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Title: Granulometric and magnetic properties of particulate matter in the Beijing subway and the implications for air quality management

Article Type: Research Paper

Keywords: Particulate matter (PM), Beijing subway, Health, Environmental magnetism, Grain size

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Abstract: The subway system is an important traffic facility in Beijing and its internal air quality is an environmental issue that could potentially affect millions of people every day. Due to the intrinsic nature of rail abrasion in subway tunnels, iron-containing particles can be generated and become suspended in the subway environment. While some studies (e.g. Li et al., 2006) have monitored the in-train levels of PM2.5s (particles < 2.5 μm), there is a lack of systematic assessment of the concentration and characteristics of iron-containing particles in the Beijing subway system. Here we report results of a study on the granulometric and magnetic properties of particulate samples collected at different localities of the Beijing subway system. Our results show that the subway samples are characterized by the presence of very fine particles. Volume proportions of 6.1 ± 1.3 % for PM2.5s and 27.5 ± 6.1 % for PM10s (particles < 10 μm) are found in the bulk subway particulate samples. These samples exhibit a strong magnetic signal, which is approximately two orders of magnitude higher than that in naturally deposited particles collected in Beijing. Fine grained ferromagnetic and ferrimagnetic minerals (e.g. iron and magnetite, respectively) are identified from mineral magnetic measurements and scanning electric microscopy. The samples collected from the Beijing stations with platform screen doors are found to be magnetically stronger and finer than those without them, suggesting that platform screen doors have failed to block the fine iron-containing particulate matters released from the rail tunnel. Given the potential health consequences of fine suspended iron-containing particles, our results have important implications for air quality management in the Beijing subway system.

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Opposed Reviewers:

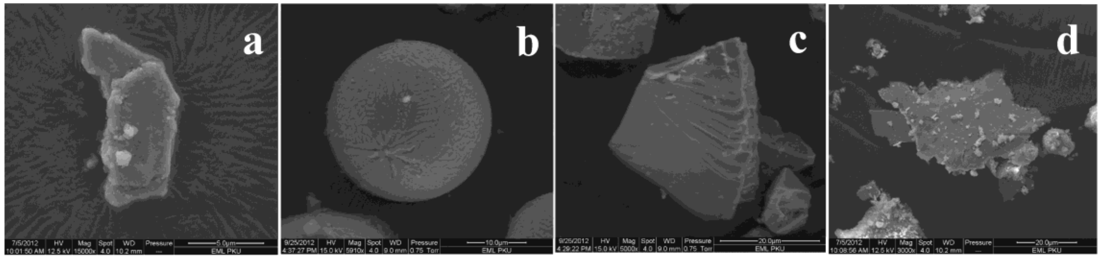
We here submit a manuscript entitled “Granulometric and magnetic properties of particulate matter in the Beijing subway and the implications for air quality management”. This work reports our finding of very fine particle matters in the Beijing subway environment, which are characterized by distinctly strong magnetic signals compared to those in naturally deposited particles. We also found variation of magnetic PM in the subway environment with platform screen door designs.

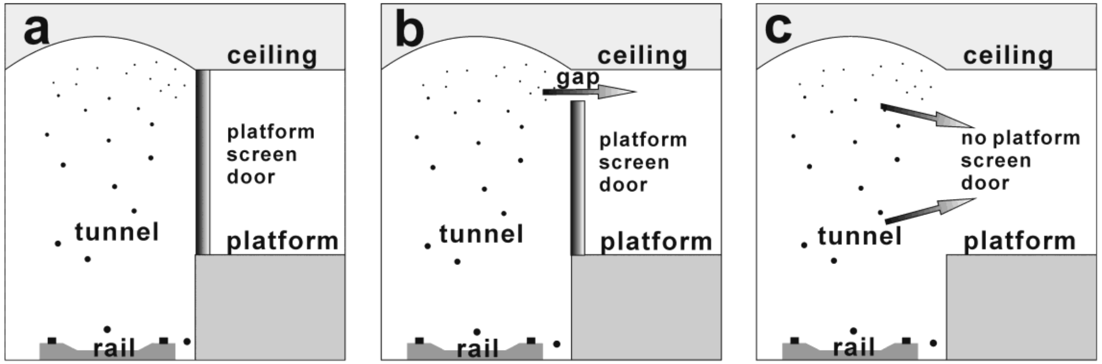
Considering the potential health consequences of fine suspended iron-containing particles identified with magnetic techniques, our results not only have important implications for air quality management in the Beijing subway system but also demonstrate a new way of assessing environmental quality in metropolitan cities with subways.

Thank you for your consideration.

*Graphical Abstract

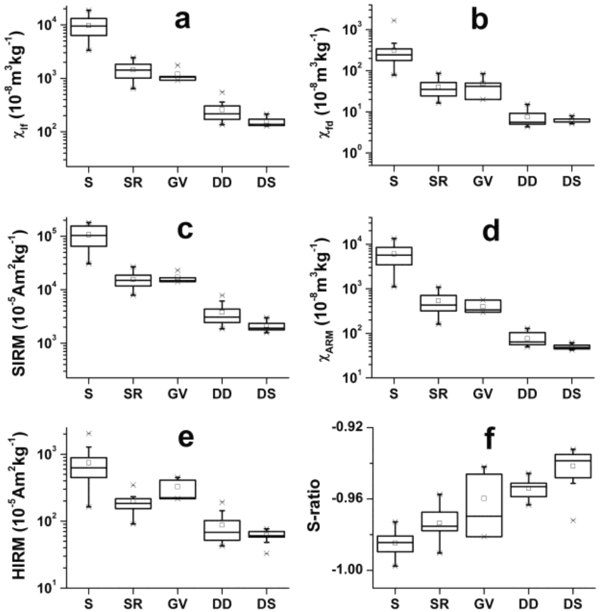
Very fine particulate matters are found in the **Beijing subway** environment.





Magnetic PM in the subway environment varies with **platform screen door designs**.

Subway samples exhibit **stronger magnetic signals** than in naturally deposited particles.



Highlights:

Very fine particulate matters are found in the Beijing subway environment;

Subway samples exhibit stronger magnetic signals than in naturally deposited particles;

Magnetic PM in the subway environment varies with platform screen door designs.

Granulometric and magnetic properties of particulate matter in the Beijing subway and the implications for air quality management

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ABSTRACT The subway system is an important traffic facility in Beijing and its internal air quality is an environmental issue that could potentially affect millions of people every day. Due to the intrinsic nature of rail abrasion in subway tunnels, iron-containing particles can be generated and become suspended in the subway environment. While some studies (e.g. Li et al., 2006) have monitored the in-train levels of PM2.5s (particles < 2.5 μm), there is a lack of systematic assessment of the concentration and characteristics of iron-containing particles in the Beijing subway system. Here we report results of a study on the granulometric and magnetic properties of particulate samples collected at different localities of the Beijing subway system. Our results show that the subway samples are characterized by the presence of very fine particles. Volume proportions of 6.1 ± 1.3 % for PM2.5s and 27.5 ± 6.1 % for PM10s (particles < 10 μm) are found in the bulk subway particulate samples. These samples exhibit a strong magnetic signal, which is approximately two orders of magnitude higher than that in naturally deposited particles collected in Beijing. Fine grained ferromagnetic and ferrimagnetic minerals (e.g. iron and magnetite, respectively) are identified from mineral magnetic measurements and scanning electric microscopy. The samples collected from the Beijing stations with platform screen doors are found to be magnetically stronger and finer than those without them, suggesting that platform screen doors have failed to block the fine iron-containing particulate matters released from the rail tunnel. Given the potential health consequences of fine suspended iron-containing particles, our results have important implications for air quality management in the Beijing subway system.

Keywords: Particulate matter (PM), Beijing subway, Health, Environmental magnetism, Grain size

1. Introduction

Over the past few years, long-lived smog conditions in Beijing with high concentrations of particulate matter (PM) have meant that air quality has become the centre of public attention (Zhang et al., 2014). Adding or extending subway and suburban railway lines is one of the key long-term measures aimed at reducing Beijing’s vehicle gas emissions, and hence to improve the urban air quality. As one of the most convenient public transportation systems for commuters, the Beijing subway has a passenger flow of ~10 million per day (www.bjsubway.com), the highest in the world. Due to the intrinsic nature of rail abrasion and train braking in the subway tunnels (Chillrud et al., 2004; Sitzmann et al., 1999), iron-containing particles can be generated and become suspended into the subway environment. Frictional abrasion contributes 40－73% of PM10s at the platforms in the Milan subway system in Italy (Colombi et al., 2013). The secondary source is the particles carried by regular passengers and air flow from the outside urban environment (Kang et al., 2008; Li et al., 2006). Earlier toxicological research has pointed out that subway PMs induce oxidative stress in cultured human lung cell (Karlsson et al., 2005, 2008) and impose strong self-limiting biological reactions on rodents (Bachoual et al., 2007), suggesting that the same PM might trigger health hazards in passengers. While lung cancer incidence was not increased (Gustavsson et al., 2008), workers in the subway service were observed to suffer a long-term inflammatory process (Bigert et al., 2008). The subway particulate matters can be accumulated and become suspended in the subway environment which could potentially affect millions of people in the city daily.

Previous attempts to assess the environmental conditions in the subway system have focused on the concentration and chemical composition of PM, with contradictory results (Aarnio et al., 2005; Boudia et al., 2006; Chillrud et al., 2004; Guo et al., 2014; Karlsson et al., 2005; Lu et al., 2015; Salma et al., 2007; Gómez-Perales et al., 2007; Querol et al., 2012) attributed to the different designs of the subway systems (Nieuwenhuijsen et al., 2007). Iron-containing minerals were found enriched in the New York (Chillrud et al., 2004), Buenos Aires (Murruni et al., 2009) and Seoul subway systems (Kang et al., 2008). Studies have been made to monitor the in-train PM2.5 levels in the Beijing subway system (Li et al., 2006, 2007). However, observations of PM concentrations alone may not be sensitive enough to characterize the intrinsic nature of the iron-containing particles.

Magnetic measurements detect and characterize iron-containing materials in natural environment with high sensitivity (e.g. Thompson and Oldfield, 1986). Numerous studies have demonstrated that magnetic techniques can be successfully used to monitor anthropogenic or geoanthropogenic pollution from surface soils (Dearing et al., 1996b;

Jordanova et al., 2003; Qiao et al., 2011; Zheng and Zhang, 2008; Zhu et al., 2010), fossil fuel combustion (Heller et al., 1998), coastal areas (Dong et al., 2014; Zhang et al., 2009), urban dust (Bucko et al., 2011; Shilton et al., 2005; Shu et al., 2000; Xie et al., 1999; Zhu et al., 2010) and historical monuments (Schiavon and Zhou, 1996). Zhang et al (2012) show the first example for the use of magnetic measurements in detecting the iron-containing particles in the Shanghai subway system.

In this study, we made granulometric and magnetic measurements of the PM collected at different localities in the Beijing subway system. The results were compared with those for the particulate matters collected from daily deposition and dust storms in the campus of Peking University (PKU). We extracted magnetic particles from the subway samples and obtained their micro-morphological and primary chemical information using scanning electron microscopy. The results provide insight about the transmission of PM within the interior of Beijing subway stations, which may be useful for improving the management of subway air quality.

2. Material and methods

2.1. Sample collection

The Beijing subway system has been in operation since 1969. Underground subway stations are dominant in downtown areas while aboveground railway stations are common in suburban districts. Independent air exchange devices are set up in most modern underground stations, with ventilation rooms outside of aboveground stations (ground vents). Suburban railway stations are usually semi-closed with regards air exchange with the outside environment. Samples of deposited PM were collected from the top of billboards and duty rooms in subway platforms with single use clean plastic brushes. The collection was processed at five subway and suburban railway lines (Lines 1, 2, 4, 5 and 13). Lines 1 and 2 are underground subway lines in which no platform screen doors are installed at their stations. Line 4 and the southern half of Line 5 are also underground subway lines but have installation of platform screen doors, although there are gaps between the ceilings and platform screen doors. The northern half of Line 5 and Line 13 are aboveground suburban railway lines in which no platform screen doors are installed. Forty-seven samples were collected from the subway system: thirty from subway stations, thirteen from suburban railway stations and four from ground vents (Fig.1). Beijing is occasionally affected by dust storms in the spring (Wang et al., 2006; Xie et al., 2005). The dust storm events are usually short but may bring high volumes of PM from central Asia and the marginal area of Beijing (cf. Pye and Zhou, 1989). Twelve samples of dust from storm events and ten from daily 'normal' deposition were collected in the campus of Peking University.

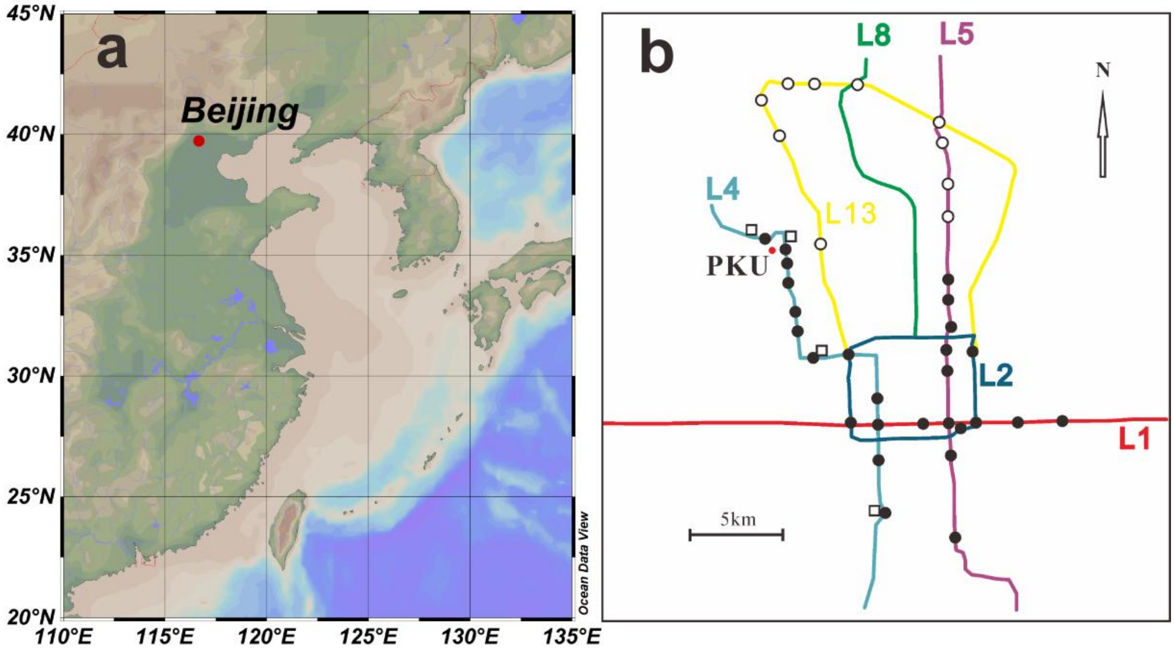

Fig. 1. Location of (a) Beijing and (b) sampling sites of subway stations (black cycles), suburban railway stations (white circles) and ground vents (squares) from five lines of the Beijing subway system. Collection of particle samples from dust storms and daily deposition was at the campus of Peking University (PKU). Note both subway stations (underground) and suburban railway stations (aboveground) exist in Line 5 as L5 (S) and L5 (SR), respectively.

2.2. Granulometric measurements

The grain size of the samples was measured with a Malvern Mastersizer 2000. In order to obtain original grain size distribution, no pretreatment procedure was applied. The median, mean (volumetrically weighted average), mode, volume proportions of PM2.5s and PM10s and gran size distribution were calculated by the Mastersizer 2000 software.

2.3. ESEM + EDS measurements

Bulk subway PM and magnetic extracts were fixed on an aluminum plate with a double faced adhesive tape, respectively. Mineral micro-morphology and primary chemical composition of subway particulate matters were obtained using a FEI Quanta 200F Environmental Scanning Electron Microscope (ESEM) equipped with an energy dispersive X-ray spectroscopy (EDS) analysis system.

2.4. Magnetic measurements

The low-field frequency (0.47 kHz) and high-field frequency (4.7 kHz) volume magnetic susceptibility (κ_{lf} and κ_{hf}) were measured with a MS2 Bartington magnetic meter. Mass susceptibility was calculated as $\chi_{lf} = \kappa_{lf} / \rho$ and $\chi_{hf} = \kappa_{hf} / \rho$. The magnetic susceptibility is primarily controlled by the concentration of ferromagnetic or ferrimagnetic materials and can be enhanced by the presence of very fine superparamagnetic (SP) magnetite (< 0.025 μm for magnetite) and coarse magnetite grains (> 10 μm).

Frequency-dependent magnetic susceptibility (χ_{fd}) was calculated as $\chi_{fd} = (\chi_{lf} - \chi_{hf})$ and percentage frequency-dependent susceptibility (χ_{fd} %) as $\chi_{fd}\ \% = (\chi_{lf} - \chi_{hf}) / \chi_{lf}$. Both χ_{fd} and χ_{fd} % are used to detect the presence of the SP magnetic grains. The frequency-dependent component is mainly controlled by ferrimagnetic grains with a diameter below 0.025 μm.

Anhysteretic remanent magnetization (ARM) was induced in a DC field of 0.04 mT imposed on a peak alternating field of 100 mT measured by a Molspin JR6 spinner magnetometer. Magnetic susceptibility of ARM (χ_{ARM}) was mass-normalized per unit bias field. The χ_{ARM} is a measure of the concentration of ferrimagnetic material and strongly grain-size dependent. It preferentially responds to single domain (SD, 0.025—1 μm for magnetite) particles.

Isothermal remanent magnetization (IRM) was acquired in fields of 20 mT, 100 mT, 300 mT and 1000 mT, then backfields of -20 mT, -100 mT and -300 mT using a Molspin pulse magnetizer and a JR6 spinner magnetometer. The maximum remanent magnetization at 1000 mT is termed the saturation isothermal remanent magnetization (SIRM). SIRM is often used to represent the saturated remanence acquired by a sample after exposure to the highest field which is usually in the range 300—1000 mT (we set 1000 mT in this study). SIRM primarily depends upon the concentration of remanence-carrying minerals. Hard IRM (HIRM) was defined as HIRM = $(\mathrm{SIRM} - \mathrm{IRM}_{-300mT}) / 2$. HIRM serves as an estimation of the total concentration of magnetic material with high coercivity. The S-ratio was obtained by dividing the “backwards” remanence (-300 mT in this study as IRM_{-300mT}) by the SIRM. It can be used to estimate the composition of the magnetic mineralogy, with values close to -1 indicating lower coercivity (“soft”) and a ferromagnetic and ferrimagnetic mineralogy (e.g. iron and magnetite) and close to zero indicating a higher coercivity (“hard”), possibly an antiferromagnetic (e.g. hematite and goethite) mineralogy.

3. Results

3.1. Grain size

The median grain size of subway PM ranges from 12.9 μm to 30.1 μm, with a mean value of 18.5 μm, which is similar to the median size of suburban railway station and dust storm particles. The median grain size of ground vent particles ranges from 12.7 μm to 13.9 μm, with a mean value of 13.3 μm, while the median of daily deposition particles ranges from 12.6 μm to 38.9 μm, with a mean value of 29.9 μm (Table 1). The ground vent particles are finer than daily deposition particles.

Table 1

Granulometric parameters of suspended particles collected inside and outside of the Beijing subway system.

		Subway station	Suburban railway station	Ground Vents	Daily Deposition	Dust Storm
Number of samples		27	6	4	7	10
Mode (μm)	Range	16.2—27.7	17.1—32.0	15.3—16.2	15.7—52.9	18.6—31.6
	Mean ± SD	21.3 ± 2.6	23.2 ± 5.3	15.7 ± 0.4	37.4 ± 12.3	23.3 ± 4.7
Median (μm)	Range	12.9—30.5	13.7—29.1	12.7—13.9	12.6—38.9	14.7—26.7
	Mean ± SD	18.5 ± 3.8	20.0 ± 5.7	13.3 ± 0.5	29.9 ± 9.6	18.6 ± 4.4
Mean (μm)	Range	15.3—55.2	16.1—43.0	14.6—17.5	15.2—54.0	18.0—40.0
	Mean ± SD	27.4 ± 9.8	26.9 ± 11.3	16.2 ± 1.3	42.4 ± 13.5	24.3 ± 8.1
< 2.5 μm (%)	Range	3.4—8.2	4.4—8.2	6.1—7.7	3.0—9.3	5.1—8.5
	Mean ± SD	6.1 ± 1.3	6.3 ± 1.4	7.1 ± 0.7	5.1 ± 2.1	7.3 ± 1.3
< 10 μm (%)	Range	14.3—38.7	15.6—35.7	34.5—38.4	10.7—39.6	18.6—34.6
	Mean ± SD	27.5 ± 6.1	25.6 ± 7.4	36.3 ± 1.9	19.3 ± 10.0	28.6 ± 5.7

Volume percentages of PM2.5s and PM10s in the subway PM (with mean values of 6.1% and 27.6%, respectively) are close to the values of particles from suburban railway stations, ground vents and dust storms, higher than for daily deposition particles (5.1% and 19.3%, respectively). All medians of sample grain size are smaller than their corresponding means (Table 1). The grain size distribution curves are negatively skewed (Fig. 2). Note that there might be subordinate modes at 0.6—0.8 μm for all five types of particles and 200 μm for daily deposition particles, indicating a possible mixing from different particle sources.

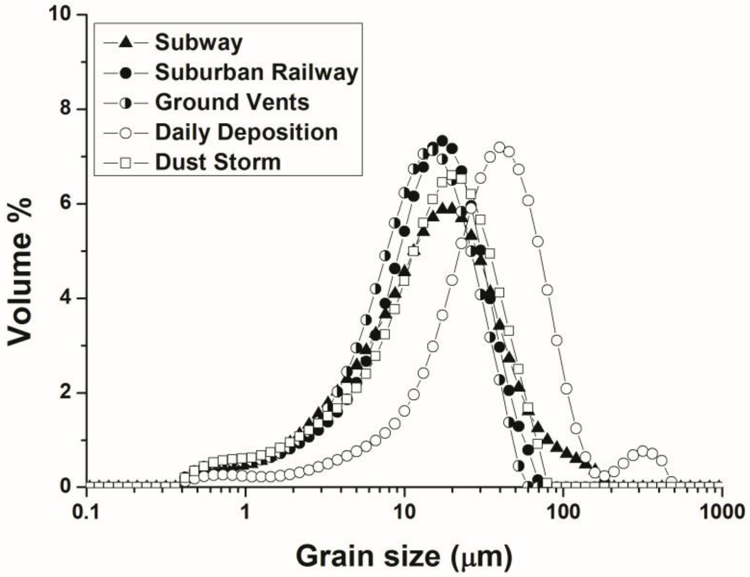

Fig. 2. Grain size distribution curves for five types of samples. Samples selected for subway stations, suburban railway stations and ground vents are at East Gate of Peking University Station on Line 4, East of Datun Rd. Station on Line 5 and Yuanmingyuan Station on Line 4, respectively.

3.2. ESEM + EDS observations

ESEM + EDS results reveal the presence of iron and magnetite in subway PM (Fig. 3) with different micro-morphological characteristics observed in subway and suburban railway PM. Irregular sheet particles (Fig. 3a) are detected in subway PM, with diameters less than 80 μm. Spherical (Fig. 3c), block (Fig. 3e) and irregular sheet (Fig. 3g) particles are detected in suburban railway particulate matters, with diameters less than 50 μm. Considerable quantities of PM2.5s and PM10s are detected throughout the samples. The EDS analysis reveals that irregular sheet particles are composed of iron or incompletely oxidized iron (Fig. 3b), while spherical and block particles are composed of iron oxide (Fig. 3d and h). Quite a few very fine particles (< 1 μm) are found attached to irregular sheet, spherical or block magnetic particles in subway and suburban railway particulate matters. It is possible that they are magnetically attached to the larger particles. Small quantities of carbonate and molybdenum oxide are also detected in subway PM. Spherical and block iron oxide particles are dominant in magnetic extracts of particles from dust storm and daily deposition samples. These particles are better sorted and rounded than subway magnetic particles which might be explained by long distance transportation. Quartz is dominant in PM from suburban railway stations, dust storms and daily deposition, while quartz and iron particles dominate in PM from underground subway station.

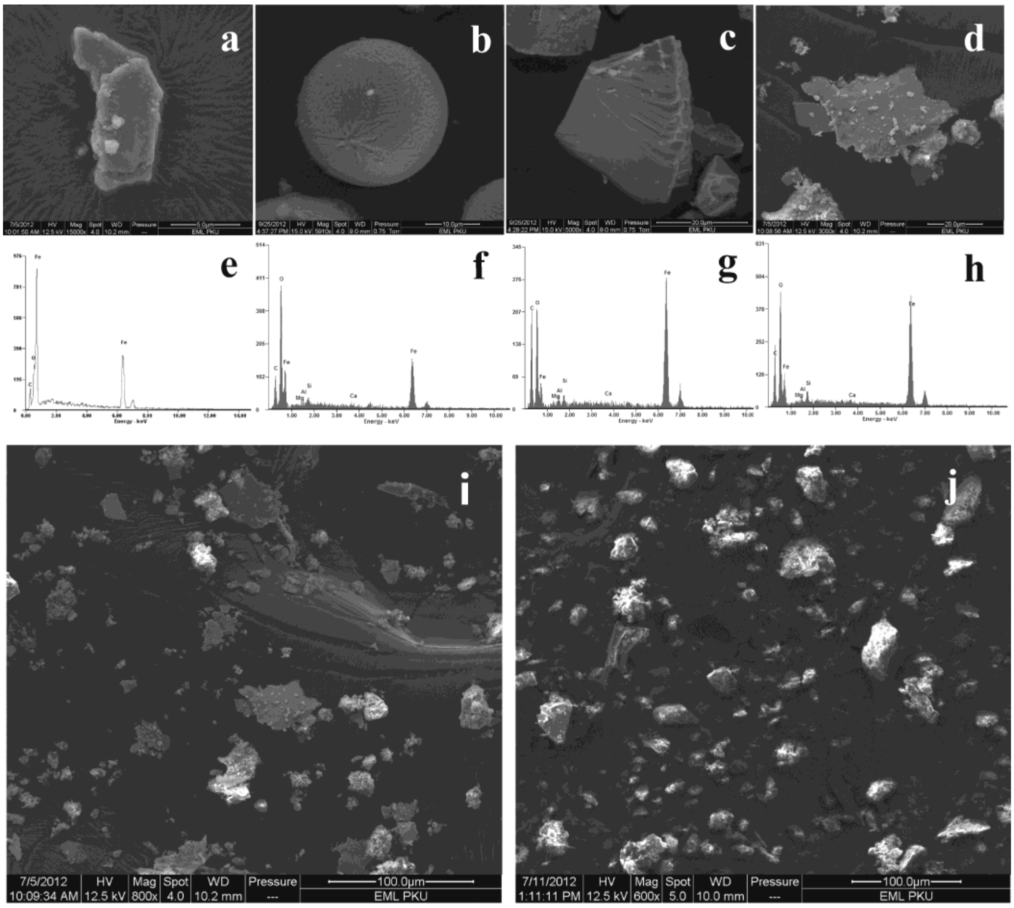

Fig.3. ESEM-EDS results of selected samples. (a) Irregular sheet iron-containing particle in subway particulate matters (Haidian Huangzhuang Station on Line 4) and (b) its primary chemical properties from EDS; (c) spherical, (e) block and (g) irregular sheet iron-containing particles in suburban railway particulate matters (Shangdi Station on Line 13) and (d, f and h, respectively) their primary chemical properties from EDS; Magnetic extracts of (i) subway particulate matters (Haidian Huangzhuang Station on Line 4) and (j) suburban railway particulate matters (Shangdi Station on Line 13) are shown. The high C and O value could be an effect of a double faced adhesive tape on the aluminum plate target.

3.3. Magnetic measurements

3.3.1. Magnetic mineral concentration

Magnetic concentration parameters (χ_{lf}, χ_{fd}, χ_{ARM}, SIRM and HIRM) vary among PM samples from subway stations, suburban railway stations, ground vents, daily deposition and dust storms (Table 2). All these parameters are approximately one order of magnitude higher in subway samples than for the suburban railway samples and two orders of magnitude higher than samples from dust storms and daily deposition. Meanwhile, the mean values of magnetic concentration parameters for particles from ground vents are close to that of the suburban railway PM.

Table 2

Magnetic parameters of suspended particles collected inside and outside of the Beijing subway system.

		Subway station [a]	Suburban railway station	Ground Vents	Daily Deposition	Dust Storm
Number of samples		30	13	4	10	12
χ_{lf} ($10^{-6}m^3\cdot kg^{-1}$)	Range	33.4—188.6	6.4—24.4	9.2—17.5	1.4—5.5	1.3—2.2
	Mean ± SD	95.1 ± 39.8	14.4 ± 5.7	11.8 ± 3.3	2.6 ± 1.2	1.6 ± 0.3
χ_{fd} ($10^{-8}m^3\cdot kg^{-1}$)	Range	78.3—506.7	16.3—86.7	20.0—85.0	4.3—15.3	5.2—8.0
	Mean ± SD	280.3 ± 136.6	40.1 ± 21.3	49.2 ± 27.0	7.4 ± 3.4	6.1 ± 0.8
χ_{fd}%	Range	1.5—4.2	1.7—3.4	2.2—4.8	2.4—3.2	3.1—5.3
	Mean ± SD	2.9 ± 0.8	2.7 ± 0.7	3.9 ± 1.2	2.9 ± 0.2	4.0 ± 0.6
χ_{ARM} ($10^{-6}m^3kg^{-1}$)	Range	11.1—134.8	1.6—10.9	3.0—5.6	0.5—1.3	0.4—0.6
	Mean ± SD	58.6 ± 30.3	5.4 ± 2.9	3.9 ± 1.2	0.8 ± 0.3	0.5 ± 0.1
SIRM ($10^{-2}Am^2kg^{-1}$)	Range	30.8—181.0	7.9—26.8	14.2—23.0	1.9—7.9	1.6—3.0
	Mean ± SD	104.8 ± 46.6	15.6 ± 5.7	17.2 ± 4.1	3.8 ± 1.9	2.2 ± 0.5
IRM_{-300mT} ($10^{-2}Am^2kg^{-1}$)	Range	-178.8—-30.1	-26.2—-7.6	-22.6—-13.4	-7.5—-1.8	-2.9—-1.4
	Mean ± SD	-103.4 ± 46.2	-15.2 ± 5.6	-16.5 ± 4.2	-3.6 ± 1.8	-2.0 ± 0.5
S-ratio	Range	-0.998—-0.973	-0.990—-0.957	-0.981—-0.942	-0.963—-0.946	-0.972—-0.932
	Mean ± SD	-0.985 ± 0.007	-0.97 ± 0.008	-0.960 ± 0.019	-0.954 ± 0.005	-0.942 ± 0.011
HIRM ($10^{-4}Am^2kg^{-1}$)	Range	16.4—204.0	9.1—34.5	21.7—45.0	4.2—19.1	3.2—7.7
	Mean ± SD	73.4 ± 43.8	19.6 ± 7.5	32.6 ± 12.3	8.7 ± 4.7	6.2 ±1.2
χ_{ARM}/SIRM ($10^{-5}A^{-1}m$)	Range	3.6—10.4	2.0—5.3	2.1—2.4	1.5—2.9	1.6—3.5
	Mean ± SD	5.6 ± 1.5	3.3 ± 1.1	2.2 ± 0.2	2.2 ± 0.5	2.4 ± 0.5
SIRM/χ_{lf} (10^3Am^{-1})	Range	7.8—16.1	8.8—12.8	13.1—15.5	13.3—17.0	12.2—14.5
	Mean ± SD	11.0 ± 2.0	11.1 ± 1.3	14.5 ± 1.1	14.4 ± 1.0	13.7 ± 0.6
χ_{ARM}/χ_{lf}	Range	0.31—1.23	0.25—0.53	0.32—0.33	0.22—0.41	0.23—0.43
	Mean ± SD	0.61 ± 0.18	0.36 ± 0.08	0.32 ± 0.00	0.31 ± 0.06	0.33 ± 0.05

[a] An abnormal subway data point is excluded which shows as high as of 1667.0 ×$10^{-8}m^3\cdot kg^{-1}$ of χ_{fd} and 10.9% of χ_{fd}%. Its median of grain size is much less than all other subway samples, which is likely derived from a distinctively different source.

The concentration of magnetic particles in a total sample can be estimated by dividing the bulk susceptibility of the sample by the susceptibility of the assumed magnetic mineral type or size (Dearing, 1999). If assuming all particles are homogeneously mixed and the magnetic mineral is pure magnetite with a magnetic susceptibility of 6.5×10^{-4} m^3 kg^{-1} (Thompson and Oldfield, 1986), the estimated mean concentration of magnetite in subway particulate matters is 14.6 ± 6.1 %. The magnetite concentrations of particles from suburban railway stations, ground vents, daily deposition and dust storms are 2.2 ± 0.9 %, 1.8 ± 0.5 %, 0.2 ± 0.2 % and 0.2 ± 0.1 %, respectively. If assuming magnetic particles are pure iron with a magnetic susceptibility of 2.8×10^{-1} m^3 kg^{-1} (Dearing, 1999), the estimated mean concentration of iron in subway particulate matters is ~0.03%.

3.3.2. Magnetic mineral grain size

The low χ_{fd} % values (1.5- 5.3%, Table 2) indicate that the SP contribution is a relatively minor to moderate part of the total magnetic susceptibility (Dearing et al 1996a) in all samples. However, higher mass specific χ_{fd} values in subway samples compared to the χ_{fd} of naturally deposited particles shows that subway samples have higher concentrations of SP magnetic particles. The χ_{fd} is well-correlated with χ_{lf} (Fig. 4a), while χ_{fd} % is independent of χ_{lf} (Fig. 4b). The χ_{ARM} values display a linear correlation with χ_{lf} (Fig. 4e), showing that the SD grains in subway particulate matters are also enriched.

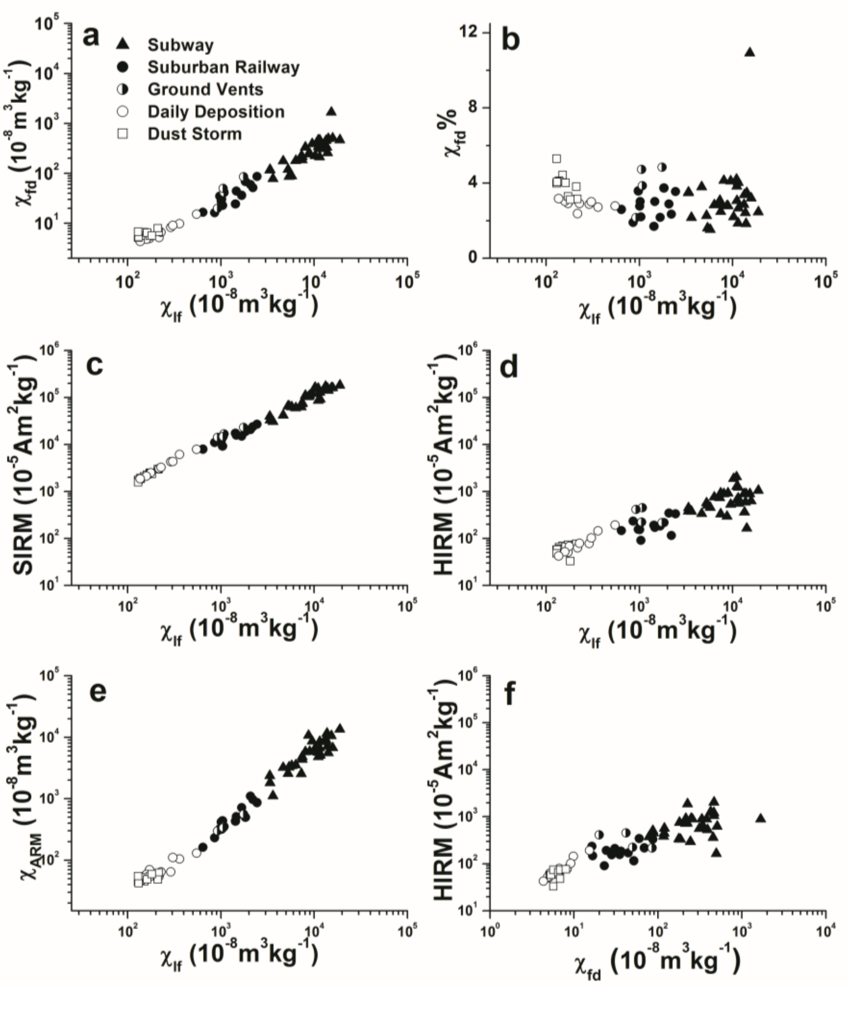

Fig. 4. Magnetic parameters are plotted as (a) χ_{fd} against χ_{lf}, (b) χ_{fd}% against χ_{lf}, (c) SIRM against χ_{lf}, (d) HIRM against χ_{lf}, (e) χ_{ARM} against χ_{lf}, and (f) HIRM against χ_{fd}. Note all coordinates of magnetic parameters are exponential except χ_{fd}%.

The χ_{fd} % versus χ_{ARM}/SIRM plot (Fig. 5) (Dearing et al., 1997), shows the subway particulate matters are dominated by multi-domain (MD, 10—100 μm for magnetite) and pseudo-single domain (PSD, 1—10 μm for magnetite) particles. The plot also confirms that the magnetic particles in the subway system are finer than the urban particles.

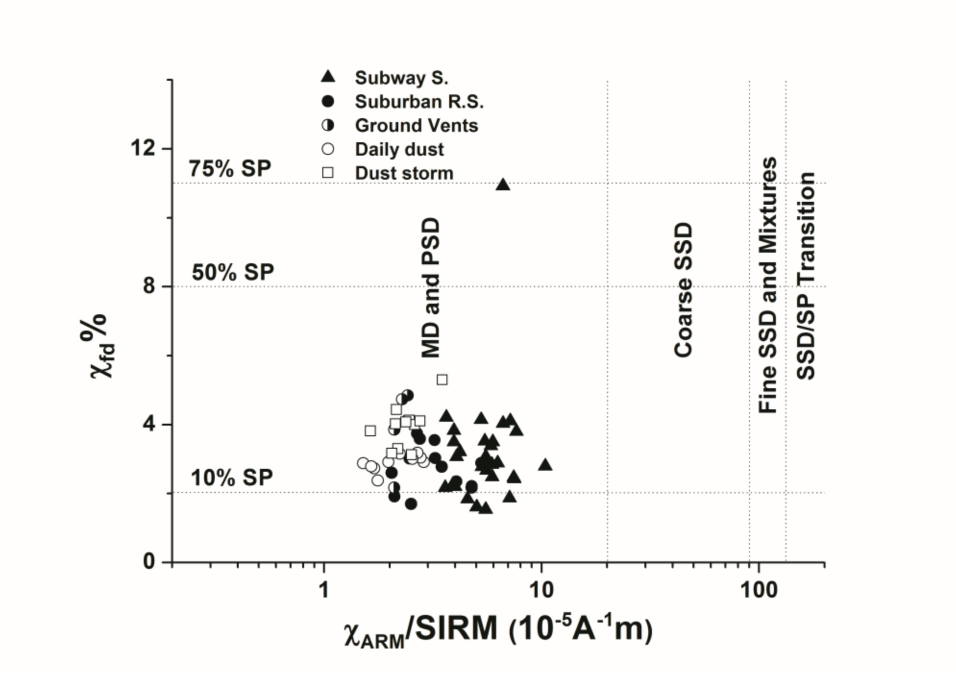

Fig. 5. A Dearing plot on a semi-log scale, showing the grain size range of the magnetic particles in the studied samples.

3.3.3. Magnetic mineral composition

The SIRM correlates well with χ_{lf} (Fig. 4c), indicating that magnetic particles are dominated by ferromagnetic and ferrimagnetic particles. The SIRM value of subway PM is two orders of magnitude higher than daily deposition particles. While the HIRM values display a correlation with χ_{lf} (Fig. 4d), the values are two orders of magnitude lower than the SIRM values, suggesting that high coercivity minerals are minor in the total range of magnetic minerals. Similar correlations exist between the HIRM and χ_{fd} (Fig. 4f). The S-ratio of subway PM ranges from -0.998 to -0.957 (dimensionless unit), with a mean value of -0.981 (Table 2). This indicates that the subway magnetic particles are dominated by low coercive ferromagnetic and ferrimagnetic particles.

4. Discussion

4.1. Possible emission pathway of subway PM

PM collected at Beijing subway is found to have extremely high magnetic concentrations compared with the local street dust and surface soil samples (Table 3). This can be attributed to internal sources of iron-rich particles (Chillrud et al., 2004; Sitzmann et al., 1999). Suburban railway PM is also found to have high magnetic concentration; lower than particles from subway stations but much higher than naturally deposited particles. ESEM results reveal the abundance of quartz in PM from suburban railway stations. The spherical oxidized iron particles which are usually enriched in the urban environment due to fuel combustion (Heller et al., 1998; Qiao et al., 2011; Schiavon and Zhou, 1996) are also observed in suburban railway but not subway PM. Colombi et al. (2013) showed that the PM10 concentrations in the Milan subway system had a decreasing trend from platform to mezzanine levels to ambient air, suggesting that air quality might be influenced by the structure of the subway system. Beijing's suburban railway system has a semi-open design with natural air exchange between the inside and outside of the station, which provides an opportunity for the mixture of anthropogenic iron-containing particles and urban naturally suspended particles.

Table 3

Magnetic susceptibilities of several types of particles in Beijing are summarized.

Sample type	Number of samples	χ_{lf} range ($10^{-8}m^3\cdot kg^{-1}$)	Mean χ_{lf} ± SD ($10^{-8}m^3\cdot kg^{-1}$)	Study
Surface soil	91	34.4—379.3	110.2 ± 62.1	Zhu et al., 2010 [a]
	63	—	≈ 200 [e]	Zheng and Zhang, 2008 [b]
	18	24.0—102.2	61.6 ± 4.6	Qiao et al., 2011 [c]
	1	—	950.6 [f]	Shen et al., 2006 [d]
Dust storms	12	129.0—216.0	144.3 ± 30.9	This study
Street dust	63	—	≈ 400 [e]	Zheng and Zhang, 2008
	160	84.5—618.8	243.6 ± 92.8	Qiao et al., 2011
	10	136.3—549.9	222.9 ± 124.7	This study
Subway PM	30	3338.3—18858.3	9867.7 ± 4051.2	This study

[a] 0—10 cm depth of urban and suburban surface soil where the bedrock is fluvial sediments.

[b] 0—1cm and 0—5cm depth of urban area surface soil.

[c] surface soil in Beijing Olympic Park.

[d] 0—3 cm depth of a polluted soil profile at an industrial area of western Beijing.

[e] estimated mean value.

[f] only a mean value is given for 0—3 cm depth samples.

A recent study demonstrated that PM emitted from a subway tunnel through ground vents could be partially removed using magnetic filters (Son et al., 2014). In our study, particles outside of the ground vents of modern subway stations show identical magnetic properties with suburban railway particles rather than naturally deposited particles. The median grain size of ground vent particles is smaller than the subway PM or naturally deposited particles. This implies that finer particles, rather than the coarser ones, may have been prior elevated by air exchange device to the ground. The proportion of underground iron-containing particles might be elevated through the air exchange system to the ground before mixing with street dust from the adjacent area. These fine iron-containing particles thus contribute to the general PM loading to the external urban environment.

Magnetic concentration parameters show a general decreasing trend from subway stations to ground vents and to naturally deposited particles (Fig. 6a—e) while the S-ratios show the opposite tendency (Fig. 6f). This suggests that the subway PM has a lower proportion of antiferromagnetic (e.g. hematite and goethite) minerals than the natural deposition particles. Overall, magnetic properties of different types of particles point to a gradual particle mixing process and a potential magnetic particle emission pathway from the subway system to the urban environment.

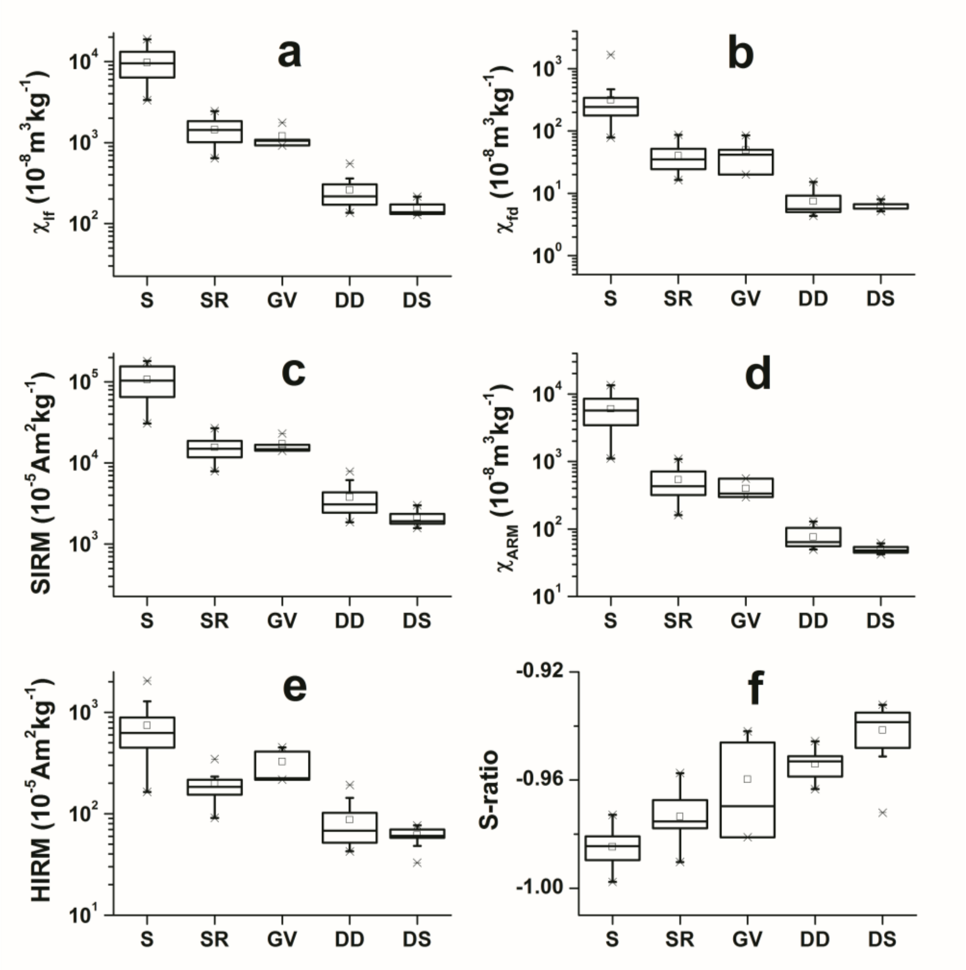

Fig. 6. Box plots of (a) χ_{lf}, (b) χ_{fd}, (c) SIRM, (d) χ_{ARM}, (e) HIRM and (f) S-ratio of five types of samples in this study. Magnetic concentration parameters of particles show a general decreasing trend from subway station (S) to suburban railway station (SR), ground vents (GV), daily deposition (DD) and dust storms (DS).

While magnetic concentration parameters (χ_{lf}, χ_{fd}, χ_{ARM}, SIRM) show identical orders of magnitude in both Beijing and Shanghai (Zhang et al., 2011) subway PM, magnetic parameter ratios like χ_{ARM}/χ_{lf}, SIRM/χ_{lf} and χ_{ARM}/SIRM are different (Fig. 7). The ratios are usually employed as magnetic grain size indicators of magnetite (Thompson and Oldfield, 1986). Both χ_{ARM} and χ_{lf} increase linearly with increasing magnetite concentration, but small SD grains are relatively more sensitive to ARM. Thus χ_{ARM}/χ_{lf} varies inversely with magnetic grain size in the 1—10μm grain-size range. The χ_{ARM}/χ_{lf} ratio (Fig. 7a) of Beijing underground subway PM (0.6, dimensionless unit) is about half of that in the Shanghai subway (1.2) (Zhang et al., 2011), which suggests magnetic particles of Beijing subway particulate matters might be coarser assuming their grain size ranges from 1μm to 10μm. The SIRM/χ_{lf} ratio is more sensitive than χ_{ARM}/χ_{lf} to changes in the proportion of large grains (> 10 μm) because the SIRM varies as a function of grain size over a wider range than χ_{ARM} (e.g. Thompson and Oldfield, 1986). The SIRM/χ_{lf} ratio (Fig. 7b) of Beijing subway PM ($11.0\times10^{3}Am^{-1}$) is slightly lower than that in Shanghai ($12.4\times10^{3}Am^{-1}$) (Zhang et al., 2011). This would imply that the grain sizes are almost identical, assuming they are above 10μm. The χ_{ARM}/SIRM is less sensitive to grain size than the other two ratios but only responds to remanence carrying magnetic material and is therefore not affected by SP and paramagnetic grains (e.g. Zhou et al., 1990). The χ_{ARM}/SIRM (Fig. 7) of Beijing subway particulate matters ($5.5\times10^{-5}A^{-1}m$) is approximately half to that in Shanghai ($9.7\times10^{-5}A^{-1}m$) (Zhang et al., 2011), which also supports the inference about the overall grain size range based on the χ_{ARM}/χ_{lf} data. Overall, the magnetic ratio comparisons suggest that magnetic particles in the Beijing subway particulate matters are coarser than those in Shanghai.

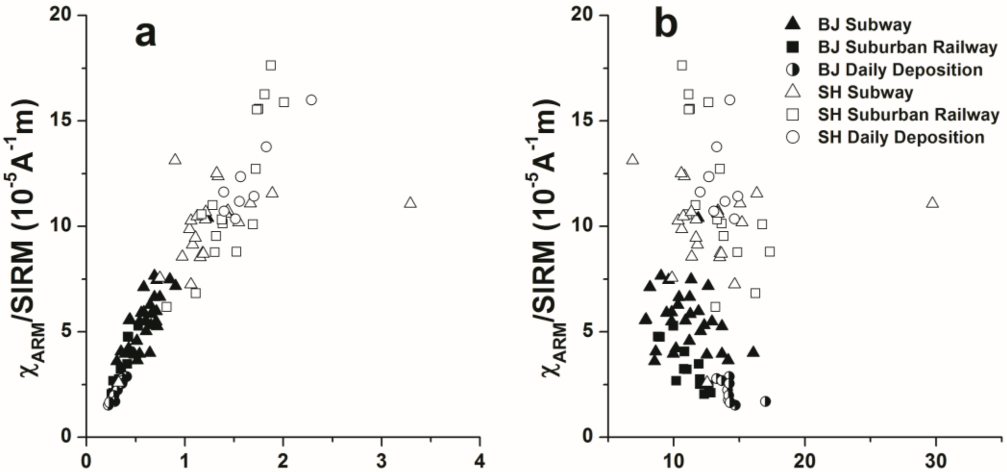

Fig. 7. Magnetic parameter ratios of χ_{ARM}/χ_{lf}, SIRM/χ_{lf} and χ_{ARM}/SIRM between Beijing (BJ) and Shanghai (SH) subway system as grain size indicators of magnetite.

4.2. Implications for subway air quality management

PM2.5s and PM10s have a damaging impact on health which might lead to short term or long term respiratory illness (Brunekreef and Holgate, 2002; Morman and Plumlee, 2014;

Pope III et al., 1991). In this study, grain size properties of Beijing subway PM show that considerable amounts of magnetic particles lie within the < 2.5 μm and < 10 μm grain size ranges. The χ_{fd} values of subway PM are two orders of magnitude higher than those of naturally deposited particles, which indicates that large amount of very fine submicron SP particles exists in subway PM: essentially PM1.0s. In the subway system of Stockholm, exposure levels of PM2.5 and PM10 are a factor of 5–10 times higher than the corresponding values measured on the busiest streets in city area (Johansson and Johansson, 2003). PM2.5s and their potential danger to the public have attracted numerous discussions, especially after the Beijing 2008 Olympic Games (Huang et al., 2012a; Huang et al., 2012b; Lin et al., 2011). Pulmonary inflammation and cardiovascular disease risk increased after air pollution control during the Olympic period (Huang et al., 2012a; Huang et al., 2012b). Moreover, enrichment of heavy metals, like Fe and others, in fine particles could also induce potential health consequences (Bachoual et al., 2007; Bigert et al., 2008; Karlsson et al., 2008; Karlsson et al., 2005; Morman and Plumlee, 2014). Considering the presence of PM sustained and accelerated by the strong air convection in the Beijing subway environment, there is immense scope for the collaboration between environmental and toxicological specialists.

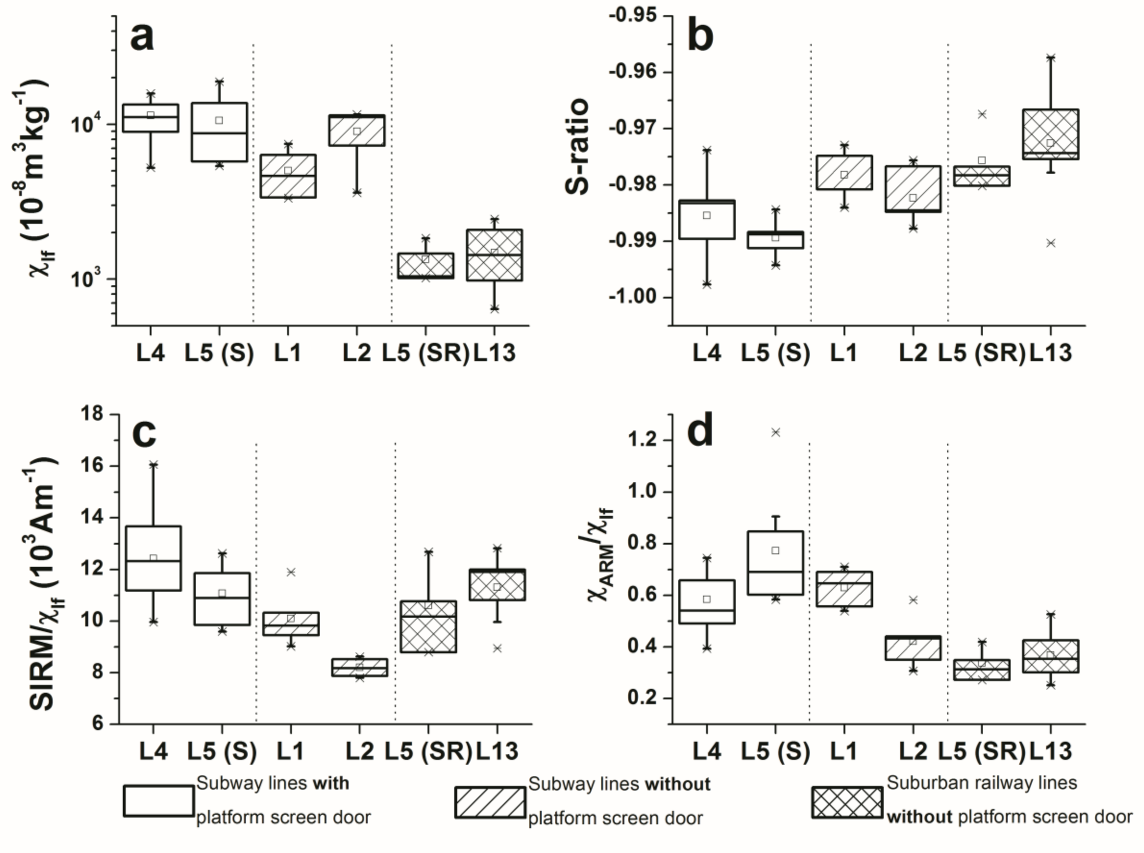

Fig. 8. Box plots of (a) χ_{lf}, (b) S-ratio, (c) SIRM/χ_{lf} and (d) χ_{ARM} /χ_{lf} of different subway lines. They can be divided into three categories according to different types of platform screen doors. Higher values of SIRM/χ_{lf} and χ_{ARM} /χ_{lf} in subway lines with platform screen doors reveal finer particles than those without platform screen doors. Both subway stations (underground) and suburban railway stations (aboveground) exist in Line 5 as L5 (S) and L5 (SR), respectively.

Magnetic and granulometric parameters vary among different types of subway lines as a result of the presence and type of platform screen doors. Barely any deposited particles are collected in subway Line 8 (the Olympic Branch Line), which is probably a result of the complete closure between the tunnel and platform by platform screen doors. Unsurprisingly, the results for Line 8 are similar to those for particles from Shanghai subway Line 1 which also has full height screen doors (Zhang et al., 2011). Elsewhere in Beijing, large amounts of particles are collected in subway Line 4 and Line 5 where the top of the platform screen door is not connected to the platform ceiling, and in Line 1 and Line 2, where there is no platform screen door between the tunnel and platform. Particles from stations with a gap between the tunnel ceilings and platform screen doors (Line 4 and part of Line 5) are magnetically stronger than those without platform screen doors (Line 1 and Line 2) (Fig. 8a), with higher concentrations of low coercive ferromagnetic and ferrimagnetic particles (Fig. 8b). The former is relatively finer than the latter as interpreted from the magnetic parameter ratios (Fig. 8c and d) and median grain size (17.4 μm and 21.5 μm, respectively).

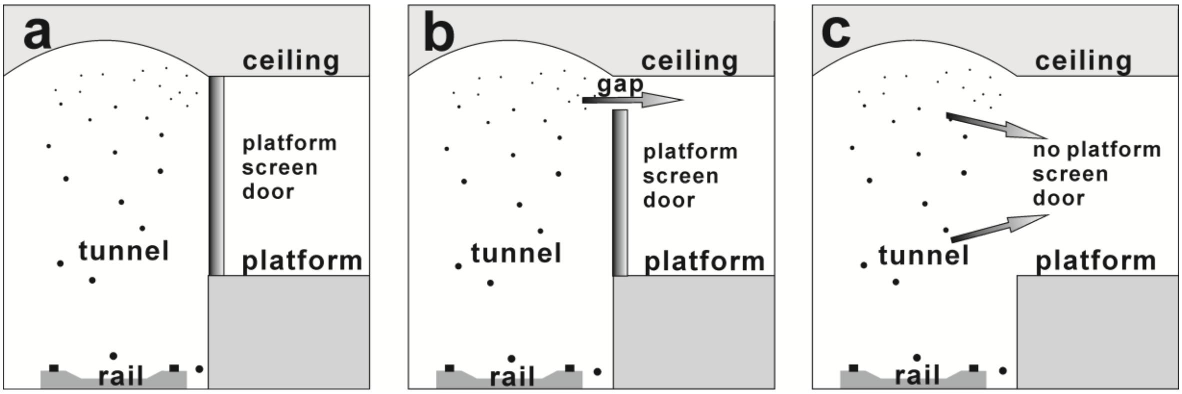

Fig. 9. Three subway particle emission scenarios from tunnel to platform according to different types of platform screen door designs. (a) Without gap between platform screen door and ceiling, iron-containing particles only suspend in tunnel. (b) With gap between platform screen door and ceiling, the fine particles could be released through the gap into the platform more easily than the coarse ones. (c) Without platform screen door, both fine and coarse particles could be released into the platform.

In the Beijing subway system, the installation of platform screen doors was supposed to be initially served as a safety measure to block passengers from the tunnel. It might reduce the emission of iron fragments if there was no gap between ceiling and screen door device (Line 8) (Fig. 9a). But platform screen doors with gaps between ceiling and screen door device cannot completely block the transport of magnetic particles from tunnel to platform. Furthermore, finer grains may preferentially move to the platform because they are held in suspension at the height of the gap between ceiling and screen door for longer than the coarser grains which might settle-out in the tunnel (Fig. 9b). Particles in the platform without platform screen doors (Line 1 and Line 2) are probably in a state of continuous suspension and settling (Fig. 9c.). In Seoul, Korea, Kim et al. (2012) found that PM10 concentrations at the platform were reduced by 16% after installation of full height platform screen doors. Therefore, the installation of full height screen doors in the Beijing subway could be an effective way to improve platform air quality, but the issue of removing the deposited subway

PM from tunnels or slowing their release through air vents to the external urban environment remain important challenges.

5. Conclusions

Granulometric and magnetic properties of PM collected from five of the Beijing Subway lines are systematically measured to assess ambient air quality in subway systems. The results reveal the presence of very fine particles in the subway environment. Mean proportions of 6.1 ± 1.3 % for < 2.5 μm and 27.5 ± 6.1 % for < 10 μm fine particles are detected in the studied samples. These samples are characterized by a strong magnetic signal which is carried by ferromagnetic and ferrimagnetic minerals as identified by mineral magnetic measurements and with ESEM-EDS. Magnetic concentration parameters (χ_{lf}, χ_{fd}, χ_{ARM}, SIRM and HIRM) of subway station PM are two orders of magnitude higher than those of surface soil and street dust in Beijing. The magnetic grain size of the subway samples are enriched in submicron SP and SD fine magnetic particles. Magnetic concentration parameters of particles show a general decreasing trend from subway station to suburban railway station, to ground vents and to naturally deposited particles. The samples collected from subway stations with gaps between ceilings and platform screen doors are finer and magnetically stronger than those without platform screen doors. Platform screen doors in Beijing subway system seem to have failed to block the fine iron-containing particles releasing from the rail tunnel. The emission of iron fragments would be reduced if full height screen doors were installed. Subway PM suspended in platforms or released through air exchanging device to urban environment may represent a health hazard to commuters and local residents. Granulometric and magnetic measurements of fine suspended particles in Beijing subway can be successfully and effectively used for monitoring subway particle pollution, thus providing reference data for the design of subway platform device and management of subway air quality. Given the potential environmental health hazards of subway PM, the results of this study provide an opportunity for further collaboration between environmental and toxicological research communities.

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