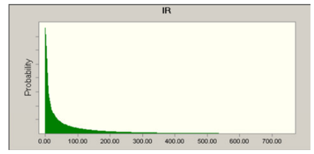
Table G- 1 Distributions and curves of intake rate (g/meal) of river pike/zander in different residential areas

Assumption: Intake rate of river pike/zander in Temirtau old town

Gamma distribution with parameters:

Location	-1.00
Scale	248.96
Shape	0.25

Selected range is from 0.00 to 800.00

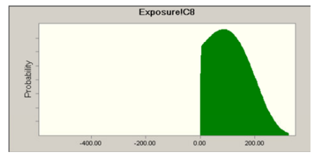


Assumption: Intake rate of river pike/zander in Temirtau new town

Minimum Extreme distribution with parameters:

Likeliest	85.40
Scale	123.84

Selected range is from 0.00 to 800.00

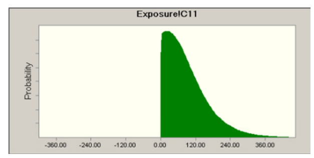


Assumption: Intake rate of river pike/zander in Chkalovo

Logistic distribution with parameters:

Mean	21.30
Scale	60.81

Selected range is from 0.00 to 800.00

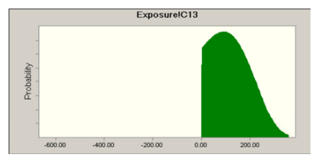


Assumption: Intake rate of river pike/zander in Gagarinskoye

Minimum Extreme distribution with parameters:

Likeliest	91.98
Scale	139.15

Selected range is from 0.00 to 800.00

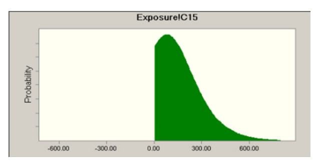


Assumption: Intake rate of river pike/zander in Samarkand

Logistic distribution with parameters:

Mean	79.49
Scale	111.41

Selected range is from 0.00 to 800.00

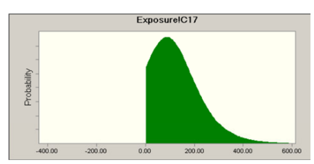


Assumption: Intake rate of river pike/zander in Rostovka

Logistic distribution with parameters:

Mean	87.94
Scale	72.16

Selected range is from 0.00 to 800.00



Assumption: Intake rate of river pike/zander in whole study areas

Logistic distribution with parameters:

Mean	28.59
Scale	62.34

Selected range is from 0.00 to 800.00

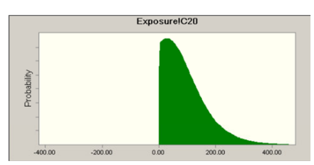


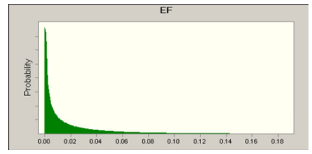
Table G- 2 Distributions and curves of intake frequency (meals/day) of river pike/zander in different residential areas

Assumption: Intake frequency of river pike/zander in Temirtau old town

Gamma distribution with parameters:

Location	0.00
Scale	0.06
Shape	0.26

Selected range is from 0.00 to 1.50

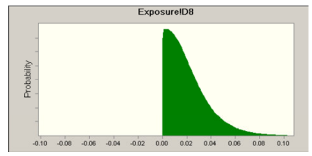


Assumption: Intake frequency of river pike/zander in Temirtau new town

Logistic distribution with parameters:

Mean	0.00
Scale	0.01

Selected range is from 0.00 to 1.50

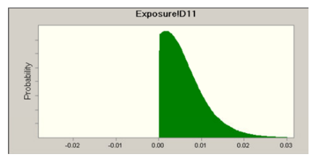


Assumption: Intake frequency of river pike/zander in Chkalovo

Logistic distribution with parameters:

Mean	0.00
Scale	0.00

Selected range is from 0.00 to 1.50

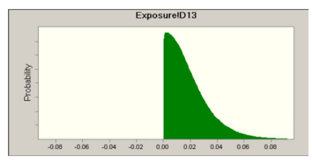


Assumption: Intake frequency of river pike/zander in Gagarinskoye

Logistic distribution with parameters:

Mean	0.00
Scale	0.01

Selected range is from 0.00 to 1.50

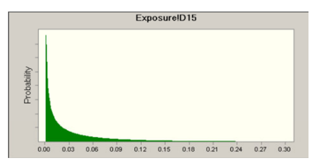


Assumption: Intake frequency of river pike/zander in Samarkand

Gamma distribution with parameters:

Location	0.00
Scale	0.10
Shape	0.28

Selected range is from 0.00 to 1.50

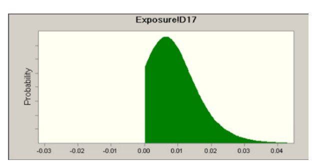


Assumption: Intake frequency of river pike/zander in Rostovka

Logistic distribution with parameters:

Mean	0.01
Scale	0.01

Selected range is from 0.00 to 1.50



Assumption: Intake frequency of river pike/zander in whole study areas

Gamma distribution with parameters:

Location	0.00
Scale	0.04
Shape	0.32

Selected range is from 0.00 to 1.50

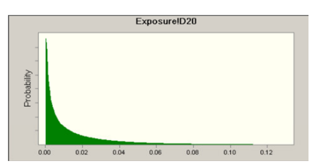


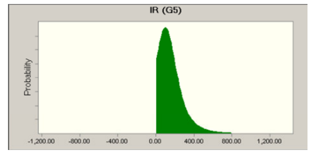
Table G- 3 Distributions and curves of intake rate (g/meal) of market pike/zander in different residential areas

Assumption: Intake rate of market pike/zander in Temirtau old town

Student's t distribution with parameters:

Midpoint	100.00
Scale	121.53
Deg. Freedom	2.95

Selected range is from 0.00 to 800.00

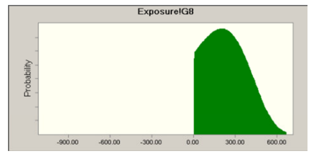


Assumption: Intake rate of market pike/zander in Temirtau new town

Minimum Extreme distribution with parameters:

Likeliest	203.76
Scale	241.51

Selected range is from 0.00 to 800.00

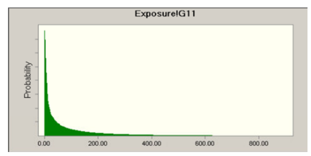


Assumption: Intake rate of market pike/zander in Chkalovo

Gamma distribution with parameters:

Location	-0.88
Scale	279.14
Shape	0.30

Selected range is from 0.00 to 800.00

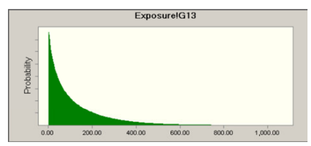


Assumption: Intake rate of market pike/zander in Gagarinskoye

Beta distribution with parameters:

Minimum	-17.54
Maximum	2,137.39
Alpha	0.58
Beta	11.16

Selected range is from 0.00 to 800.00

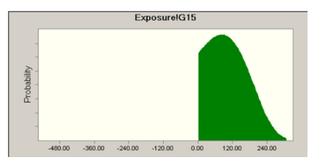


Assumption: Intake rate of market pike/zander in Samarkand

Minimum Extreme distribution with parameters:

Likeliest	82.57
Scale	116.30

Selected range is from 0.00 to 800.00

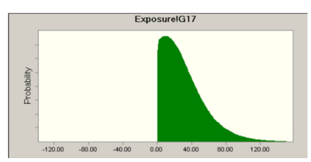


Assumption: Intake rate of market pike/zander in Rostovka

Logistic distribution with parameters:

Mean	9.57
Scale	20.42

Selected range is from 0.00 to 800.00



Assumption: Intake rate of market pike/zander in whole study areas

Beta distribution with parameters:

Minimum	-10.30
Maximum	3,240.00
Alpha	0.38
Beta	12.75

Selected range is from 0.00 to 800.00

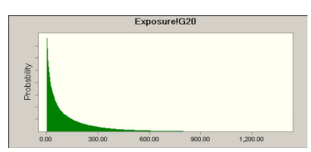


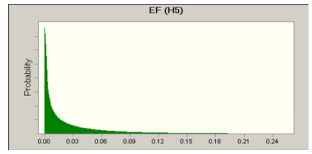
Table G- 4 Distributions and curves of intake frequency (meals/day) of market pike/zander in different residential areas

Assumption: Intake frequency of market pike/zander in Temirtau old town

Gamma distribution with parameters:

Location	0.00
Scale	0.08
Shape	0.34

Selected range is from 0.00 to 1.50

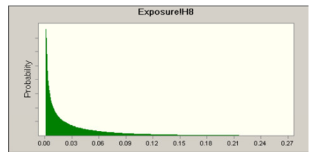


Assumption: Intake frequency of market pike/zander in Temirtau new town

Gamma distribution with parameters:

Location	0.00
Scale	0.08
Shape	0.32

Selected range is from 0.00 to 1.50

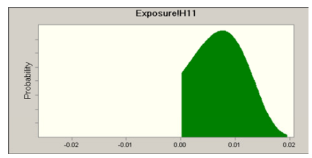


Assumption: Intake frequency of market pike/zander in Chkalovo

Minimum Extreme distribution with parameters:

Likeliest	0.01
Scale	0.01

Selected range is from 0.00 to 1.50

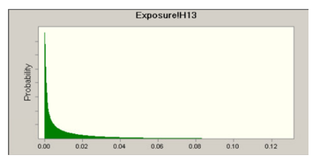


Assumption: Intake frequency of market pike/zander in Gagarinskoye

Gamma distribution with parameters:

Location	0.00
Scale	0.04
Shape	0.28

Selected range is from 0.00 to 1.50

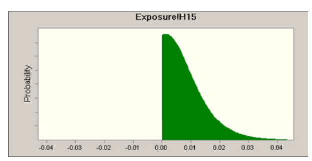


Assumption: Intake frequency of market pike/zander in Samarkand

Logistic distribution with parameters:

Mean	0.00
Scale	0.01

Selected range is from 0.00 to 1.50

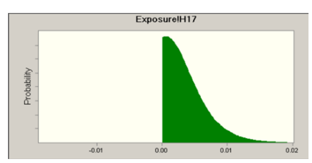


Assumption: Intake frequency of market pike/zander in Rostovka

Logistic distribution with parameters:

Mean	0.00
Scale	0.00

Selected range is from 0.00 to 1.50



Assumption: Intake frequency of market pike/zander in whole study areas

Gamma distribution with parameters:

Location	0.00
Scale	0.04
Shape	0.36

Selected range is from 0.00 to 1.50

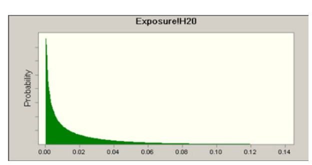


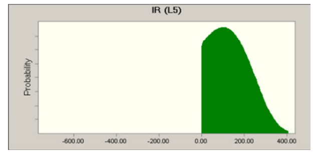
Table G- 5 Distributions and curves of intake rate (g/meal) of river bream/roach in different residential areas

Assumption: Intake rate of river bream/roach in Temirtau old town

Minimum Extreme distribution with parameters:

Likeliest	101.64
Scale	158.41

Selected range is from 0.00 to 800.00

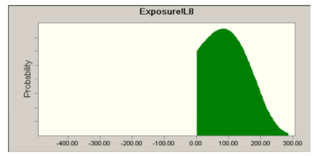


Assumption: Intake rate of river bream/roach in Temirtau new town

Minimum Extreme distribution with parameters:

Likeliest	83.22
Scale	105.06

Selected range is from 0.00 to 800.00

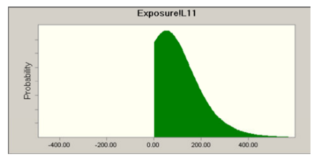


Assumption: Intake rate of river bream/roach in Chkalovo

Logistic distribution with parameters:

Mean	53.15
Scale	74.95

Selected range is from 0.00 to 800.00

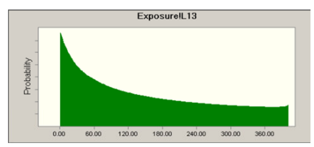


Assumption: Intake rate of river bream/roach in Gagarinskoye

Beta distribution with parameters:

Minimum	-27.53
Maximum	401.20
Alpha	0.37
Beta	0.96

Selected range is from 0.00 to 800.00

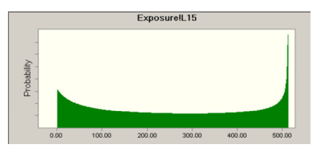


Assumption: Intake rate of river bream/roach in Samarkand

Beta distribution with parameters:

Minimum	-28.14
Maximum	513.09
Alpha	0.45
Beta	0.63

Selected range is from 0.00 to 800.00

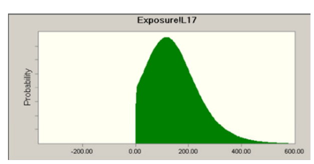


Assumption: Intake rate of river bream/roach in Rostovka

Logistic distribution with parameters:

Mean	116.23
Scale	66.75

Selected range is from 0.00 to 800.00



Assumption: Intake rate of river bream/roach in whole study areas

Beta distribution with parameters:

Minimum	-15.48
Maximum	533.91
Alpha	0.3
Beta	1.31

Selected range is from 0.00 to 800.00

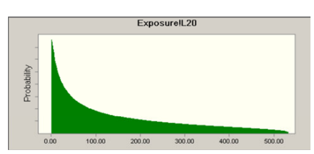


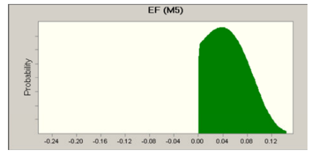
Table G- 6 Distributions and curves of intake frequency (meals/day) of river bream/roach in different residential areas

Assumption: Intake frequency of river bream/roach in Temirtau old town

Minimum Extreme distribution with parameters:

Likeliest	0.04
Scale	0.06

Selected range is from 0.00 to 1.50

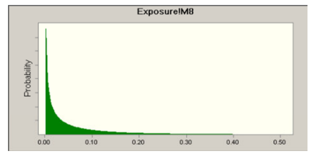


Assumption: Intake frequency of river bream/roach in Temritau new town

Gamma distribution with parameters:

Location	0.00
Scale	0.17
Shape	0.26

Selected range is from 0.00 to 1.50

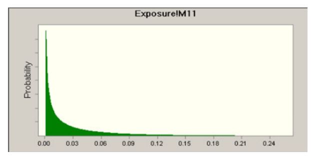


Assumption: Intake frequency of river bream/roach in Chkalovo

Gamma distribution with parameters:

Location	0.00
Scale	0.08
Shape	0.28

Selected range is from 0.00 to 1.50

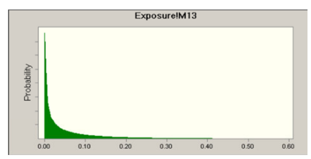


Assumption: Intake frequency of river bream/roach in Gagarinskoye

Gamma distribution with parameters:

Location	0.00
Scale	0.19
Shape	0.29

Selected range is from 0.00 to 1.50

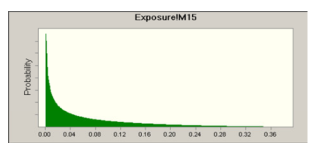


Assumption: Intake frequency of river bream/roach in Samarkand

Beta distribution with parameters:

Minimum	0.00
Maximum	0.40
Alpha	0.38
Beta	2.34

Selected range is from 0.00 to 1.50

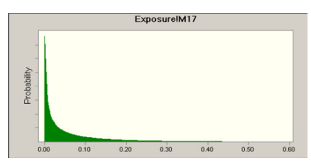


Assumption: Intake frequency of river bream/roach in Rostovka

Gamma distribution with parameters:

Location	0.00
Scale	0.17
Shape	0.34

Selected range is from 0.00 to 1.50



Assumption: Intake frequency of river bream/roach in whole study areas

Gamma distribution with parameters:

Location	0.00
Scale	0.13
Shape	0.31

Selected range is from 0.00 to 1.50

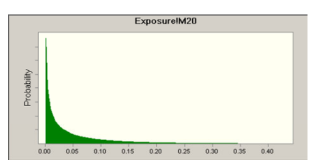


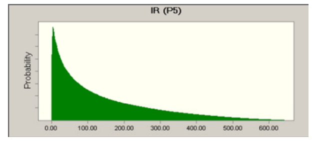
Table G- 7 Distributions and curves of intake rate (g/meal) of market bream/roach in different residential areas

Assumption: Intake rate of market bream/roach in Temritau old town

Beta distribution with parameters:

Minimum	-20.17
Maximum	671.10
Alpha	0.44
Beta	2.27

Selected range is from 0.00 to 800.00

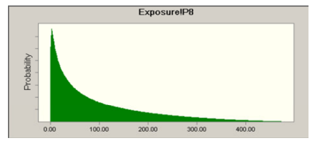


Assumption: Intake rate of market bream/roach in Temritau new town

Beta distribution with parameters:

Minimum	-12.75
Maximum	503.64
Alpha	0.36
Beta	2.31

Selected range is from 0.00 to 800.00

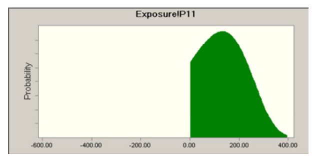


Assumption: Intake rate of market bream/roach in Chkalovo

Minimum Extreme distribution with parameters:

Likeliest	131.87
Scale	136.59

Selected range is from 0.00 to 800.00

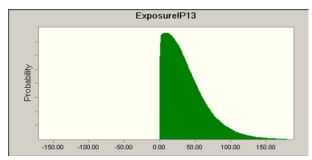


Assumption: Intake rate of market bream/roach in Gagarinskoye

Logistic distribution with parameters:

Mean	8.67
Scale	24.86

Selected range is from 0.00 to 800.00

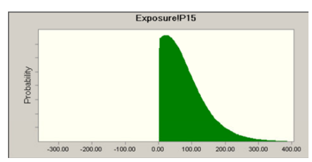


Assumption: Intake rate of market bream/roach in Samarkand

Logistic distribution with parameters:

Mean	21.60
Scale	52.81

Selected range is from 0.00 to 800.00

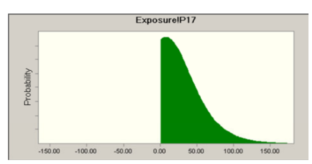


Assumption: Intake rate of market bream/roach in Rostovka

Logistic distribution with parameters:

Mean	7.73
Scale	23.95

Selected range is from 0.00 to 800.00



Assumption: Intake rate of market bream/roach in whole study areas

Logistic distribution with parameters:

Mean	26.72
Scale	50.08

Selected range is from 0.00 to 800.00

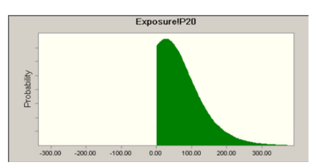


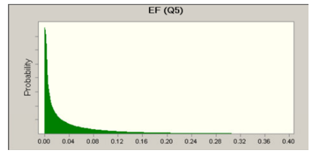
Table G- 8 Distributions and curves of intake frequency (meals/day) of market bream/roach in different residential areas

Assumption: Intake frequency of market bream/roach in Temirtau old town

Gamma distribution with parameters:

Location	0.00
Scale	0.12
Shape	0.29

Selected range is from 0.00 to 1.50

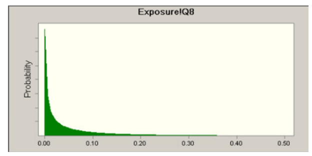


Assumption: Intake frequency of market bream/roach in Temirtau new town

Gamma distribution with parameters:

Location	0.00
Scale	0.16
Shape	0.27

Selected range is from 0.00 to 1.50

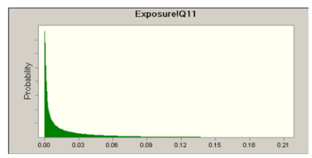


Assumption: Intake frequency of market bream/roach in Chkalovo

Gamma distribution with parameters:

Location	0.00
Scale	0.07
Shape	0.26

Selected range is from 0.00 to 1.50

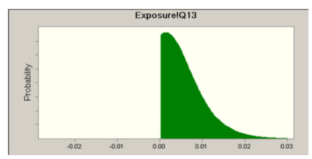


Assumption: Intake frequency of market bream/roach in Gagarinskoye

Logistic distribution with parameters:

Mean	0.00
Scale	0.00

Selected range is from 0.00 to 1.50

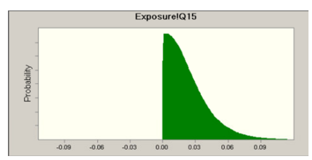


Assumption: Intake frequency of market bream/roach in Samarkand

Logistic distribution with parameters:

Mean	0.00
Scale	0.02

Selected range is from 0.00 to 1.50

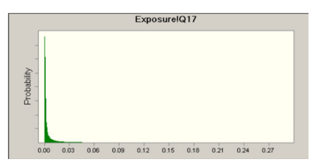


Assumption: Intake frequency of market bream/roach in Rostovka

Weibull distribution with parameters:

Location	0.00
Scale	0.00
Shape	0.31

Selected range is from 0.00 to 1.50



Assumption: Intake frequency of market bream/roach in whole study areas

Gamma distribution with parameters:

Location	0.00
Scale	0.08
Shape	0.29

Selected range is from 0.00 to 1.50

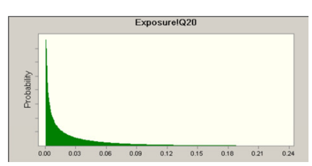


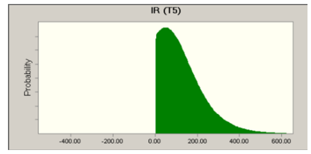
Table G- 9 Distributions and curves of intake rate (g/meal) of river carp in different residential areas

Assumption: Intake rate of river carp in Temirtau old town

Logistic distribution with parameters:

Mean	48.50
Scale	83.18

Selected range is from 0.00 to 800.00

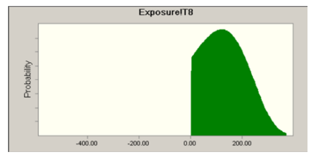


Assumption: Intake rate of river carp in Temirtau new town

Minimum Extreme distribution with parameters:

Likeliest	120.50
Scale	130.16

Selected range is from 0.00 to 800.00

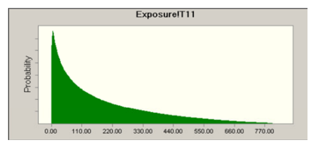


Assumption: Intake rate of river carp in Chkalovo

Beta distribution with parameters:

Minimum	-25.64
Maximum	877.51
Alpha	0.45
Beta	2.36

Selected range is from 0.00 to 800.00

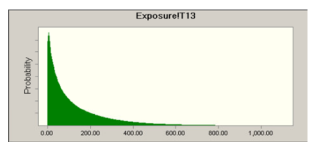


Assumption: Intake rate of river carp in Gagarinskoye

Beta distribution with parameters:

Minimum	-14.81
Maximum	1,838.01
Alpha	0.48
Beta	8.16

Selected range is from 0.00 to 800.00

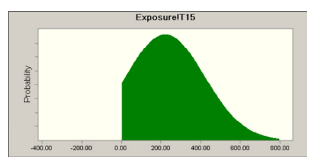


Assumption: Intake rate of river carp in Samarkand

Normal distribution with parameters:

Mean	220.55
Std. Dev.	197.87

Selected range is from 0.00 to 800.00

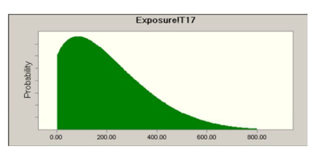


Assumption: Intake rate of river carp in Rostovka

Beta distribution with parameters:

Minimum	-50.15
Maximum	1,140.11
Alpha	1.66
Beta	6.27

Selected range is from 0.00 to 800.00



Assumption: Intake rate of river carp in whole study areas

Normal distribution with parameters:

Mean	118.49
Std. Dev.	164.32

Selected range is from 0.00 to 800.00

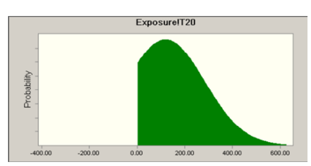


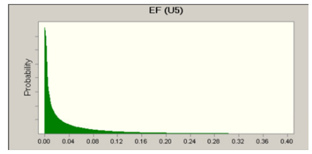
Table G- 10 Distributions and curves of intake frequency (meals/day) of river carp in different residential areas

Assumption: Intake frequency of river carp in Temirtau old town

Gamma distribution with parameters:

Location	0.00
Scale	0.13
Shape	0.27

Selected range is from 0.00 to 1.50

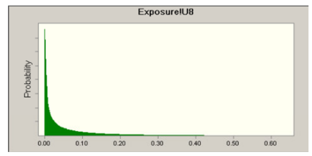


Assumption: Intake frequency of river carp in Temirtau new town

Gamma distribution with parameters:

Location	0.00
Scale	0.21
Shape	0.25

Selected range is from 0.00 to 1.50

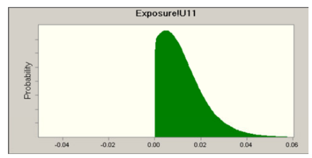


Assumption: Intake frequency of river carp in Chkalovo

Logistic distribution with parameters:

Mean	0.00
Scale	0.01

Selected range is from 0.00 to 1.50

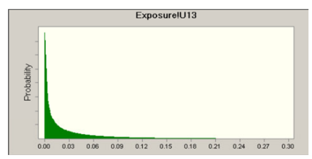


Assumption: Intake frequency of river carp in Gagarinskoye

Gamma distribution with parameters:

Location	0.00
Scale	0.10
Shape	0.28

Selected range is from 0.00 to 1.50

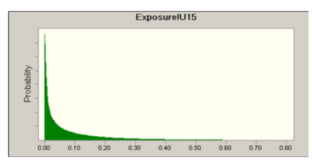


Assumption: Intake frequency of river carp in Samarkand

Gamma distribution with parameters:

Location	0.00
Scale	0.22
Shape	0.39

Selected range is from 0.00 to 1.50

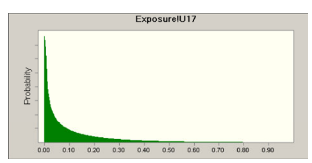


Assumption: Intake frequency of river carp in Rostovka

Gamma distribution with parameters:

Location	0.00
Scale	0.26
Shape	0.43

Selected range is from 0.00 to 1.50



Assumption: Intake frequency of river carp in whole study areas

Gamma distribution with parameters:

Location	0.00
Scale	0.16
Shape	0.33

Selected range is from 0.00 to 1.50

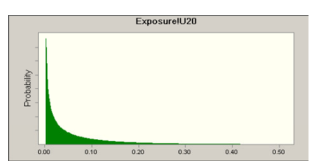


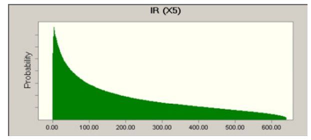
Table G- 11 Distributions and curves of intake rate (g/meal) of market carp in different residential areas

Assumption: Intake rate of market carp in Temirtau old town

Beta distribution with parameters:

Minimum	-22.56
Maximum	642.04
Alpha	0.42
Beta	1.36

Selected range is from 0.00 to 800.00

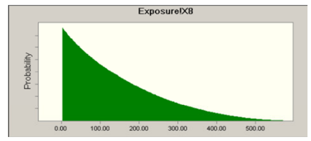


Assumption: Intake rate of market carp in Temirtau new town

Beta distribution with parameters:

Minimum	-44.83
Maximum	619.83
Alpha	0.89
Beta	3.20

Selected range is from 0.00 to 800.00

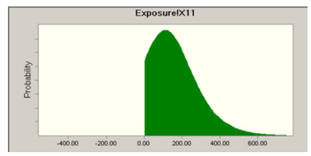


Assumption: Intake rate of market carp in Chkalovo

Logistic distribution with parameters:

Mean	111.31
Scale	92.66

Selected range is from 0.00 to 800.00

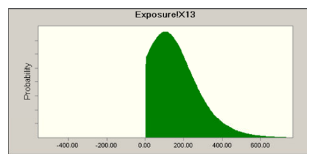


Assumption: Intake ratey of market carp in Gagarinskoye

Logistic distribution with parameters:

Mean	103.79
Scale	91.20

Selected range is from 0.00 to 800.00

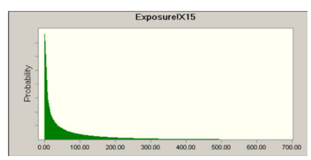


Assumption: Intake rate of market carp in Samarkand

Gamma distribution with parameters:

Location	-0.83
Scale	214.32
Shape	0.29

Selected range is from 0.00 to 800.00

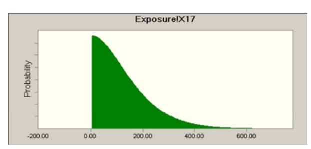


Assumption: Intake rate of market carp in Rostovka

Beta distribution with parameters:

Minimum	-196.21
Maximum	6,218.54
Alpha	4.19
Beta	100

Selected range is from 0.00 to 800.00



Assumption: Intake rate of market carp in whole study areas

Logistic distribution with parameters:

Mean	7.14
Scale	26.62

Selected range is from 0.00 to 800.00

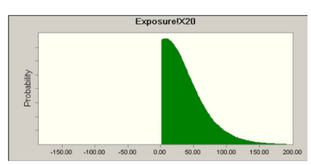


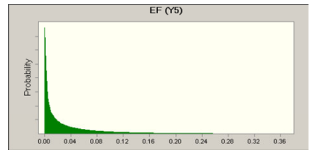
Table G- 12 Distributions and curves of intake frequency (meals/day) of market carp in different residential areas

Assumption: Intake frequency of market carp in Temirtau old town

Gamma distribution with parameters:

Location	0.00
Scale	0.11
Shape	0.31

Selected range is from 0.00 to 1.50

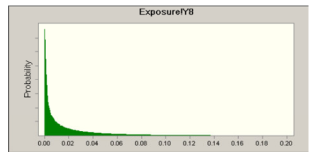


Assumption: Intake frequency of market carp in Temirtau new town

Gamma distribution with parameters:

Location	0.00
Scale	0.06
Shape	0.31

Selected range is from 0.00 to 1.50

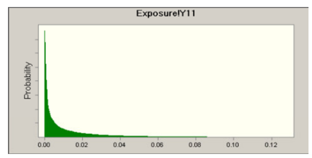


Assumption: Intake frequency of market carp in Chkalovo

Gamma distribution with parameters:

Location	0.00
Scale	0.04
Shape	0.30

Selected range is from 0.00 to 1.50

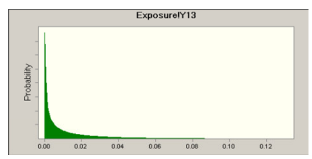


Assumption: Intake frequency of market carp in Gagarinskoye

Gamma distribution with parameters:

Location	0.00
Scale	0.04
Shape	0.29

Selected range is from 0.00 to 1.50

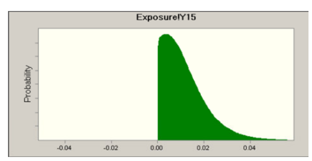


Assumption: Intake frequency of market carp in Samarkand

Logistic distribution with parameters:

Mean	0.00
Scale	0.01

Selected range is from 0.00 to 1.50

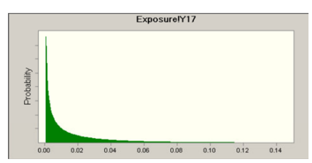


Assumption: Intake frequency of market carp in Rostovka

Gamma distribution with parameters:

Location	0.00
Scale	0.05
Shape	0.27

Selected range is from 0.00 to 1.50



Assumption: Intake frequency of market carp in whole study areas

Minimum Extreme distribution with parameters:

Likeliest	0.01
Scale	0.01

Selected range is from 0.00 to 1.50

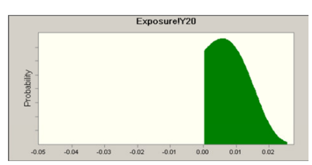


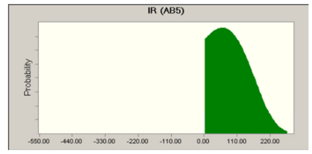
Table G- 13 Distributions and curves of intake rate (g/meal) of river crayfish in different residential areas

Assumption: Intake rate of river crayfish in Temirtau old town

Minimum Extreme distribution with parameters:

Likeliest	60.74
Scale	112.14

Selected range is from 0.00 to 500.00

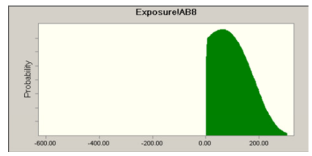


Assumption: Intake rate of river crayfish in Temirtau new town

Minimum Extreme distribution with parameters:

Likeliest	61.65
Scale	126.03

Selected range is from 0.00 to 500.00

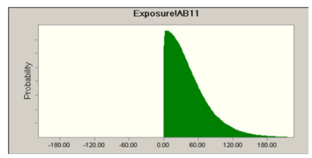


Assumption: Intake rate of river crayfish in Chkalovo

Logistic distribution with parameters:

Mean	3.95
Scale	30.52

Selected range is from 0.00 to 500.00

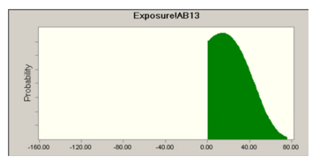


Assumption: Intake rate of river crayfish in Gagarinskoye

Minimum Extreme distribution with parameters:

Likeliest	14.15
Scale	31.96

Selected range is from 0.00 to 500.00

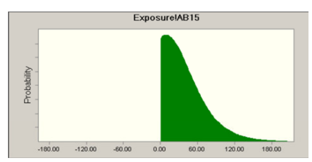


Assumption: Intake rate of river crayfish in Samarkand

Logistic distribution with parameters:

Mean	8.72
Scale	28.45

Selected range is from 0.00 to 500.00

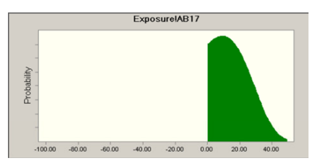


Assumption: Intake rate of river crayfish in Rostovka

Minimum Extreme distribution with parameters:

Likeliest	9.27
Scale	20.83

Selected range is from 0.00 to 500.00



Assumption: Intake rate of river crayfish in whole study areas

Logistic distribution with parameters:

Mean	2.05
Scale	17.49

Selected range is from 0.00 to 500.00

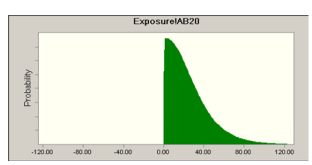


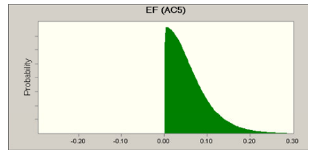
Table G- 14 Distributions and curves of intake frequency (meals/day) of river crayfish in different residential areas

Assumption: Intake frequency of river crayfish in Temirtau old town

Logistic distribution with parameters:

Mean	0.00
Scale	0.04

Selected range is from 0.00 to 1.50

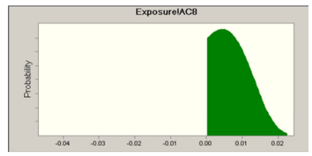


Assumption: Intake frequency of river crayfish in Temirtau new town

Minimum Extreme distribution with parameters:

Likeliest	0.00
Scale	0.01

Selected range is from 0.00 to 1.50

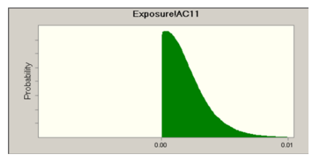


Assumption: Intake frequency of river crayfish in Chkalovo

Logistic distribution with parameters:

Mean	0.00
Scale	0.00

Selected range is from 0.00 to 1.50

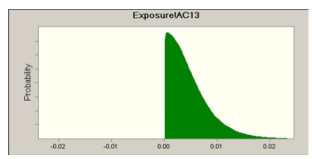


Assumption: Intake frequency of river crayfish in Gagarinskoye

Logistic distribution with parameters:

Mean	0.00
Scale	0.00

Selected range is from 0.00 to 1.50

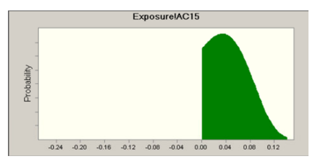


Assumption: Intake frequency of river crayfish in Samarkand

Minimum Extreme distribution with parameters:

Likeliest	0.03
Scale	0.06

Selected range is from 0.00 to 1.50

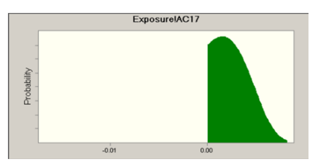


Assumption: Intake frequency of river crayfish in Rostovka

Minimum Extreme distribution with parameters:

Likeliest	0.00
Scale	0.00

Selected range is from 0.00 to 1.50



Assumption: Intake frequency of river crayfish in whole study areas

Logistic distribution with parameters:

Mean	0.00
Scale	0.01

Selected range is from 0.00 to 1.50

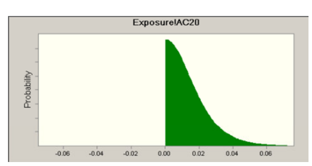


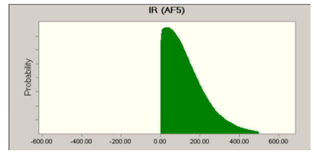
Table G- 15 Distributions and curves of intake rate (g/meal) of market crayfish in different residential areas

Assumption: Intake rate of market crayfish in Temirtau old town

Logistic distribution with parameters:

Mean	32.00
Scale	89.74

Selected range is from 0.00 to 500.00

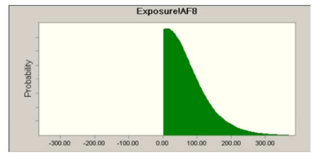


Assumption: Intake rate of market crayfish in Temirtau new town

Logistic distribution with parameters:

Mean	11.34
Scale	51.78

Selected range is from 0.00 to 500.00

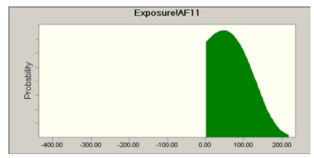


Assumption: Intake rate of market crayfish in Chkalovo

Minimum Extreme distribution with parameters:

Likeliest	47.02
Scale	88.41

Selected range is from 0.00 to 500.00



Assumption: Intake rate of market crayfish in whole study areas

Logistic distribution with parameters:

Mean	4.18
Scale	33.73

Selected range is from 0.00 to 500.00

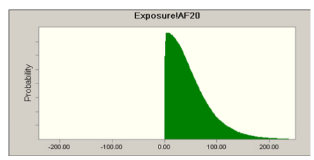


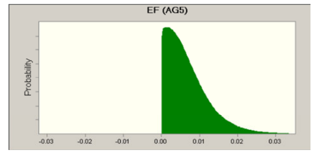
Table G- 16 Distributions and curves of intake frequency (meals/day) of market crayfish in different residential areas

Assumption: Intake frequency of market crayfish in Temirtau old town

Logistic distribution with parameters:

Mean	0.00
Scale	0.00

Selected range is from 0.00 to 1.50

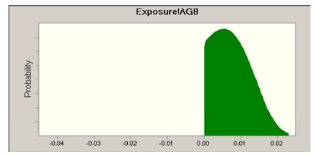


Assumption: Intake frequency of market crayfish in Temirtau new town

Minimum Extreme distribution with parameters:

Likeliest	0.01
Scale	0.01

Selected range is from 0.00 to 1.50

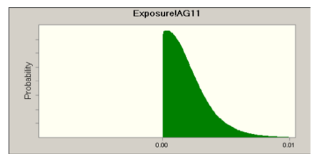


Assumption: Intake frequency of market crayfish in Chkalovo

Logistic distribution with parameters:

Mean	0.00
Scale	0.00

Selected range is from 0.00 to 1.50



Assumption: Intake frequency of market crayfish in whole study areas

Normal distribution with parameters:

Mean	0.00
Std. Dev.	0.01

Selected range is from 0.00 to 1.50

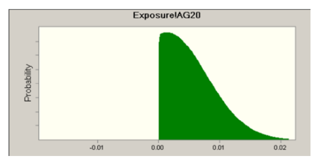


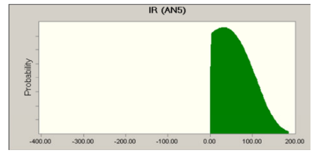
Table G- 17 Distributions and curves of intake rate (g/meal) of market shellfish in different residential areas

Assumption: Intake rate of market shellfish in Temirtau old town

Minimum Extreme distribution with parameters:

Likeliest	31.62
Scale	79.90

Selected range is from 0.00 to 500.00

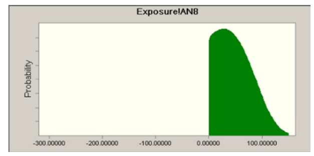


Assumption: Intake rate of market shellfish in Temirtau new town

Minimum Extreme distribution with parameters:

Likeliest	27.69
Scale	63.39

Selected range is from 0.00000 to 500.00000



Assumption: Intake rate of market shellfish in whole study areas

Minimum Extreme distribution with parameters:

Likeliest	19.62
Scale	61.64

Selected range is from 0.00 to 500.00

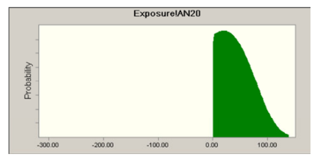


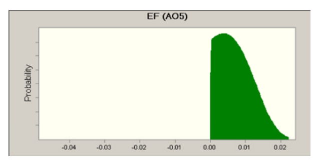
Table G- 18 Distributions and curves of intake frequency (meals/day) of market shellfish in different residential areas

Assumption: Intake frequency of market shellfish in Temirtau old town

Minimum Extreme distribution with parameters:

Likeliest	0.00
Scale	0.01

Selected range is from 0.00 to 1.50

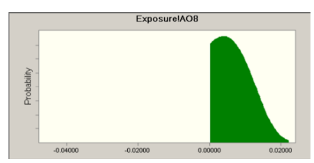


Assumption: Intake frequency of market shellfish in Temirtau new town

Minimum Extreme distribution with parameters:

Likeliest	0.00
Scale	0.01

Selected range is from 0.00000 to 1.50000



Assumption: Intake frequency of market shellfish in whole study areas

Logistic distribution with parameters:

Mean	0.00
Scale	0.00

Selected range is from 0.00 to 1.50

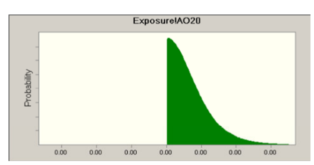


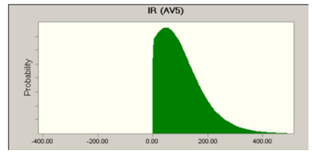
Table G- 19 Distributions and curves of intake rate (g/meal) of market herring in different residential areas

Assumption: Intake rate of market herring in Temirtau old town

Logistic distribution with parameters:

Mean	47.48
Scale	64.10

Selected range is from 0.00 to 800.00

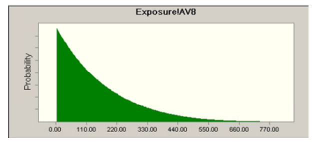


Assumption: Intake rate of market herring in Temirtau new town

Beta distribution with parameters:

Minimum	-40.71
Maximum	1,090.78
Alpha	0.96
Beta	6.16

Selected range is from 0.00 to 800.00

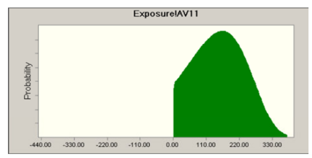


Assumption: Intake rate of market herring in Chkalovo

Minimum Extreme distribution with parameters:

Likeliest	163.09
Scale	111.80

Selected range is from 0.00 to 800.00

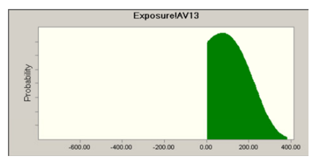


Assumption: Intake rate of market herring in Gagarinskoye

Minimum Extreme distribution with parameters:

Likeliest	74.34
Scale	159.23

Selected range is from 0.00 to 800.00

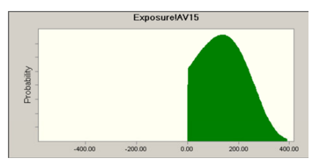


Assumption: Intake rate of market herring in Samarkand

Minimum Extreme distribution with parameters:

Likeliest	136.85
Scale	131.66

Selected range is from 0.00 to 800.00

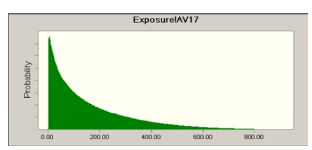


Assumption: Intake rate of market herring in Rostovka

Beta distribution with parameters:

Minimum	-15.66
Maximum	1,064.69
Alpha	0.63
Beta	4.05

Selected range is from 0.00 to 800.00



Assumption: Intake rate of market herring in whole study areas

Beta distribution with parameters:

Minimum	-28.13
Maximum	889.68
Alpha	0.58
Beta	4.20

Selected range is from 0.00 to 800.00

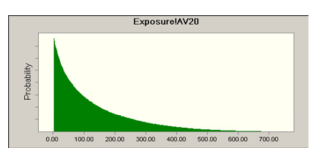


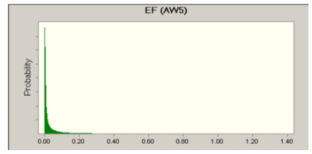
Table G- 20 Distributions and curves of intake frequency (meals/day) of market herring in different residential areas

Assumption: Intake frequency of market herring in Temirtau old town

Weibull distribution with parameters:

Location	0.00
Scale	0.01
Shape	0.36

Selected range is from 0.00 to 1.50

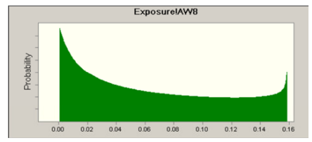


Assumption: Intake frequency of market herring in Temirtau new town

Beta distribution with parameters:

Minimum	-0.01
Maximum	0.16
Alpha	0.36
Beta	0.82

Selected range is from 0.00 to 1.50

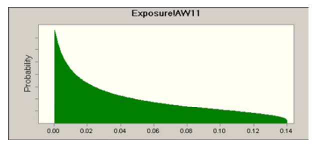


Assumption: Intake frequency of market herring in Chkalovo

Beta distribution with parameters:

Minimum	-0.01
Maximum	0.14
Alpha	0.44
Beta	1.27

Selected range is from 0.00 to 1.50

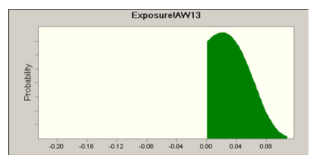


Assumption: Intake frequency of market herring in Gagarinskoye

Minimum Extreme distribution with parameters:

Likeliest	0.02
Scale	0.04

Selected range is from 0.00 to 1.50

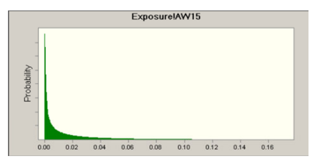


Assumption: Intake frequency of market herring in Samarkand

Gamma distribution with parameters:

Location	0.00
Scale	0.06
Shape	0.26

Selected range is from 0.00 to 1.50

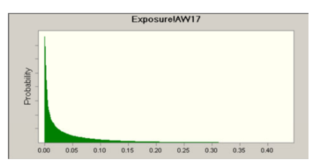


Assumption: Intake frequency of market herring in Rostovka

Gamma distribution with parameters:

Location	0.00
Scale	0.13
Shape	0.37

Selected range is from 0.00 to 1.50



Assumption: Intake frequency of market herring in whole study areas

Gamma distribution with parameters:

Location	0.00
Scale	0.08
Shape	0.38

Selected range is from 0.00 to 1.50

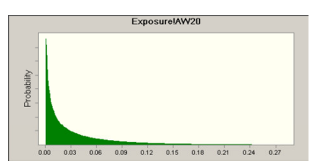


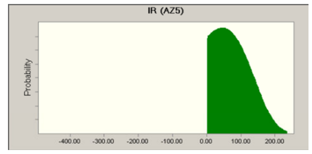
Table G- 21 Distributions and curves of intake rate (g/meal) of river perch in different residential areas

Assumption: Intake rate of river perch in Temritau old town

Minimum Extreme distribution with parameters:

Likeliest	45.87
Scale	98.40

Selected range is from 0.00 to 800.00

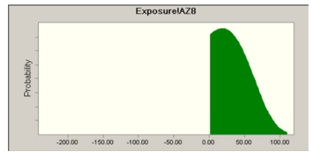


Assumption: Intake rate of river perch in Temritau new town

Minimum Extreme distribution with parameters:

Likeliest	18.58
Scale	47.52

Selected range is from 0.00 to 800.00

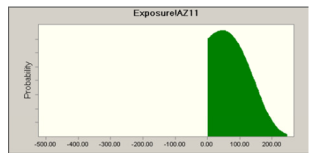


Assumption: Intake rate of river perch in Chkalovo

Minimum Extreme distribution with parameters:

Likeliest	46.35
Scale	104.17

Selected range is from 0.00 to 800.00

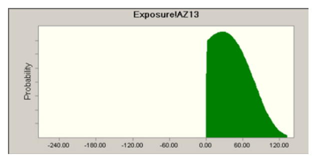


Assumption: Intake rate of river perch in Gagarinskoye

Minimum Extreme distribution with parameters:

Likeliest	26.58
Scale	55.00

Selected range is from 0.00 to 800.00

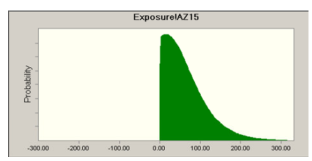


Assumption: Intake rate of river perch in Samarkand

Logistic distribution with parameters:

Mean	15.00
Scale	43.56

Selected range is from 0.00 to 800.00

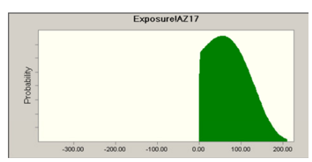


Assumption: Intake rate of river perch in Rostovka

Minimum Extreme distribution with parameters:

Likeliest	55.70
Scale	80.44

Selected range is from 0.00 to 800.00



Assumption: Intake rate of river perch in whole study areas

Minimum Extreme distribution with parameters:

Likeliest	47.77
Scale	99.09

Selected range is from 0.00 to 800.00

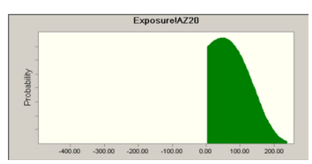


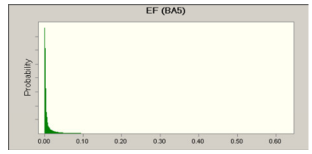
Table G- 22 Distributions and curves of intake frequency (meals/day) of river perch in different residential areas

Assumption: Intake frequency of river perch in Temirtau old town

Weibull distribution with parameters:

Location	0.00
Scale	0.00
Shape	0.28

Selected range is from 0.00 to 1.50

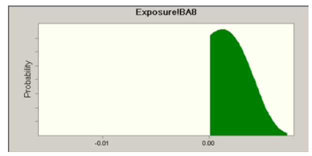


Assumption: Intake frequency of river perch in Temirtau new town

Minimum Extreme distribution with parameters:

Likeliest	0.00
Scale	0.00

Selected range is from 0.00 to 1.50

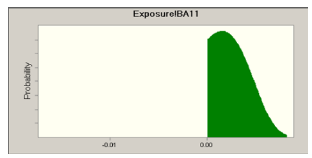


Assumption: Intake frequency of river perch in Chkalovo

Minimum Extreme distribution with parameters:

Likeliest	0.00
Scale	0.00

Selected range is from 0.00 to 1.50

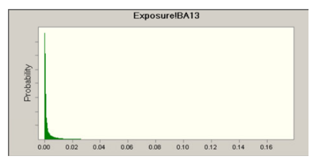


Assumption: Intake frequency of river perch in Gagarinskoye

Weibull distribution with parameters:

Location	0.00
Scale	0.00
Shape	0.28

Selected range is from 0.00 to 1.50

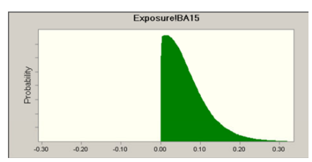


Assumption: Intake frequency of river perch in Samarkand

Logistic distribution with parameters:

Mean	0.01
Scale	0.04

Selected range is from 0.00 to 1.50

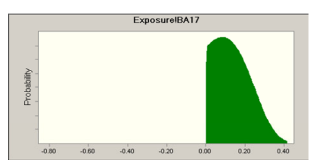


Assumption: Intake frequency of river perch in Rostovka

Minimum Extreme distribution with parameters:

Likeliest	0.08
Scale	0.17

Selected range is from 0.00 to 1.50



Assumption: Intake frequency of river perch in whole study areas

Logistic distribution with parameters:

Mean	0.00
Scale	0.02

Selected range is from 0.00 to 1.50

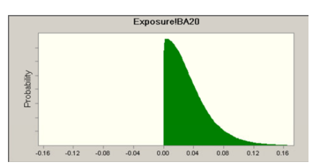


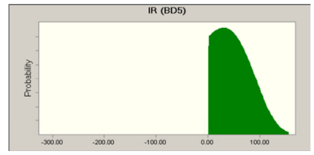
Table G- 23 Distributions and curves of intake rate (g/meal) of market perch in different residential areas

Assumption: Intake rate of market perch in Temirtau old town

Minimum Extreme distribution with parameters:

Likeliest	30.04
Scale	65.13

Selected range is from 0.00 to 800.00

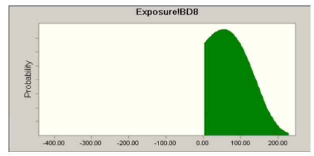


Assumption: Intake rate of market perch in Temirtau new town

Minimum Extreme distribution with parameters:

Likeliest	54.03
Scale	90.48

Selected range is from 0.00 to 800.00

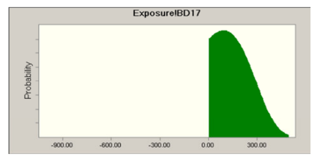


Assumption: Intake rate of market perch in Rostovka

Minimum Extreme distribution with parameters:

Likeliest	92.70
Scale	208.34

Selected range is from 0.00 to 800.00



Assumption: Intake rate of market perch in whole study areas

Minimum Extreme distribution with parameters:

Likeliest	46.08
Scale	141.34

Selected range is from 0.00 to 800.00

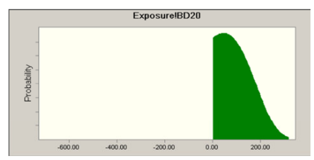


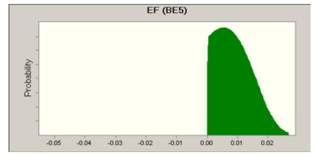
Table G- 24 Distributions and curves of intake frequency (meals/day) of market perch in different residential areas

Assumption: Intake frequency of market perch in Temirtau old town

Minimum Extreme distribution with parameters:

Likeliest	0.01
Scale	0.01

Selected range is from 0.00 to 1.50

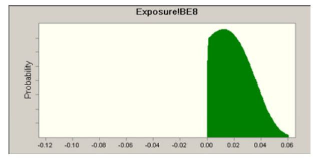


Assumption: Intake frequency of market perch in Temirtau new town

Minimum Extreme distribution with parameters:

Likeliest	0.01
Scale	0.03

Selected range is from 0.00 to 1.50

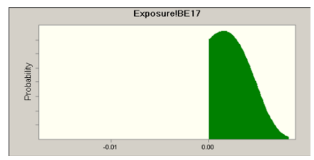


Assumption: Intake frequency of market perch in Rostovka

Minimum Extreme distribution with parameters:

Likeliest	0.00
Scale	0.00

Selected range is from 0.00 to 1.50



Assumption: Intake frequency of market perch in whole study areas

Minimum Extreme distribution with parameters:

Likeliest	0.01
Scale	0.02

Selected range is from 0.00 to 1.50

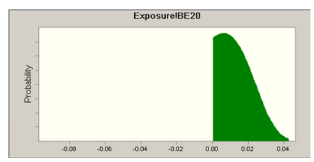


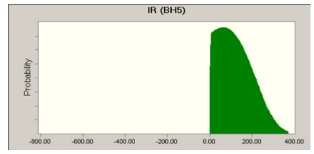
Table G- 25 Distributions and curves of intake rate (g/meal) of other river fish in different residential areas

Assumption: Intake rate of other river fish in Temirtau old town

Minimum Extreme distribution with parameters:

Likeliest	63.25
Scale	159.80

Selected range is from 0.00 to 800.00

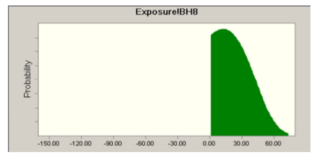


Assumption: Intake rate of other river fish in Temirtau new town

Minimum Extreme distribution with parameters:

Likeliest	12.27
Scale	31.54

Selected range is from 0.00 to 800.00

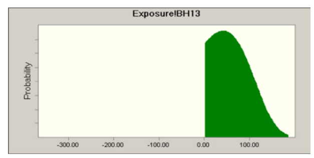


Assumption: Intake rate of other river fish in Gagarinskoye

Minimum Extreme distribution with parameters:

Likeliest	41.70
Scale	74.59

Selected range is from 0.00 to 800.00

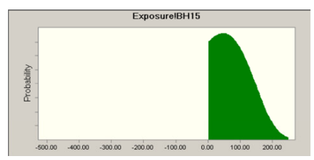


Assumption: Intake rate of other river fish in Samarkand

Minimum Extreme distribution with parameters:

Likeliest	47.16
Scale	104.90

Selected range is from 0.00 to 800.00

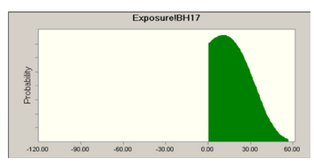


Assumption: Intake rate of rother river fish in Rostovka

Minimum Extreme distribution with parameters:

Likeliest	10.62
Scale	23.87

Selected range is from 0.00 to 800.00



Assumption: Intake rate of other river fish in whole study areas

Logistic distribution with parameters:

Mean	0.52
Scale	9.60

Selected range is from 0.00 to 800.00

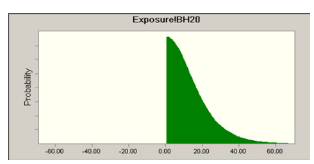


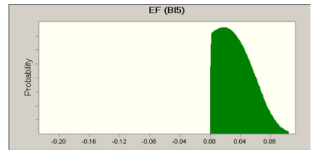
Table G- 26 Distributions and curves of intake frequency (meals/day) of other river fish in different residential areas

Assumption: Intake frequency of other river fish in Temirtau old town

Minimum Extreme distribution with parameters:

Likeliest	0.02
Scale	0.04

Selected range is from 0.00 to 1.50

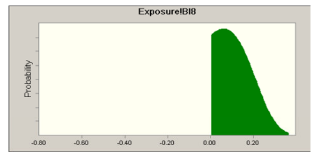


Assumption: Intake frequency of other river fish in Temirtau new town

Minimum Extreme distribution with parameters:

Likeliest	0.06
Scale	0.16

Selected range is from 0.00 to 1.50

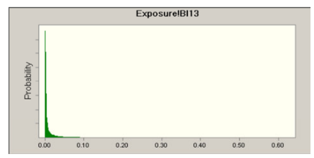


Assumption: Intake frequency of other river fish in Gagarinskoye

Weibull distribution with parameters:

Location	0.00
Scale	0.00
Shape	0.28

Selected range is from 0.00 to 1.50

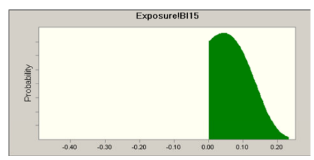


Assumption: Intake frequency of other river fish in Samarkand

Minimum Extreme distribution with parameters:

Likeliest	0.04
Scale	0.10

Selected range is from 0.00 to 1.50

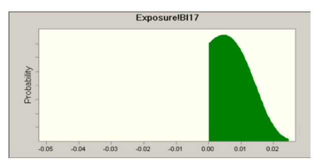


Assumption: Intake frequency of other river fish in Rostovka

Minimum Extreme distribution with parameters:

Likeliest	0.00
Scale	0.01

Selected range is from 0.00 to 1.50



Assumption: Intake frequency of other river fish in whole study areas

Logistic distribution with parameters:

Mean	0.00
Scale	0.01

Selected range is from 0.00 to 1.50

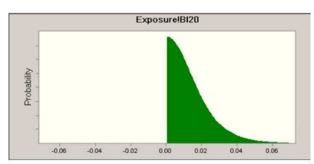


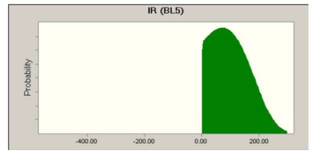
Table G- 27 Distributions and curves of intake rate (g/meal) of other market fish in different residential areas

Assumption: Intake rate of other market fish in Temirtau old town

Minimum Extreme distribution with parameters:

Likeliest	71.50
Scale	117.19

Selected range is from 0.00 to 800.00

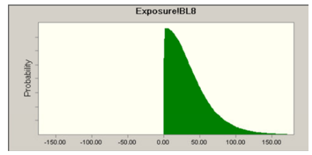


Assumption: Intake rate of other market fish in Temirtau new town

Logistic distribution with parameters:

Mean	2.91
Scale	24.43

Selected range is from 0.00 to 800.00

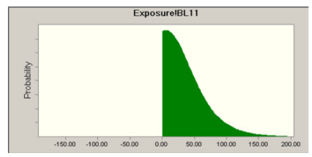


Assumption: Intake rate of other market fish in Chkalovo

Logistic distribution with parameters:

Mean	5.76
Scale	27.31

Selected range is from 0.00 to 800.00

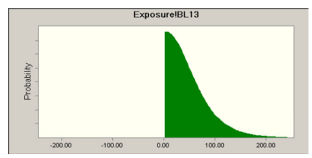


Assumption: Intake rate of other market fish in Gagarinskoye

Logistic distribution with parameters:

Mean	3.89
Scale	34.41

Selected range is from 0.00 to 800.00

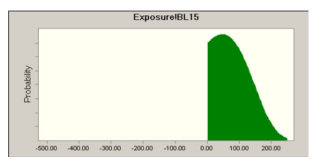


Assumption: Intake rate of other market fish in Samarkand

Minimum Extreme distribution with parameters:

Likeliest	47.16
Scale	104.90

Selected range is from 0.00 to 800.00

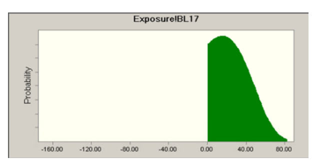


Assumption: Intake rate of other market fish in Rostovka

Minimum Extreme distribution with parameters:

Likeliest	15.45
Scale	34.72

Selected range is from 0.00 to 800.00



Assumption: Intake rate of other market fish in whole study areas

Minimum Extreme distribution with parameters:

Likeliest	69.04
Scale	164.64

Selected range is from 0.00 to 800.00

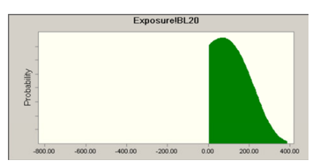


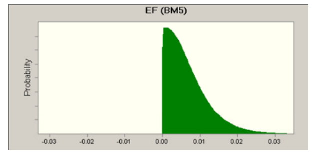
Table G- 28 Distributions and curves of intake frequency (meals/day) of other market fish in different residential areas

Assumption: Intake frequency of other market fish in Temirtau old town

Logistic distribution with parameters:

Mean	0.00
Scale	0.00

Selected range is from 0.00 to 1.50

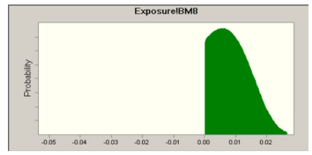


Assumption: Intake frequency of other market fish in Temirtau new town

Minimum Extreme distribution with parameters:

Likeliest	0.01
Scale	0.01

Selected range is from 0.00 to 1.50

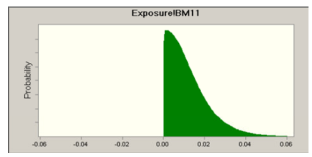


Assumption: Intake frequency of other market fish in Chkalovo

Logistic distribution with parameters:

Mean	0.00
Scale	0.01

Selected range is from 0.00 to 1.50

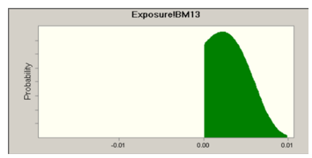


Assumption: Intake frequency of other market fish in Gagarinskoye

Minimum Extreme distribution with parameters:

Likeliest	0.00
Scale	0.00

Selected range is from 0.00 to 1.50

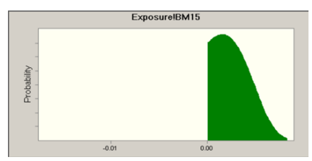


Assumption: Intake frequency of other market fish in Samarkand

Minimum Extreme distribution with parameters:

Likeliest	0.00
Scale	0.00

Selected range is from 0.00 to 1.50

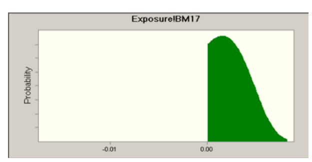


Assumption: Intake frequency of other market fish in Rostovka

Minimum Extreme distribution with parameters:

Likeliest	0.00
Scale	0.00

Selected range is from 0.00 to 1.50



Assumption: Intake frequency of other market fish in whole study areas

Logistic distribution with parameters:

Mean	0.00
Scale	0.00

Selected range is from 0.00 to 1.50

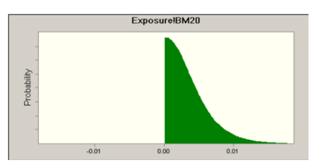


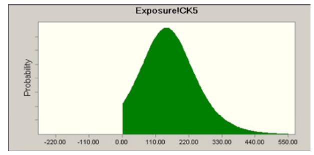
Table G- 29 Distributions and curves of intake rate (g/meal) of beef in different residential areas

Assumption: Intake rate of beef in Temirtau old town

Logistic distribution with parameters:

Mean	146.53
Scale	58.45

Selected range is from 0.00 to 1,000.00

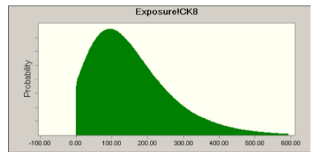


Assumption: Intake rate of beef in Temirtau new town

Maximum Extreme distribution with parameters:

Likeliest	93.68
Scale	94.39

Selected range is from 0.00 to 1,000.00

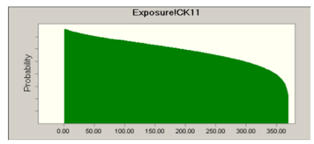


Assumption: Intake rate of beef in Chkalovo

Beta distribution with parameters:

Minimum	-32.53
Maximum	369.65
Alpha	0.94
Beta	1.18

Selected range is from 0.00 to 1,000.00

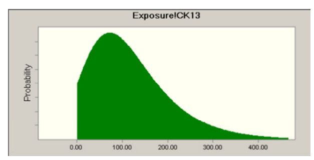


Assumption: Intake rate of beef in Gagarinskoye

Maximum Extreme distribution with parameters:

Likeliest	72.38
Scale	74.34

Selected range is from 0.00 to 1,000.00

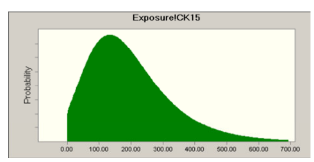


Assumption: Intake rate of beef in Samarkand

Maximum Extreme distribution with parameters:

Likeliest	132.82
Scale	105.75

Selected range is from 0.00 to 1,000.00

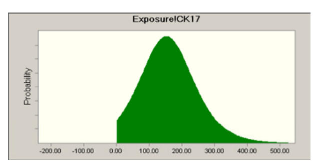


Assumption: Intake rate of beef in Rostovka

Logistic distribution with parameters:

Mean	152.83
Scale	54.01

Selected range is from 0.00 to 1,000.00



Assumption: Intake rate of beef in whole study areas

Logistic distribution with parameters:

Mean	139.71
Scale	64.52

Selected range is from 0.00 to 1,000.00

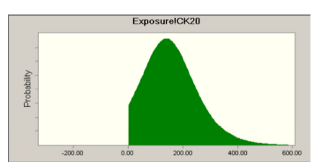


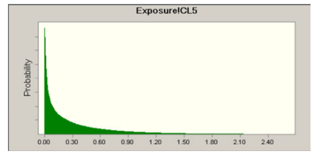
Table G- 30 Distributions and curves of intake frequency (meals/day) of beef in different residential areas

Assumption: Intake frequency of beef in Temirtau old town

Gamma distribution with parameters:

Location	0.00
Scale	0.64
Shape	0.54

Selected range is from 0.00 to 3.00

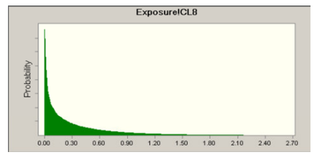


Assumption: Intake frequency of beef in Temirtau new town

Gamma distribution with parameters:

Location	0.00
Scale	0.65
Shape	0.54

Selected range is from 0.00 to 3.00

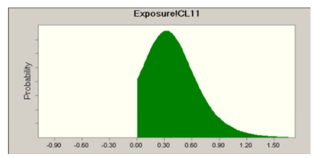


Assumption: Intake frequency of beef in Chkalovo

Logistic distribution with parameters:

Mean	0.32
Scale	0.19

Selected range is from 0.00 to 3.00

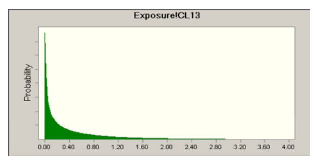


Assumption: Intake frequency of beef in Gagarinskoye

Gamma distribution with parameters:

Location	0.00
Scale	1.08
Shape	0.43

Selected range is from 0.00 to 3.00

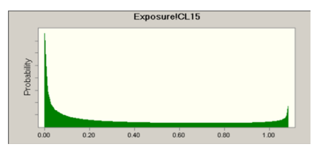


Assumption: Intake frequency of beef in Samarkand

Beta distribution with parameters:

Minimum	0.00
Maximum	1.08
Alpha	0.34
Beta	0.66

Selected range is from 0.00 to 3.00

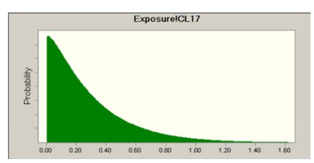


Assumption: Intake frequency of beef in Rostovka

Weibull distribution with parameters:

Location	-0.01
Scale	0.30
Shape	1.07

Selected range is from 0.00 to 3.00



Assumption: Intake frequency of beef in whole study areas

Gamma distribution with parameters:

Location	0.00
Scale	0.64
Shape	0.56

Selected range is from 0.00 to 3.00

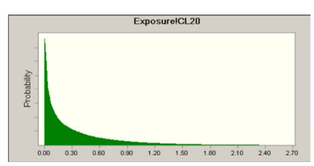


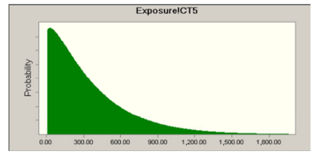
Table G- 31 Distributions and curves of intake rate (g/meal) of dairy in different residential areas

Assumption: Intake rate of dairy in Temirtau old town

Weibull distribution with parameters:

Location	-12.13
Scale	372.60
Shape	1.10

Selected range is from 0.00 to 3,000.00

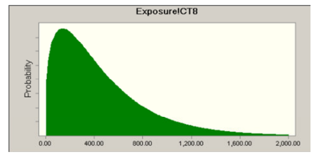


Assumption: Intake rate of dairy in Temirtau new town

Gamma distribution with parameters:

Location	-6.25
Scale	317.23
Shape	1.46

Selected range is from 0.00 to 3,000.00

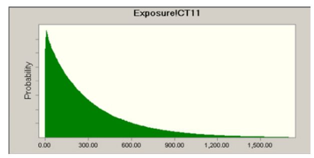


Assumption: Intake rate of dairy in Chkalovo

Gamma distribution with parameters:

Location	-1.88
Scale	327.35
Shape	0.95

Selected range is from 0.00 to 3,000.00

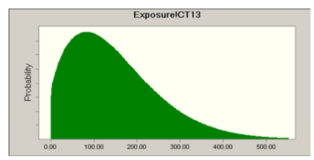


Assumption: Intake rate of dairy in Gagarinskoye

Weibull distribution with parameters:

Location	-14.83
Scale	180.27
Shape	1.59

Selected range is from 0.00 to 3,000.00

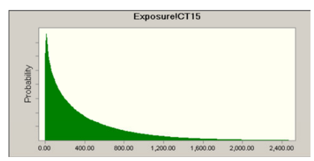


Assumption: Intake rate of dairy in Samarkand

Gamma distribution with parameters:

Location	-3.75
Scale	521.03
Shape	0.78

Selected range is from 0.00 to 3,000.00

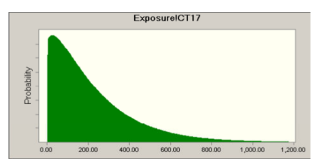


Assumption: Intake rate of dairy in Rostovka

Weibull distribution with parameters:

Location	-11.10
Scale	236.28
Shape	1.13

Selected range is from 0.00 to 3,000.00



Assumption: Intake rate of dairy in whole study areas

Gamma distribution with parameters:

Location	-3.75
Scale	332.76
Shape	0.96

Selected range is from 0.00 to 3,000.00

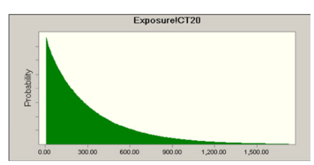


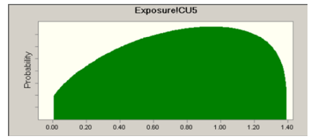
Table G- 32 Distributions and curves of intake frequency (meals/day) of dairy in different residential areas

Assumption: Intake frequency of dairy in Temirtau old town

Beta distribution with parameters:

Minimum	-0.05
Maximum	1.39
Alpha	1.59
Beta	1.26

Selected range is from 0.00 to 3.00

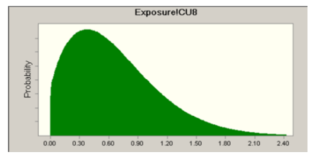


Assumption: Intake frequency of dairy in Temirtau new town

Weibull distribution with parameters:

Location	-0.07
Scale	0.80
Shape	1.61

Selected range is from 0.00 to 3.00

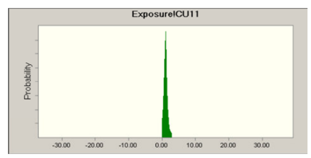


Assumption: Intake frequency of dairy in Chkalovo

Student's t distribution with parameters:

Midpoint	1.00
Scale	0.36
Deg. Freedom	1.12

Selected range is from 0.00 to 3.00

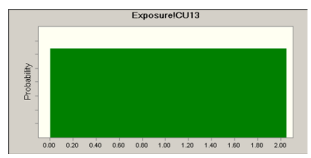


Assumption: Intake frequency of dairy in Gagarinskoye

Uniform distribution with parameters:

Minimum	-0.05
Maximum	2.05

Selected range is from 0.00 to 3.00

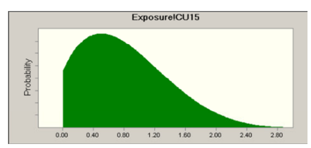


Assumption: Intake frequency of dairy in Samarkand

Beta distribution with parameters:

Minimum	-0.25
Maximum	3.31
Alpha	2.07
Beta	5.02

Selected range is from 0.00 to 3.00

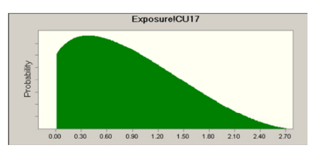


Assumption: Intake frequency of dairy in Rostovka

Beta distribution with parameters:

Minimum	-0.14
Maximum	2.80
Alpha	1.38
Beta	2.80

Selected range is from 0.00 to 3.00



Assumption: Intake frequency of dairy in whole study areas

Beta distribution with parameters:

Minimum	-0.16
Maximum	3.31
Alpha	2.03
Beta	4.72

Selected range is from 0.00 to 3.00

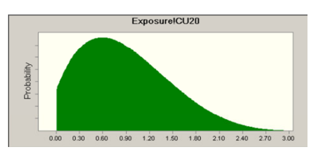


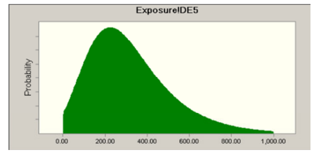
Table G- 33 Distributions and curves of intake rate (g/meal) of tomatoes in different residential areas

Assumption: Intake rate of tomatoes in Temirtau old town

Maximum Extreme distribution with parameters:

Likeliest	223.84
Scale	160.06

Selected range is from 0.00 to 1,000.00

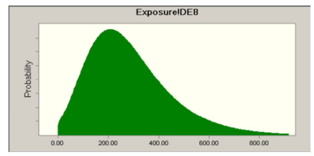


Assumption: Intake rate of tomatoes in Temirtau new town

Maximum Extreme distribution with parameters:

Likeliest	207.02
Scale	134.02

Selected range is from 0.00 to 1,000.00

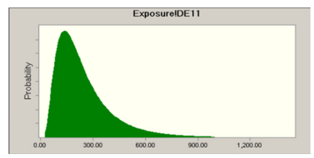


Assumption: Intake rate of tomatoes in Chkalovo

Lognormal distribution with parameters:

Mean	249.07
Std. Dev.	172.84

Selected range is from 0.00 to 1,000.00

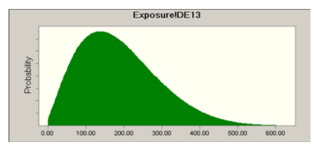


Assumption: Intake rate of tomatoes in Gagarinskoye

Beta distribution with parameters:

Minimum	-13.08
Maximum	832.31
Alpha	2.58
Beta	8.32

Selected range is from 0.00 to 1,000.00

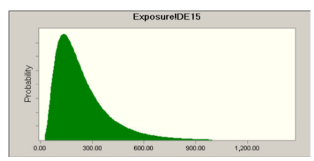


Assumption: Intake rate of tomatoes in Samarkand

Lognormal distribution with parameters:

Mean	244.93
Std. Dev.	174.05

Selected range is from 0.00 to 1,000.00

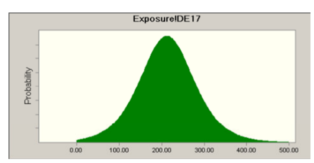


Assumption: Intake rate of tomatoes in Rostovka

Logistic distribution with parameters:

Mean	212.18
Scale	41.50

Selected range is from 0.00 to 1,000.00



Assumption: Intake rate of tomatoes in whole study areas

Maximum Extreme distribution with parameters:

Likeliest	182.49
Scale	116.15

Selected range is from 0.00 to 1,000.00

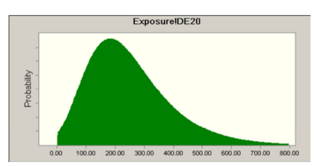


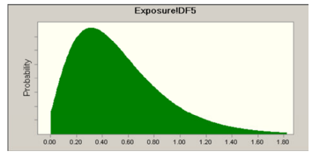
Table G- 34 Distributions and curves of intake frequency (meals/day) of tomatoes in different residential areas

Assumption: Intake frequency of tomatoes in Temirtau old town

Gamma distribution with parameters:

Location	-0.07
Scale	0.21
Shape	2.78

Selected range is from 0.00 to 3.00

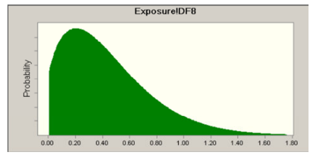


Assumption: Intake frequency of tomatoes in Temirtau new town

Weibull distribution with parameters:

Location	-0.03
Scale	0.52
Shape	1.47

Selected range is from 0.00 to 3.00

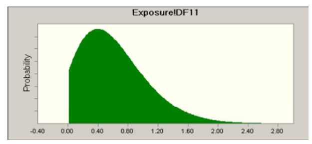


Assumption: Intake frequency of tomatoes in Chkalovo

Beta distribution with parameters:

Minimum	-0.38
Maximum	7.29
Alpha	3.86
Beta	26.51

Selected range is from 0.00 to 3.00

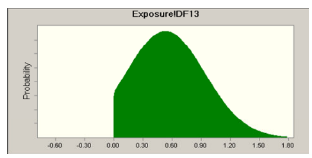


Assumption: Intake frequency of tomatoes in Gagarinskoye

Normal distribution with parameters:

Mean	0.53
Std. Dev.	0.41

Selected range is from 0.00 to 3.00

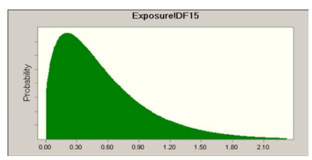


Assumption: Intake frequency of tomatoes in Samarkand

Gamma distribution with parameters:

Location	-0.02
Scale	0.35
Shape	1.64

Selected range is from 0.00 to 3.00

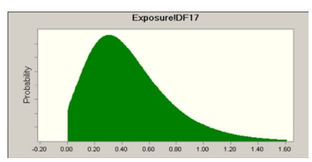


Assumption: Intake frequency of tomatoes in Rostovka

Maximum Extreme distribution with parameters:

Likeliest	0.30
Scale	0.25

Selected range is from 0.00 to 3.00



Assumption: Intake frequency of tomatoes in whole study areas

Gamma distribution with parameters:

Location	-0.03
Scale	0.31
Shape	1.73

Selected range is from 0.00 to 3.00

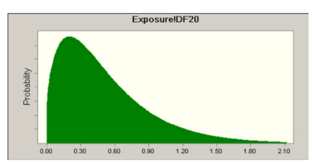


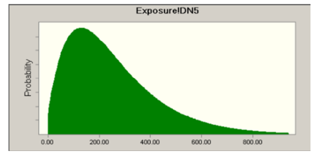
Table G- 35 Distributions and curves of intake rate (g/meal) of cucumbers in different residential areas

Assumption: Intake rate of cucumbers in Temirtau old town

Gamma distribution with parameters:

Location	-15.00
Scale	123.32
Shape	2.16

Selected range is from 0.00 to 1,000.00

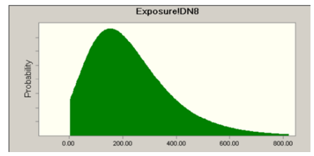


Assumption: Intake rate of cucumbers in Temirtau new town

Maximum Extreme distribution with parameters:

Likeliest	152.53
Scale	126.12

Selected range is from 0.00 to 1,000.00

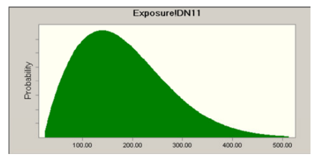


Assumption: Intake rate of cucumbers in Chkalovo

Weibull distribution with parameters:

Location	24.17
Scale	178.48
Shape	1.82

Selected range is from 0.00 to 1,000.00

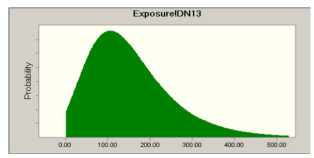


Assumption: Intake rate of cucumbers in Gagarinskoye

Maximum Extreme distribution with parameters:

Likeliest	105.51
Scale	79.94

Selected range is from 0.00 to 1,000.00

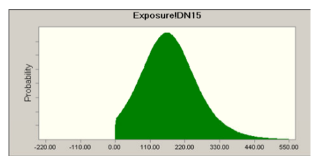


Assumption: Intake rate of cucumbers in Samarkand

Logistic distribution with parameters:

Mean	162.86
Scale	55.90

Selected range is from 0.00 to 1,000.00

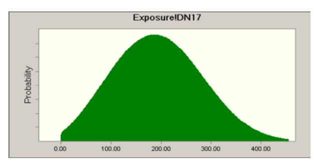


Assumption: Intake rate of cucumbers in Rostovka

Weibull distribution with parameters:

Location	-43.30
Scale	265.56
Shape	2.90

Selected range is from 0.00 to 1,000.00



Assumption: Intake rate of cucumbers in whole study areas

Maximum Extreme distribution with parameters:

Likeliest	138.84
Scale	100.62

Selected range is from 0.00 to 1,000.00

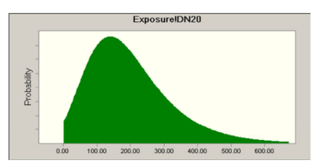


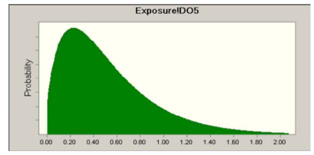
Table G- 36 Distributions and curves of intake frequency (meals/day) of cucumbers in different residential areas

Assumption: Intake frequency of cucumbers in Temirtau old town

Gamma distribution with parameters:

Location	-0.02
Scale	0.29
Shape	1.83

Selected range is from 0.00 to 3.00

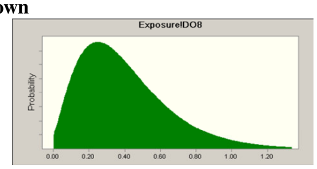


Assumption: Intake frequency of cucumbers in Temirtau new town

Gamma distribution with parameters:

Location	-0.04
Scale	0.15
Shape	2.91

Selected range is from 0.00 to 3.00

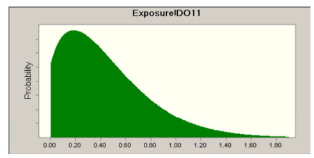


Assumption: Intake frequency of cucumbers in Chkalovo

Weibull distribution with parameters:

Location	-0.05
Scale	0.54
Shape	1.42

Selected range is from 0.00 to 3.00

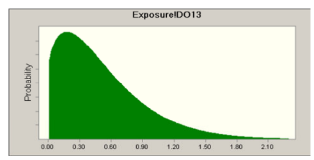


Assumption: Intake frequency of cucumbers in Gagarinskoye

Weibull distribution with parameters:

Location	-0.03
Scale	0.59
Shape	1.33

Selected range is from 0.00 to 3.00

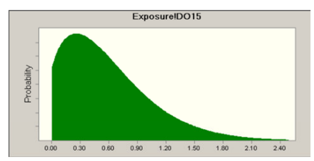


Assumption: Intake frequency of cucumbers in Samarkand

Weibull distribution with parameters:

Location	-0.07
Scale	0.73
Shape	1.46

Selected range is from 0.00 to 3.00

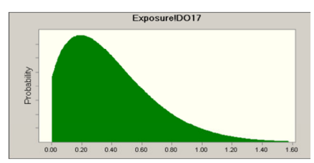


Assumption: Intake frequency of cucumbers in Rostovka

Weibull distribution with parameters:

Location	-0.05
Scale	0.49
Shape	1.51

Selected range is from 0.00 to 3.00

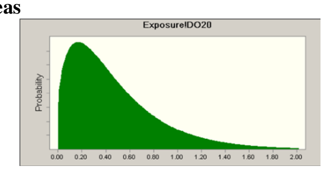


Assumption: Intake frequency of cucumbers in whole study areas

Gamma distribution with parameters:

Location	-0.02
Scale	0.31
Shape	1.61

Selected range is from 0.00 to 3.00



APPENDIX H: Distributions and curves of probabilistic simulation results in the six residential areas

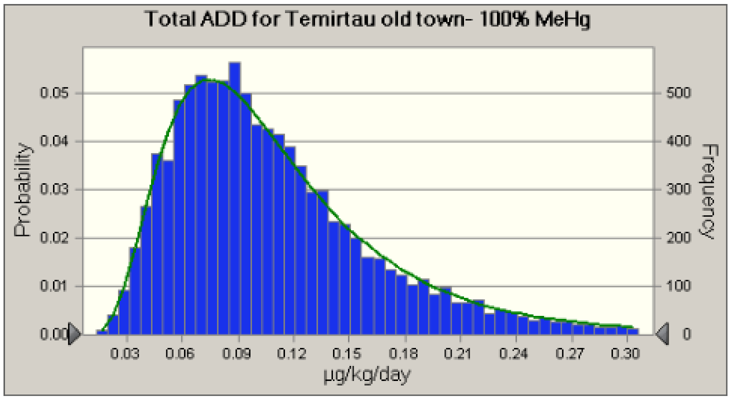


Figure H- 1 Distributions and curves of ADDs with the assumption of total Hg as MeHg in Temirtau old town

Table H- 1 Statistical data of simulated ADDs with the assumption of total Hg as MeHg in Temirtau old town

Statistics	Forecast values	Percentiles	Forecast values	Percentiles	Forecast values
Distribution	Lognormal	10%	0.05	60%	0.11
Mean	0.11	20%	0.06	70%	0.13
SD	0.07	30%	0.08	80%	0.15
Minimum	0.01	40%	0.09	90%	0.19
Maximum	1.44	50%	0.10	100%	1.44

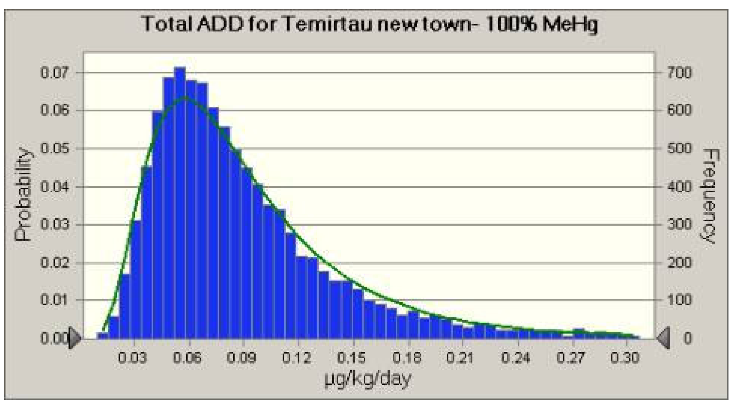


Figure H- 2 Distributions and curves of ADDs with the assumption of total Hg as MeHg in Temirtau new town

Table H- 2 Statistical data of simulated ADDs with the assumption of total Hg as MeHg in Temirtau new town

Statistics	Forecast values	Percentiles	Forecast values	Percentiles	Forecast values
Distribution	Lognormal	10%	0.04	60%	0.09
Mean	0.09	20%	0.05	70%	0.10
SD	0.08	30%	0.06	80%	0.12
Minimum	0.01	40%	0.07	90%	0.16
Maximum	2.72	50%	0.08	100%	2.72

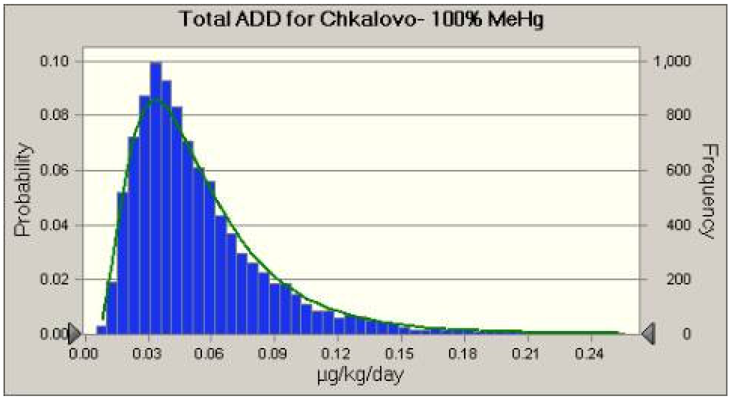


Figure H- 3 Distributions and curves of ADDs with the assumption of total Hg as MeHg in Chkalovo

Table H- 3 Statistical data of simulated ADDs with the assumption of total Hg as MeHg in Chkalovo

Statistics	Forecast values	Percentiles	Forecast values	Percentiles	Forecast values
Distribution	Lognormal	10%	0.02	60%	0.05
Mean	0.06	20%	0.03	70%	0.06
SD	0.07	30%	0.03	80%	0.08
Minimum	0.01	40%	0.04	90%	0.10
Maximum	4.18	50%	0.05	100%	4.18

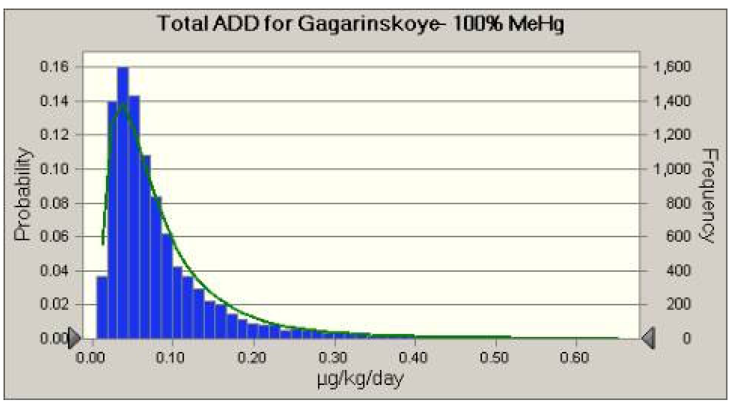


Figure H- 4 Distributions and curves of ADDs with the assumption of total Hg as MeHg in Gagarinskoye

Table H- 4 Statistical data of simulated ADDs with the assumption of total Hg as MeHg in Gagarinskoye

Statistics	Forecast values	Percentiles	Forecast values	Percentiles	Forecast values
Distribution	Lognormal	10%	0.03	60%	0.07
Mean	0.10	20%	0.03	70%	0.09
SD	0.20	30%	0.04	80%	0.12
Minimum	0.00	40%	0.05	90%	0.18
Maximum	9.36	50%	0.06	100%	9.36

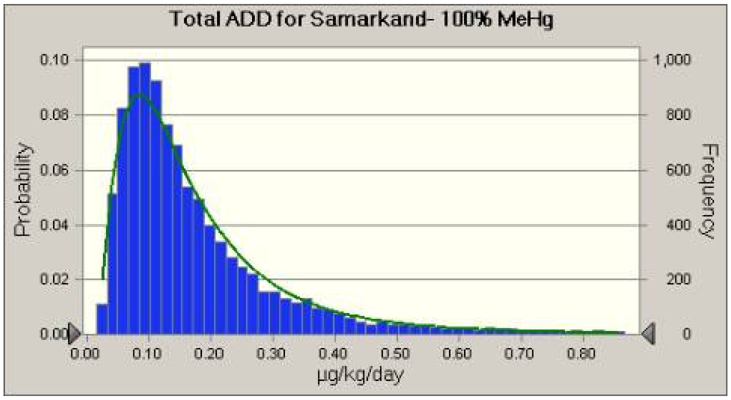


Figure H- 5 Distributions and curves of ADDs with the assumption of total Hg as MeHg in Samarkand

Table H- 5 Statistical data of simulated ADDs with the assumption of total Hg as MeHg in Samarkand

Statistics	Forecast values	Percentiles	Forecast values	Percentiles	Forecast values
Distribution	Lognormal	10%	0.06	60%	0.16
Mean	0.19	20%	0.08	70%	0.20
SD	0.24	30%	0.09	80%	0.25
Minimum	0.01	40%	0.11	90%	0.36
Maximum	12.24	50%	0.13	100%	12.24

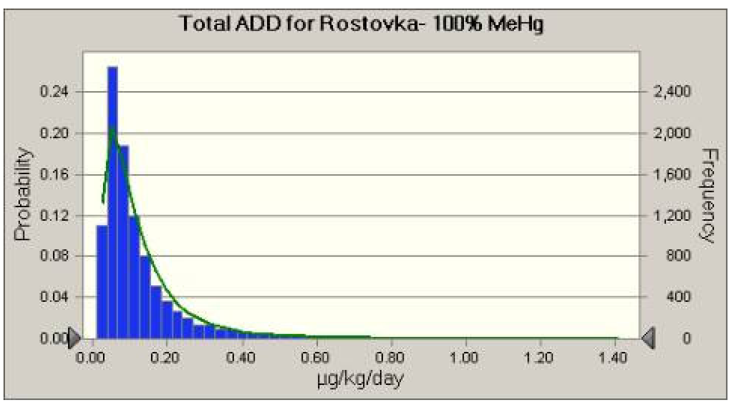


Figure H- 6 Distributions and curves of ADDs with the assumption of total Hg as MeHg in Rostovka

Table H- 6 Statistical data of simulated ADDs with the assumption of total Hg as MeHg in Rostovka

Statistics	Forecast values	Percentiles	Forecast values	Percentiles	Forecast values
Distribution	Lognormal	10%	0.04	60%	0.10
Mean	0.14	20%	0.05	70%	0.13
SD	0.46	30%	0.06	80%	0.17
Minimum	0.01	40%	0.07	90%	0.27
Maximum	41.53	50%	0.08	100%	41.53

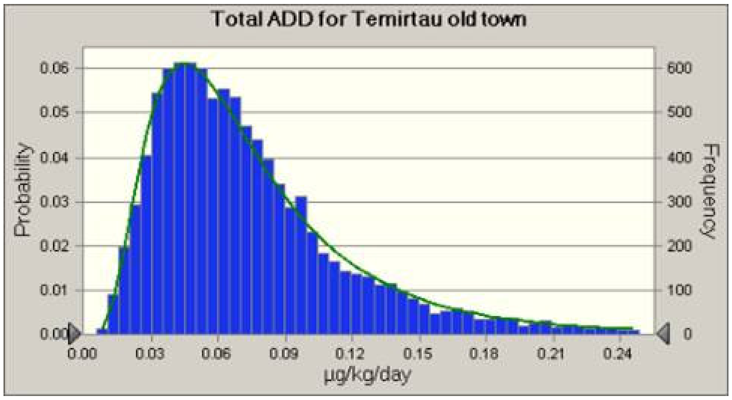


Figure H- 7 Distributions and curves of ADDs with the assumption of different proportions of MeHg in exposure media in Temirtau old town

Table H- 7 Statistical data of simulated ADDs with the assumption of different proportions of MeHg in exposure media in Temirtau old town

Statistics	Forecast values	Percentiles	Forecast values	Percentiles	Forecast values
Distribution	Lognormal	10%	0.03	60%	0.07
Mean	0.08	20%	0.04	70%	0.09
SD	0.06	30%	0.05	80%	0.10
Minimum	0.01	40%	0.06	90%	0.14
Maximum	2.51	50%	0.06	100%	2.51

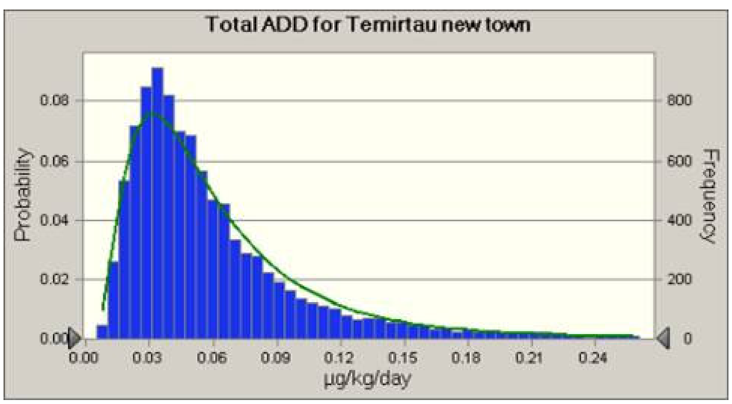


Figure H- 8 Distributions and curves of ADDs with the assumption of different proportions of MeHg in exposure media in Temirtau new town

Table H- 8 Statistical data of simulated ADDs with the assumption of of different proportions of MeHg in exposure media in Temirtau new town

Statistics	Forecast values	Percentiles	Forecast values	Percentiles	Forecast values
Distribution	Lognormal	10%	0.02	60%	0.06
Mean	0.07	20%	0.03	70%	0.07
SD	0.07	30%	0.03	80%	0.09
Minimum	0.00	40%	0.04	90%	0.12
Maximum	1.68	50%	0.05	100%	1.68

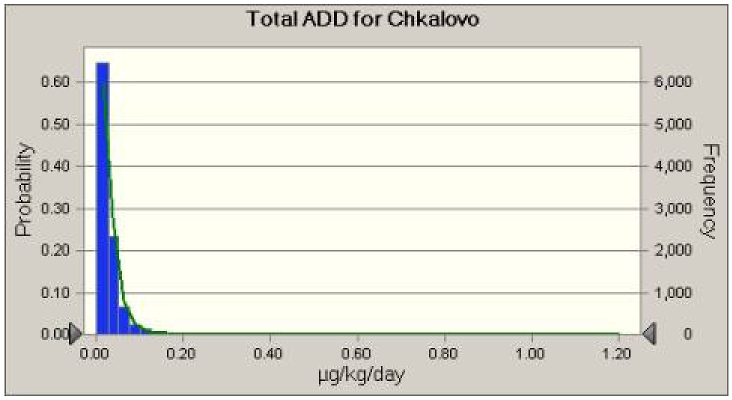


Figure H- 9 Distributions and curves of ADDs with the assumption of different proportions of MeHg in exposure media in Chkalovo

Table H- 9 Statistical data of simulated ADDs with the assumption of of different proportions of MeHg in exposure media in Chkalovo

Statistics	Forecast values	Percentiles	Forecast values	Percentiles	Forecast values
Distribution	Lognormal	10%	0.01	60%	0.02
Mean	0.04	20%	0.01	70%	0.03
SD	0.42	30%	0.02	80%	0.04
Minimum	0.00	40%	0.02	90%	0.06
Maximum	29.78	50%	0.02	100%	29.78

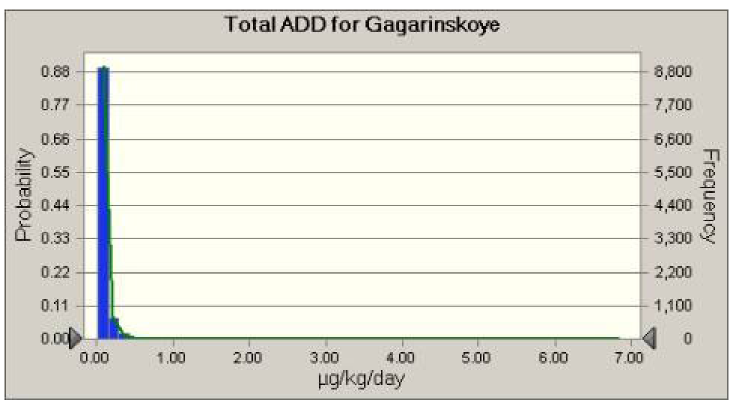


Figure H- 10 Distributions and curves of ADDs with the assumption of different proportions of MeHg in exposure media in Gagarinskoye

Table H- 10 Statistical data of simulated ADDs with the assumption of different proportions of MeHg in exposure media in Gagarinskoye

Statistics	Forecast values	Percentiles	Forecast values	Percentiles	Forecast values
Distribution	Lognormal	10%	0.01	60%	0.04
Mean	0.11	20%	0.02	70%	0.06
SD	2.44	30%	0.02	80%	0.08
Minimum	0.00	40%	0.03	90%	0.15
Maximum	207.51	50%	0.04	100%	207.51

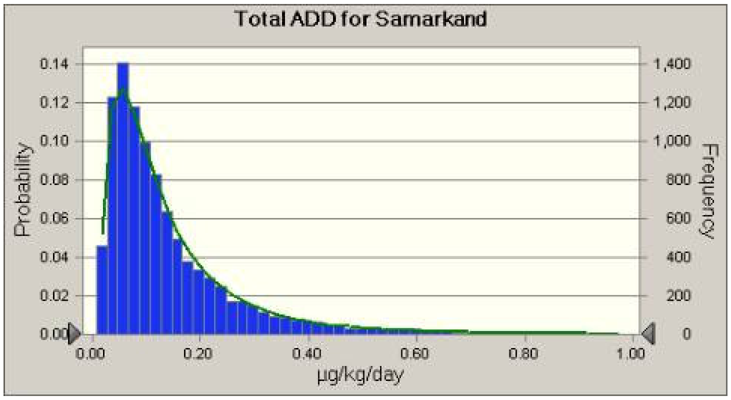


Figure H- 11 Distributions and curves of ADDs with the assumption of different proportions of MeHg in exposure media in Samarkand

Table H- 11 Statistical data of simulated ADDs with the assumption of different proportions of MeHg in exposure media in Samarkand

Statistics	Forecast values	Percentiles	Forecast values	Percentiles	Forecast values
Distribution	Lognormal	10%	0.04	60%	0.12
Mean	0.16	20%	0.05	70%	0.16
SD	0.30	30%	0.06	80%	0.21
Minimum	0.01	40%	0.08	90%	0.31
Maximum	21.69	50%	0.10	100%	21.69

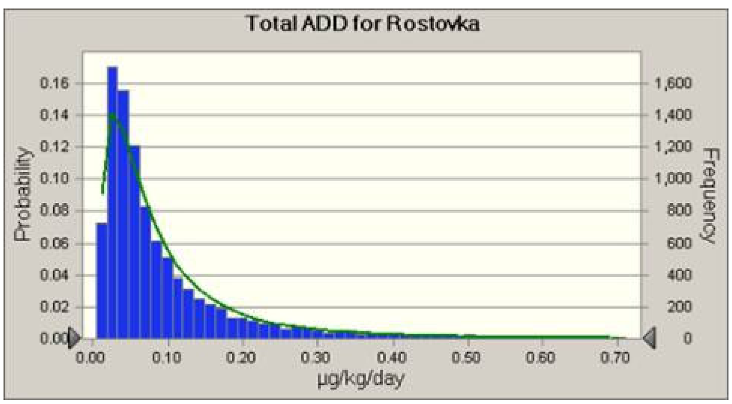


Figure H- 12 Distributions and curves of ADDs with the assumption of different proportions of MeHg in exposure media Rostovka

Table H- 12 Statistical data of simulated ADDs with the assumption of different proportions of MeHg in exposure media in Rostovka

Statistics	Forecast values	Percentiles	Forecast values	Percentiles	Forecast values
Distribution	Lognormal	10%	0.02	60%	0.08
Mean	0.12	20%	0.03	70%	0.10
SD	0.21	30%	0.04	80%	0.14
Minimum	0.00	40%	0.05	90%	0.24
Maximum	8.41	50%	0.06	100%	8.41

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