Systems scale assessment of the sustainability implications of emerging green initiatives

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Abstract (149 words) - This paper demonstrates a systems framework for assessment of environmental impacts from 'green initiatives', through a case study of meso-scale, anthropogenic-biogenic interactions. The following cross-sectoral green initiatives, combining the emerging trends in the North East region of the United Kingdom, have been considered - increasing the vegetation cover; decarbonising road transport; decentralising energy production through biomass plants. Two future scenarios are assessed - Baseline_2020 (projected emissions from realisation of policy instruments); Aggressive_2020 (additional emissions from realisation of green initiatives). Resulting trends from the Aggressive_2020 scenario suggest an increase in emissions of pollutant precursors, including biogenic volatile organic compounds and nitrogen dioxide over the base case by up to 20% and 5% respectively. This has implications for enhanced daytime ozone and secondary aerosols formation by up to 15% and over 5% respectively. Associated land cover changes show marginal decrease of ambient temperature but modest reductions in ammonia and ambient particulates.

Capsule abstract (137 characters) – Systems scale implication for air pollution was assessed across three sectors of emerging green initiatives– energy, transport and ecosystem.

Keywords- air quality; bio energy; electric vehicles; green infrastructure; meso-scale; secondary aerosols; systems framework.

1. Introduction

Alongside conventional management of urban growth through efficient designing of the built form and transportation (so called "grey infrastructure"), developing green infrastructures is being considered as a cost-effective means for decoupling climate change impacts from urban sustainability (Pataki et al., 2011; Llausàs and Roe, 2012), primarily owing to their economy of scale and multi functionality (DCLG, 2007; CABE, 2010; TCPA, 2011). The concept of green infrastructure (GI) as a spatial planning tool has been adapted for quite some time (PCSD, 1999; McDonald et al., 2006), however, currently there has been re-energised emphasis on planning of 'garden cities' in the UK at a city-regional level (TCPA, 2011). The scope of greening is multi-faceted and extends beyond mere increase in green space cover to more unconventional measures, such as introduction of low/ zero emission transportation (electric/ fuel cell vehicles), production of renewable (so called 'green') energy from local resources, etc. For example, as part of the '2050 Vision for a Green Europe' sustainably grown domestic biomass is projected to provide up to 10% of the UK's energy needs by 2050 and significantly contribute to the reduction of greenhouse gas (GHG) emissions through penetration of low emission vehicles reaching up to 50% of the fleet (DECC, 2010). Furthermore, the UK Renewable Energy Roadmap places bioenergy at the forefront of the Government's plans to meet the Renewable Energy Directive objectives in 2020 (DECC, 2012).

Almost all the proposed green initiatives entail air quality implications on a systems scale which need addressing. Studies suggest that controlling air quality, especially in urban areas, will become more difficult in the future and under scenarios of climate change than it is now (Steiner et al., 2006; Nagendra et al., 2012), mainly owing to exacerbation of the local effects of climate change on meteorology, energy, emissions, photochemistry, and air quality. Currently (2012) there is a knowledge gap in ascertaining the interplay between different anthropogenic and biogenic components of green initiatives to ensure sustainable development through amelioration of local (and regional) air quality while minimising climate change impacts. Although piecemeal assessments of air quality implications from conventional biomass processing technologies (Tiwary et al., 2010) and adoption of zero-carbon transport technologies supported on fossil-based electricity grid (Williams, 2007) do exist, future implications for local air quality of enhanced greening of the entire urban form is not yet fully understood at systems level from landscape interactions perspective (Taha, 2008; Fowler et al., 2009). Evaluation of the 'true sustainability potentials' of combining these green initiatives therefore requires a paradigm shift in scoping of both the direct and the second-order environmental impacts, adequately quantifying the contributions from inherent anthropogenic and biogenic components using an integrated 'whole systems' approach.

Until recently simulations studying the implications of climate change adaptation strategies (in terms of urban surface modifications) have mainly evaluated the ozone impacts (Taha 2008) whilst the potential impacts on aerosol formation have received less attention, apart from a recent study on continental scale modelling of anthropogenic-biogenic emissions interactions (Sartelet et al., 2012). Besides, the impacts of the proposed control strategies, under future-year emission scenarios of climate change, are little understood. This paper takes a novel approach to systems analysis by assessing the landscape interactions between the anthropogenic and biogenic components of emerging green initiatives, essentially envisaged on the basis of plausible future scenarios. The assessment is mainly confined to pollutants that are currently of particular concern, including both primary and secondary particulate matter (PM, considered here as the combined pool of PM_{10}, $PM_{2.5}$ i.e. particles with aerodynamic diameters <10 µm and <2.5 µm respectively), ozone (O_3) and nitrogen dioxide (NO_2). The first part provides an overview of the assessments framework, highlighting the key components of green initiatives scoped in this study, alongwith their air quality implications. The second part demonstrates its application to a case study, through the development of representative scenarios using appropriate land use and emissions data sources. The subsequent parts provide the methodological approach adopted, followed by a discussion of the results and their policy relevance.

2. Materials and methods

2.1 Assessment framework

A theoretical framework is developed for systems scale assessment (**Figure 1**), encompassing plausible combinations of emerging green initiatives that will be implemented over the next 10-20 years (around 2020/30 horizons). It draws together the evidence-base from available literature on cross-disciplinary climate change and urban sustainability research, applying a cross-sectoral approach to three broad categories of green initiatives, including – a) use of vegetation; b) low-emission personal transport; and c) renewable energy from biomass. Each initiative (shown in boldface text) is characterised by a set of positive and negative environmental impacts (shown in italicised text) with their resulting air quality implications. These depend on the activities involved and their influence on either formation or removal of air pollutants. This was considered as an essential first step towards scoping the systems framework of landscape interactions between biogenic emissions, primarily biogenic volatile organics (bVOCs) and the anthropogenic emissions from fossil-fuel combustion (mainly from transport and energy sources) in future Green Cities. This step informed the subsequent modelling exercise in evenly incorporating both the positive and the negative effects in order to assess the overall sustainability implications of the green initiatives.

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Enhancing the Green Infrastructure, through planting vegetation in open spaces and on buildings (and rooftops), is expected to lower the air temperature and ameliorate the urban heat island effects, with the biological activity of plants and the surface albedo playing a crucial role (DCLG, 2006; Susca et al., 2011). However, the implication for air quality is considered to be heavily dependent on the species planted. Large scale commercial plantations of bioenergy crops such as poplar and willows, whilst fulfilling the current drive for energy sustainability from renewable biomass (Karp and Shield, 2008; Lovett et al., 2009; DECC, 2012) would exacerbate the bVOC (mainly isoprene and monoterpene) emissions (Williams, 2007). Furthermore, stress-induced bVOC emissions under an aggressive climate can enhance photochemical secondary organic aerosol (SOA) formation (Kiendler-Scharr et al., 2011). On the other hand, efficient surface modification strategies using, for example, urban reforestation of low- or no-bVOC emitters can produce net improvements to the air quality (Taha, 2008; Morani et al., 2011).

Uptake of greener transportation technologies, through a combination of low-emitting internal combustion and electric/fuel-cell traction, is projected to reduce primary emissions of CO, NO_x and PM from vehicle use. However, such initiatives can be considered green only to the extent that the source of energy supply is renewable. Fuel cell powered vehicles may still be associated with pollutant emissions in peri-urban regions if the hydrogen is generated by fossil fuel sources, which would contribute to additional aerosols from atmospheric reactions of SO_2, NO_x, NH_3 and VOCs, originating from the refineries. Incorporating biomass into the future energy mix is meant to keep the decarbonised energy generation affordable but at the cost of air quality (DECC, 2012). Systems level assessments of different fuel mixes have found increased N_2O, NH_3 and primary PM from the harvest phase and enhanced NO_x, CH_4 and secondary aerosol (SA) formation potentials from the combustion phase (Ciambrone, 1997; Tiwary et al., 2010).

The effect of land cover modification on ambient temperature is another key driver to energy demand (for cooling/ heating) and corresponding pollutant emissions (both in terms of primary components from associated activities as well as the photochemical precursors, i.e. chemicals that lead to tropospheric ozone production). While climate change effects are projected to contribute to aggravation of the urban heat island effects (and an overall increase in ambient temperature), specifically for the business as usual cases in built-up areas (Iamarino et al., 2011), lower air temperatures, resulting from vegetation cover modifications and retrofitting initiatives to enhance evapotranspiration and albedo-effects have shown reduction in cooling electricity demand (Susca et al., 2011) and reduced meteorology-dependent emissions from anthropogenic and biogenic sources (Williams, 2007). This in turn would contribute to reduced rates of ground-level O_3 formation and/or accumulation (Sartelet et al., 2012). However, potential increases in O_3 (negative impacts) can still arise from a combination of conditions that allow this pollutant to accumulate. Chemistry and atmospheric carrying-capacity aside, these include reduced mixing and dilution caused by lower winds and decreased boundary-layer depth (Taha, 2008). The latter would have further indirect impact on modifying local pollution concentrations.

2.2 Case study - site and scenario descriptions

The North East England has been used to demonstrate the application of the proposed meso-scale assessment framework, comprising of several cities and satellite market towns. **Figure 2** shows the spatial context of the region relative to the national boundaries, with the land cover map of Newcastle city centre in the inset, indicating the availability of green space towards the north with large potentials for greening of open spaces and marginal land. The region, consisting of four counties – Northumberland, Tyne and Wear, Durham and Tees Valley, is characterised by marked variations in land use, ranging from large patches of open areas in the north to built-up commercial/residential areas in the centre and heavy industries in its south. This serves as an ideal test bed for assessing the air quality implication of landscape interactions of the constituent anthropogenic and biogenic components. Pertinent to the greening of the transport fleet, the Tyne and Wear County of the North East is currently witnessing a huge emphasis on the promotion of low-carbon electric vehicles through the UK government funded ‘Plugged in places’ scheme (DfT, 2011). Parts of the region also have been earmarked for strategic plantations to provide renewable resources for decentralised energy generation in local biomass plants (either already in operation or currently pending planning approval) (Defra, 2009).

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Based on the assessment framework described earlier, the overall aim of the case study is to understand the systems scale interactions of anthropogenic and biogenic emissions from plausible green initiatives in three cross-disciplinary strands, comprising of the following activities – a) Energy (biomass co-firing power plants, district heating boilers); b) Transport (emissions reductions from conventional combustion technologies and penetration of low-emitting vehicles) and c) Ecosystem (a mix of bVOC active/inactive vegetation, including grassland/ woodland/ mixed forest mosaic). Two scenarios are developed to assess the air quality implications of land use changes towards future greening up to 2020, reflected in all three strands. – i) Baseline_2020, considers the baseline assumptions for 2020 emissions using the government forecasts from policy implementations (Defra 2011); ii) Aggressive_2020, includes enhanced emissions from bioenergy cropping on available land in the case study area and considers the associated life cycle emissions - from harvest, transport through to biomass combustion for energy generation.

2.3 Data preparation

2.3.1 Land cover

In the Baseline_2020 scenario the default information from Land Cover Mapping (LCM, 2007) data is applied. For Aggressive_2020 the LCM data is modified, reflecting projected greenspace plantations in the North East up to 2020, based on the energy cropping feasibility assessment (Defra, 2009). The projected yield estimates for Short Rotation Coppice (SRC) and Switchgrass at 5km resolution, published by the Government (**Figure 3**, left and middle panels), are applied to obtain the effective greenspace cover for 2020 (**Figure 3**, right panel). It involved overlaying the energy cropping data over the baseline LCM vegetation features in a geographical information system (GIS) map layer to estimate the percentage ground cover of greenspace for each ward.

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The yield estimates have taken into account potential impact of climate change, consistent with the 2020 scenario of the UK Climate Impact Programme (UKCIP) from established cropping in deep moisture retentive soils, assuming drier summers with rainfall less evenly distributed throughout the year. It is assumed that SRC willow (*Salix viminalis x*) be planted in the inner city and parklands whereas Switchgrass (*Miscanthus giganteus*) be planted on idle/marginal and arable land following the recommended best practice (Gallagher, 2008; Lovett et al., 2009). The location of the proposed and operating biomass power stations shown in **Figure 3** are obtained from the National Infrastructure Planning portal (shown as large red circles), whereas the locations of the community boilers are limited to the three higher educational institutions in the Newcastle city-region (shown as small dots).

2.3.2 Emissions

The Baseline_2020 scenario is developed for projected point and area source emissions up to 2020, obtained from the National Atmospheric Emissions Inventory, UK (NAEI, 2012). These cover the National Emissions Ceiling Directive (NECD) pollutants - nitrogen oxides (NAEI provides estimates of NO_x as NO_2), sulphur dioxide (SO_2), and ammonia (NH_3) (**Figures 4 a-c**; expressed as annual tonneNitrogen (tN) or tonneSulphur (tS) per square kilometre, t km^{-2} yr^{-1}**)**. These emissions estimates are based on the Updated Energy Projections, UEP43 (DECC, 2012) and have taken into account the impact of policy instruments affecting the emissions over this period, including penetration of electric vehicles, reduction of emissions from conventional diesel and petrol vehicle fleet, modification to agricultural emissions from technological advancement in processing and non-road transport. For Aggressive_2020 the emissions inventory include the following additional emissions – biogenic: bVOCs and NH_3 to the local environment from biomass cultivation; anthropogenic: NO_2, PM, SO_2 from harvest, processing and haulage of the biomass as well as the stack emissions from biomass burning in the power plants (**Figure 5 a-c** respectively, in terms of the same units as used for Baseline_2020**)**.

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The activities include three power stations, sited in the suburbs (Porth Blyth, Tyne and Lynemouth plants with respective capacities of 100, 295, 420 MWe, feeding on a 50:50 fuel mix of SRC woodchips and switchgrass, using a circulating fluidised bed combustor) and three community-scale boilers, located at each of the three higher educational institutes in the city of Newcastle upon Tyne, assumed to be cofiring gas and woodchips. The power stations and the community boilers are assumed to be in operation for 90% and 60% of the year respectively; the power stations are considered to be operated for the majority of the year while the community boilers were assumed to be operated mainly during the winter months. The biogenic emissions are based on the productivity estimates reported in the regional biomass yield maps (Defra, 2009). The anthropogenic emissions are estimated for biomass storage and utilisation; haulage and off-road vehicle transportation of fuel; combustion in biomass plants (Tiwary et al., 2010; NAEI, 2012). It is envisaged limited supply of locally sourced biomass, at least in the near future, will push a significant rise in import of the fuel in the region from overseas (or from elsewhere in the UK) via ship (RES, 2010). This may further enhance the associated anthropogenic emissions (sulphur and active nitrogen in the atmosphere) (Dore *et al.*, 2007). However, in this study offshore transport is not included and all the biomass is assumed to be sourced locally via ground transport. The bVOC emissions from the rest of the vegetation cover for the two scenarios have been estimated for isoprene and monoterpenes on the basis of the land cover data (as detailed in Section 2.4.2).

2.4 Anthropogenic-biogenic interactions modelling

A two-stage modelling was adopted - first, a detailed spatially-resolved approach to understand the implications of anthropogenic-biogenic interactions of the sources and sinks, using the emissions inventory prepared for the North East Region (**section 2.3.2**) and second, a more strategic approach, essentially up scaling the trends from the regional emissions for the reference years to understand their national implications on pollutant formation and removal, specifically for secondary pollutants, ozone and aerosols. A brief survey of available tools capable of modelling meso-scale anthropogenic-biogenic interactions was carried out to establish their merits and limitations. Based on previous applications and capabilities two tools, namely FRAME and WRF/Chem, were adopted for the respective tasks outlined above. The complementary capabilities of these two models enabled quantification of pollutant concentrations, both from altered anthropogenic/biogenic activities and the associated land cover changes, as follows - the FRAME set up allowed spatially-resolved estimation of the secondary inorganic aerosols and PM_{10} deposition to vegetation surfaces; the WRF/Chem set up enabled the simulation of the meteorological variables and the photochemical transformation of pollutant precursors for estimation of tropospheric ozone and secondary organic aerosols. A brief overview of the two models is presented below.

2.4.1 FRAME

The FRAME (Fine Resolution Atmospheric Multi-pollutant Exchange) model was initially developed specifically to simulate the concentration and deposition of ammonia (Singles et al., 1998). Subsequently it was modified to be a multi-pollutant model, including detailed treatment of oxidised nitrogen and sulphur (Fournier et al., 2004) and applied to estimate acid deposition and the exceedance of critical loads. FRAME offers a spatially-resolved mechanism for modelling future emissions scenarios from altered land cover as well as for capturing the photochemical interactions of precursors contributing to secondary pollution formation (Dore et al., 2007). FRAME is capable of estimating the ‘net’ concentrations at a meso-scale by accounting for the distributed fluxes of pollutants to land and vegetation surfaces, which is relevant to understanding the systems implications from green initiatives scoped in this study. FRAME was successfully applied to – a) Estimate the secondary pollution formation potential from a biomass plant system (Tiwary et al., 2011); b) Assess the exceedance of critical loads for nitrogen and acid deposition (Matejko *et al.*, 2009), and c) Estimate ammonia concentrations and deposition of reduced nitrogen in the North China Plains (Zhang *et al.*, 2011). In this study, the focus is on the reaction of acid gases (H_2SO_4 and HNO_3) with ammonia to form secondary inorganic aerosols (SIAs, ammonium sulphate and ammonium nitrate). The high resolution (1 km) version of the FRAME model, developed by Hallsworth *et al.* (2010), has been applied in this study to set up the regional model. The emissions data from the National Atmospheric Emissions Inventory (http://naei.defra.gov.uk/) are gridded into sector-dependent vertical model layers for the 11 sectors proposed by SNAP (Selected Nomenclature for Air Pollution) for NO_2, SO_2 and NH_3. Point source emissions are treated individually with a plume rise model (Vieno *et al.*, 2010) using stack parameters (temperature and exit velocity of emissions and stack height and diameter). Vertical diffusion in the air column is calculated using K-theory eddy diffusivity and solved with the Finite Volume Method. Deposition of NO_2, SO_2 and NH_3 is calculated through specific parameterisation for common landscapes in the region – including, forest, moorland, improved grassland, arable, urban and water.

A further development of the model code for this study included extending the existing canopy resistance formulation of dry deposition of gases to aerosol particles. This scheme uses a 'big-leaf resistance' analogy model for dry deposition of particles (Smith et al., 2000). The first term in the resistance analogy concerns the transport of particles from the well-mixed planetary boundary layer to the immediate vicinity of the surface and is controlled by turbulent diffusion according to the wind velocity and aerodynamic roughness of the surface. The second term concerns the transport of molecules through the viscous, quasi-laminar boundary layer of air close to the surface by diffusion and depends on the physical properties of the particles. The third term in the resistance analogy is dependent on the gravitational settling velocity which is a function of particulate mass. This resulted in vegetation specific deposition velocities for fine (ammonium nitrate and ammonium sulphate aerosol) and coarse (large nitrate aerosol) particulates, permitting calculation of the effect of increased forest cover on removal of particulate matter from the atmosphere by dry deposition.

2.4.2 WRF/Chem

The meso-scale emissions interactions were evaluated from the online Weather Research and Forecasting model coupled with Chemistry (WRF/Chem) (url: http://www.acd.ucar.edu/wrf-chem). WRF/Chem was considered adequate, having been used extensively for studying interactions of bVOC emissions with anthropogenic emissions (Eder et al. 2005, Carmichael et al. 2009; Zhang et al 2010) and the model outputs have been statistically evaluated against observations, specifically for regulated air pollutants, and found to be in good agreement (Grell et al., 2005; Zhang et al., 2010; Hu et al., 2012). The air quality component of the model is fully consistent with the meteorological component; both components use the same transport scheme (mass and scalar preserving), the same grid (horizontal and vertical components), and the same physics schemes for subgrid-scale transport (Grell et al., 2005). The components also use the same time step, hence no temporal interpolation is needed.

Physics options chosen for the simulations include the Dudhia shortwave radiation algorithm, the rapid radiative transfer model (RRTM) for longwave radiation, the WRF Single-Moment 6-class (WSM6) microphysics scheme, the Noah land-surface scheme, and the Yonsei University (YSU) planetary boundary layer (PBL) schemes. The National Centres for Environmental Prediction (NCEP) Final (FNL) Global Forecast System (GFS) operational analyses were used for the initial and boundary conditions of all meteorological variables. Chemical options chosen for the simulations included the Regional Atmospheric Chemistry Mechanism (RACM) gas-phase mechanism (Stockwell et al., 1997), Modal Aerosol Dynamics Model for Europe (Ackermann et al., 1998) with secondary organic aerosols incorporated into the module through the Secondary Organic Aerosol Model (MADE/SORGAM) (Schell et al., 2001) aerosol parameterisation, the Wesley (1989) dry deposition method. The Initial and boundary conditions for the chemical species were extracted from the output of the global model MOZART4 (Emmons et al., 2010). The anthropogenic emissions dataset (CO, NO_2, SO_2, NH_3, PM_{10}, $PM_{2.5}$) for the United Kingdom was sourced from the NAEI 2020 projections and structured in the format prescribed by the Emission Database for Global Atmospheric Research (EDGAR) system (http://www.mnp.nl/edgar). Based on the land cover information for the two scenarios the model computed the bVOC emissions using established schemes (Guenther et al., 1994; Chung et al., 2011). **Table 1** shows the species-specific emission potentials for the trees considered in this study under standard, daylight regime for temperate climate suiting the UK, drawn from the literature (Owen et al., 2003, Stewart et al., 2003). The default (Guenther) scheme in WRF/Chem allocates the monoterpenes emission to isoprene (Grell et al., 2005; Fast et al., 2006) because it is only coupled with the gas-phase mechanism of the Regional Acid Deposition Model (RADM) that does not include monoterpenes.

It is understood that all aspects of meteorology change, in varying degrees, as a result of surface modifications; however, air temperature has been used as a suitable index, given its relevance to urban heat island mitigation. Following earlier work (Taha, 2008), air temperature difference from the two scenarios was assessed at 2m height at 1100hrs.

3. Results and discussions

3.1 Quality assurance

As a first step, the model performance of the aerosols outputs was benchmarked against monitored data from the UK Acid Gases and Aerosols Monitoring Network (AGANet) for secondary aerosols for 2008. The AGANet provides monthly speciated measurements of ambient air pollution (including aerosol components NO_3^-, SO_4^{--}, Cl^-, Na^+, Mg^{++}, Ca^{++} and NH_4^+) as part of the UK Eutrophying and Acidifying Atmospheric Pollutants (UKEAP) network to provide temporal and spatial patterns and trends and to provide a long-term dataset for comparing results with dispersion models (Defra, 2008). Annual average concentrations from 25 sites for NO_3^- and SO_4^{--} aerosol were compared with modelled outputs (**Figure 6**).

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Overall, performance of the model showed good agreement with measurements for both NO_3^- and SO_4^{--} (R^2 values of 0.92 and 0.69 respectively), with slight over prediction in general in both cases at higher concentrations. **Table 2** shows the comparative statistics for SO_4^{--}, NO_3^- and NH_4^+; a stronger agreement of the model estimates with AGANet measurements for NO_3^- and NH_4^+ is confirmed on the basis of R^2 values of over 0.9 in both cases. On the other hand, the FAC2 analysis (i.e. measure of the number of sample points that fall below or above 1:2 and 2:1 lines) shows a wider scatter of values for NH_4^+. There seems to be a tendency for underestimation of the annual NO_3^- and NH_4^+ concentrations, indicated by negative normalised mean bias values. Also, the lower normalised mean gross error (NMGE) and root mean square error (RMSE) values for NO_3^- and NH_4^+ compared to SO_4^{--} indicate the better agreement of the estimates for the first two and less so for the latter.

Recent estimates from an enhanced version of the UK photochemical trajectory model for secondary inorganic aerosols (SIAs) have reported over predictions of Cl^-, NO_3^- and SO_4^{--} while comparing with AGANet measurements (Beddows et al., 2012), mainly attributed to differences in the chemical and the meteorological processes included in the model from those of the air masses sampled.

3.2 Anthropogenic-biogenic interactions

It is noteworthy that the scope of anthropogenic-biogenic interactions presented here is geared to capturing the overall patterns of plausible emissions scenario and therefore ought to be more impressionistic rather than precise at all levels. The results presented essentially account for the altered anthropogenic and biogenic emissions profile from biomass harvesting and processing as well as their utilisation in local power stations to support the increased energy demand for electric vehicle infrastructure over the period. The spatial mapping of the resulting SIAs over the North East model domain (**Figure 7**) shows an increase in concentration for the Aggressive_2020, with prominent increases in the lower and lower-middle parts, marked with high population and industrial activities. This is along the lines of earlier findings suggesting secondary aerosols, formed from precursor emissions of SO_2, NO_x and NH_3, constitute a significant fraction of $PM_{2.5}$ in ambient air (Tiwary et al., 2011; Ciambrone, 1997). Such situations may pose potential risks of adverse human heath implication from enhanced exposure to fine particles and poor visibility over the years.

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The relative changes (in percentages) for the Agrressive_2020 scenario over the Basline_2020 for the study region is shown in the bar chart (**Figure 8**). It illustrates the overall impacts (i.e. net effects) for the parameters studied (black), calculated as the differences between the estimated increases (pitted white) and the estimated reductions (pitted black) in the respective parameters (the latter was only estimated for NO_2 and PM_{10}). These estimates cover the three components of greening considered - enhanced green infrastructure development (including biomass harvesting); green transport; renewable energy initiatives. They essentially involve a combination of emissions sources of primary pollutants and precursors of the secondary pollutants as well as emissions sinks from enhanced deposition associated with land cover changes (based on the study framework, **Figure 1**). Only a marginal change in ambient temperature (estimated at 2m height at 1100hrs) of -0.05% is noted. However, ambient NO_2 is estimated to rise by up to 5% (net, i.e. discounting for the deposition to the increased vegetation cover, shown using the negative bar), apparently holding potentials for eroding the forecasted reduction of overall NO_2 by 2020 of as much as 30% (from 2010 levels) for the UK (NAEI, 2012). Also, bVOCs (limited to estimates for monoterpenes and isoprene) are found to rise by up to 20%.This is mainly attributed to the significant proportion of bVOC-active SRC willow (*Salix viminalis x*) plantations in the city-regions, serving the local bioenergy demands; the majority of trees species in the base case, primarily used for street plantations are bVOC-benign (see **Table 1**), viz. Sycamore maple (*Acer pseudoplatanus*), Lime (*Tilia cordata*), Turkish Hazel (*Corylus colurna*), Hornbeam (*Carpinus betulus*) etc.

The plot in **Figure 8** further shows enhanced daytime ozone and secondary aerosols (ultrafine particles, UFPs) formation by up to 15% and over 5% respectively, mainly from increased anthropogenic-biogenic emissions interactions between NO_2 and bVOC near large town centres. This is consistent with a recent study which found SOA formation potentials from baseline anthropogenic-biogenic emissions interactions in Europe to be relatively lower than in North America owing to less abundance of bVOC active species in Europe (Sartelet et al., 2012). However, this may significantly change with increase in plantation of isoprene- and monoterpene-active species in and around cities along side rise in anthropogenic emissions of NO_x from enhanced uptake of diesel cars.

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Table 3 provides regression statistics comparing the modelled outputs from the two scenarios. The strength of coefficients of determination (R^2) signify the inverse relation to the extent of modification of the respective environmental variable as a consequence of the interventions assumed in the aggressive scenario, i.e. smaller R^2 and p-values indicate greater relative changes from the base case. For example, based on the p-value statistics the most significant changes from these interventions would be observed in bVOC concentrations. For the rest of the pollutants this is shown to be varying moderately and there seems to be negligible influence on the ambient daytime temperature. This substantiates our findings from the simulations shown in **Figure 8** statistically, that of marginal change in ambient temperature and considerable changes for bVOC, NO_2, O_3 and particulates (both PM_{10} and Ultrafines).

<place Table 3 here>

The interactions between precursors are influenced by reaction time delays and therefore have more prominent national impacts on the resulting pollutant concentrations and a relatively small impact locally. The output from scaling up the regional emissions scenario to the national scale in the next step in WRF-Chem provided spatial mapping of the modifications to surface layer ozone (**Figure 9** shows the model outputs for the two scenarios at 1400hrs on 4th July, representing a typical summer afternoon). The simulated changes in ozone concentrations account for all positive and negative interactions of meteorology, emissions, and photochemistry, thus providing estimates of the net effects.

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Our results regarding the potential enhancement in O_3 and SAs in Aggressive_2020 case due to increased anthropogenic-biogenic emissions interactions are consistent with a recent study (Sartelet et al., 2012), which reported greater potential for O_3 and PM_{10} reductions near large town centres by removal of anthropogenic emissions whilst greater potential for SOA reductions by removal of biogenic emissions.

3.3. Effect of land cover changes

Land cover modifications have been strongly associated with potential reduction in air temperature over an area; the spatial distribution of such temperature dip matches closely with the nature of surface modifications and their distributions (Morani et al., 2011; Susca et al., 2011; Taha 2008). The land cover alterations assumed in this study, however, are mainly associated with enhancing the green cover from bio energy cropping, thus showing only marginal daytime temperature reduction (**Figure 8**). This is along the lines of earlier finding showing relatively smaller surface modifications having only minor impacts on air temperature, e.g., decreases of around 0.5 – 1°C (Taha, 2008). Nevertheless, this would still have implications for prolonging the photochemistry during the daylight hours, leading to alteration in the trends for syntheses of precursors and secondary pollutants.

An important effect of increasing the tree cover, shown in this study is that the uptake from vegetation has relatively small impact on reduction in PM_{10} and a much greater impact on reducing NH_3 concentrations (**Figure 10**). This is attributed to the modelling assumptions in FRAME, resulting in an order of magnitude higher dry deposition velocities for NH_3 to tree canopies than grasslands. This may also be of significance in relation to greening of urban areas due to recent increases in emissions of NH_3 from vehicle exhausts caused by the introduction of catalytic convertors. The lower deposition velocities for PM_{10} is because their removal from the atmosphere is primarily due to washout by precipitation, with dry deposition of aerosol particles estimated to contribute only approximately 5% of the total national deposition of sulphur and nitrogen to the UK (RoTAP, 2012). Although planting vegetation in urban areas in the UK is shown to be effective in reducing PM_{10} concentrations, attributable mainly to the higher deposition velocities of larger particles (McDonald et al. 2012), experimental studies have demonstrated that particulate deposition velocities vary by two orders of magnitude across the full range of sizes in the atmosphere (Seinfield and Pandis, 1998). The effect of urban greening on particulate matter concentrations through increased tree planting is therefore sensitive to the nature of the particle source. This has interesting consequences for understanding the implications for future provision of ecosystem services from plants, both in terms of human health benefits and potential loss of biodiversity through eutrophication.

<place Fig 10 here>

In a pertinent policy context for evaluating systems scale sustainability implications from land cover changes our study shows a clear need for monitoring not just regulated pollutants at urban hotspots but also the much illusive rural emissions of important precursors, attributable mainly to greening initiatives. This is deemed essential for understanding their regional contributions to ozone and secondary aerosol formation from photochemical interactions. Another important point to consider is to incorporate the emerging global trends in land cover changes while simulating future emissions scenarios. This is because while emissions in the UK and its immediate European neighbours is expected to decrease over the longer time scale, the relative (and in some cases the absolute) contributions from emissions in the rest of the northern hemisphere are likely to increase. Therefore, in assessing air quality in the UK, say for a 2050 scenario, adequate consideration is needed for hemispheric, if not global emissions (Williams, 2007). For example, increases in anthropogenic emissions from China and India have been shown to enhance urban air quality issues in the US (Zhang et al. 2010). This is also evidenced by a recent Review of Transboundary Air Pollution (RoTAP, 2012), which found background ozone concentrations to have grown by about 15% despite decline in peak ground level ozone concentrations by about 30% in the UK and continental Europe. This is primarily explained through increases in precursor emissions elsewhere in the northern hemisphere.

We acknowledge that an evaluation of systems scale sustainability implications, based on the outlined assessment framework, is subjected to uncertainties in both model assumptions and the underlying emissions data. A quantitative analysis of these uncertainties was considered too complex to be considered within the scope of our study. However, for the benefit of readers in interpretation of the results, some qualitative comments on the magnitude of the uncertainties of different factors as well as the combined impacts of different sources of uncertainty have been included here. As described from the outset, the assessment outcomes are primarily a function of emissions projections and hence subjected to several sources of uncertainties – including, forecast error in demand equations for the emissions predictions; uncertainty in future policy impacts; uncertainty in modelling assumptions of drivers of economic growth, energy and transport demands, etc.

Over the study period the uncertainty in NH_3 emissions is going to be associated mainly with two factors – one, alteration to agricultural practices, for example the switch in fertiliser use from ammonium nitrate to urea; two, varied intensities of green infrastructure development, for example extension (or replacement) of existing green space with bio energy cropping with higher rate of NH_3 cycling into the local environment. Non-agricultural sources, including higher emissions from lean-burning fuels and fuel-efficient petrol cars are expected to make only a small proportion of emissions and so assumptions on energy use would make little difference to NH_3 emissions (NAEI, 2012). On the other hand, the NO_2 emissions are strongly dependent on the assumptions made about the future transport and energy demands in the literature. For example, owing to uncertainties in energy efficiency measures and impacts of climate policies beyond 2030 significant increases in NO_2 emissions can still occur from higher natural gas use in power stations despite substantial reduction in fossil-based transport NO_2 emissions from switching to cleaner technologies. Uncertainties in the emissions of PM and bVOCs are mainly going to be driven by assumptions on land cover changes (for example development of Green Infrastructure) and fuel use in response to variation in energy demand, in particular the effect of renewable energy policy on the level of biomass uptake beyond 2020. It is envisaged, uncertainties in SO_2 emissions would be dominated by assumptions about the future levels of direct coal use, implementation of clean coal technologies and the operating characteristics of the Flue-gas desulfurization (FGD), in both power generation and industry (NAEI, 2012). In addition, there is also an uncertainty in SO_2 emissions from crude oil refineries with varying levels of sulphur.

4. Conclusions and future directions

The combined effects of emerging green initiatives, as scoped through the systems level assessment framework adopted in this study, show tendency to aggravate air quality issues, both at local/regional and national scales. The case study indicates rises of up to 5% and 20% in NO_2 and bVOC respectively for Aggressive_2020 over Baseline_2020 scenario. This has implications for enhanced formation of daytime ozone and UFPs by up to 15% and over 5% respectively. The integrated assessment framework demonstrates the need for whole-system thinking in ascertaining the sustainability of future land cover modifications, incorporating the green components while optimising the inter-dependence of the resource utilisation, emissions and photochemical interactions of precursors. Our study shows a clear footprint of secondary aerosols concentrations from increased biomass production and utilisation, which has a spatial distribution across the study area at relatively low levels of concentration (primarily attributed to the abundance of the precursors and the time scale required for chemical reactions to take place). Modified land cover results in enhanced deposition of N-compounds (NO_2 and NH_3) and particulates on taller vegetation more than on grasslands. Evidently, this has positive implication from human exposure perspective but adverse ecological implications to biota through eutrophication and potential loss of biodiversity. This presents interesting research questions worth investigating in the future.

Overall, this study presents the steps needed to evaluate systems scale interaction of anthropogenic-biogenic emissions from the green initiatives considered. However, wide scale adoption of the proposed approach is needed if the demands for delivering sustainability are to be met in the future. For example, there is a need to compare the impact of land cover change to local pollution source/sink generation as well as the influence on local micro climate from excessive reliance on imported bioenergy resources, electric vehicles and associated infrastructure. Consistent with other researchers' recommendation we re-emphasise that multi-episodic and seasonal evaluations are essential to assess the potential impacts of the proposed strategies, covering a range of meteorological and emission conditions, including average seasonal conditions (in addition to the worst-case scenarios). Another interesting dimension to take the work forward would be to assess the ecological and human health impacts from enhanced biomass production, namely a) Exacerbation of secondary aerosol concentrations, with consequential effect on visibility and human health; b) Enhanced deposition of oxidised nitrogen, affecting eutrophication and loss of biodiversity and urban ecosystem functions; and c) Alteration in the pollution profile for the whole urban-rural system, resulting mainly from increased haulage of the feedstock and penetration of low emission vehicles.

5. Acknowledgments

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Table captions

Table 1. Species-specific bVOC emission potentials (µg gdw^{-1} h^{-1}) for the trees considered in this study for standard conditions.

Table 2. Model performance metrics for aerosols output for 2008 [*N* = number of complete pairs of measured and calculated values; NMB = Normalised mean bias; NMGE = normalised mean gross error; RMSE = root mean square error (µg m^{-3}); FAC2 = A count fraction of points within 0.5 and 2 times the observation; R^2 = Coefficient of Determination].

Table 3. Comparison between Baseline_2020 and Aggressive_2020 outputs of representative environmental variables using regression statistics (with 95% confidence interval, *N* = 538) [R^2 = Coefficient of Determination; p-value = Probability measure of statistical significance; St dev = Standard deviation].

Figure captions

Fig 1. Theoretical framework illustrating the positive and negative implications for air quality from emerging green initiatives (2020/30 horizons).

Fig 2. Case study site – the North East region of England [note: the references to colour in this figure legend can be found on the web version of this article]. © Crown Copyright Ordnance Survey Map.

Fig 3. Estimated greenspace cover for the North East England (percentage greenspace at ward-level) for Aggressive_2020 scenario (right panel). [note: the left two panels show the yield maps for short rotation coppiced (SRC) and miscanthus plantations]. © Crown Copyright Ordnance Survey Map.

Fig 4. Emissions for Baseline_2020 scenario based on projected emissions– (a) NO_2; (b) SO_2; (c) NH_3.

Fig 5. Additional emissions for Aggressive_2020 scenario from biomass harvest, transportation and combustion – (a) NO_2; (b) SO_2; (c) NH_3.

Fig 6. Benchmarking model output against the monitored annual average aerosol concentrations from AGANet.

Fig 7. Spatial mapping of secondary inorganic aerosol (SIA) formation potentials over the North East England case study area from FRAME [note: the references to colour in this figure legend can be found on the web version of this article].

Fig 8. Systems scale implications of green initiatives from Aggressive_2020 compared to Baseline_2020 (expressed as percentage change).

Fig 9. UK map for alteration in tropospheric ozone concentrations from Aggressive_2020 compared to Baseline_2020 [note: the references to colour in this figure legend can be found on the web version of this article].

Fig 10. Attenuation in pollution concentration in North East England from enhanced uptake by vegetation cover for Aggressive_2020 compared to Baseline_2020 from deposition processes (both wet and dry) – (a) PM_{10}; (b) NH_3.

Figure 1

Negative	Green initiative	Positive
• *Greater foliage* → more bVOCs → more ozone → more aerosolisation • *More fertilisation* → more NH_3, N_2O	**Green Infrastructure*** (greening built-up areas, open spaces/ parklands)	• *Lower air temperature* → less bVOCs → less aerosolisation • *Higher dry deposition* → less PM_{10}, NH_3, NO_2
• *Higher fossil fuel use on urban/rural fringes#* → more NO_2, SO_2, HCl → more aerosolisation	**Greener transport** (uptake of Electric/ Fuel cell vehicles)	• *Lower fossil fuel use in cities* → less NO_2, CO_2, CO, NH_3, VOCs
• *Biomass harvesting*, transportation, processing, combustion* → more NO_2, PM_{10}, CH_4	**Bio energy** (generating energy from locally sourced biomass)	• *Lower fossil fuel use* → less CO_2

** includes positive and negative air quality implications from Green Infrastructure development.*

 # assumes combustion-based electricity generation from bioenergy resources and hydrogen production from fossil fuels.

Figure 2

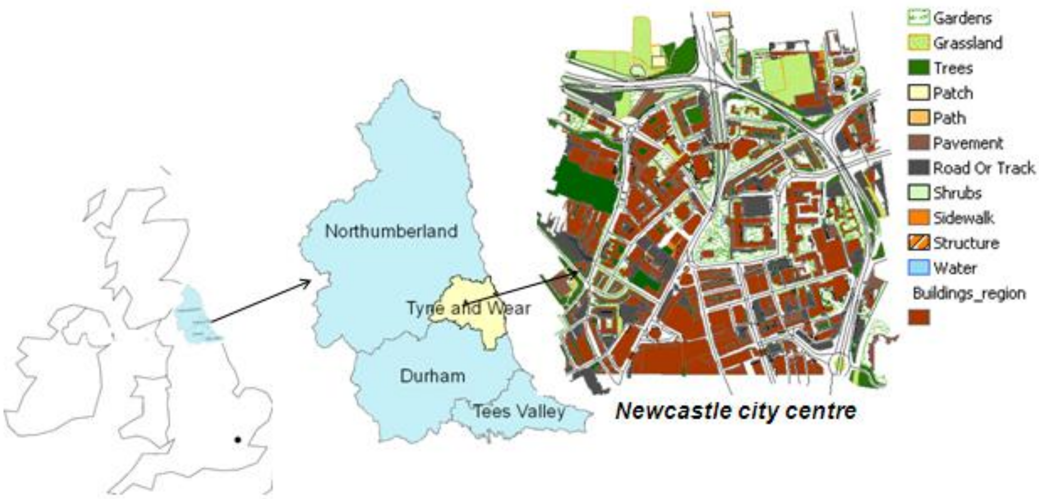


Figure 3

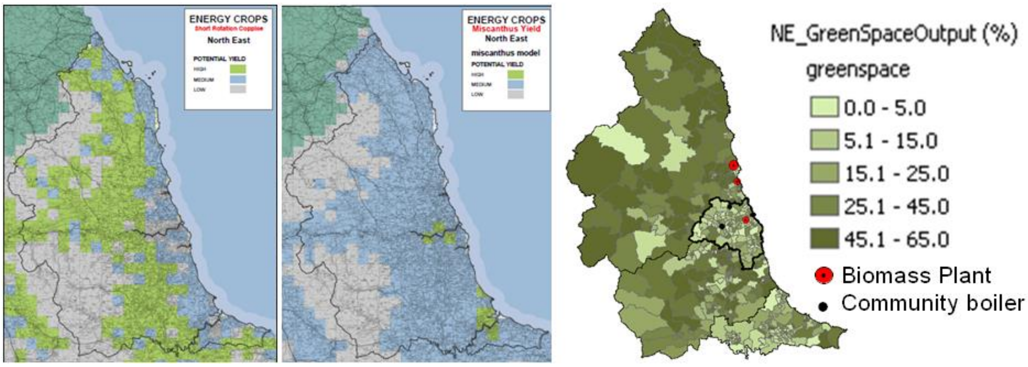


Figure 4

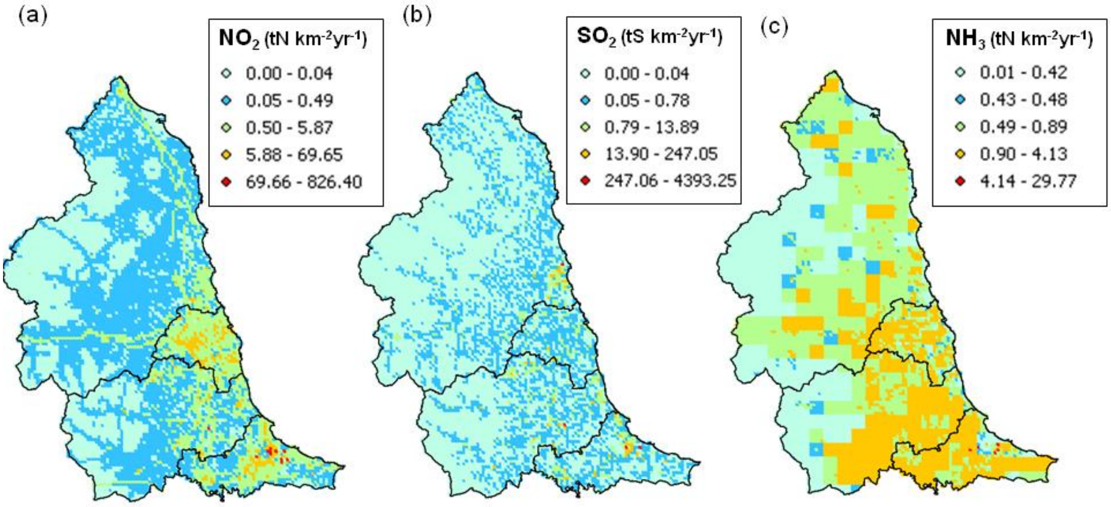


Figure 5

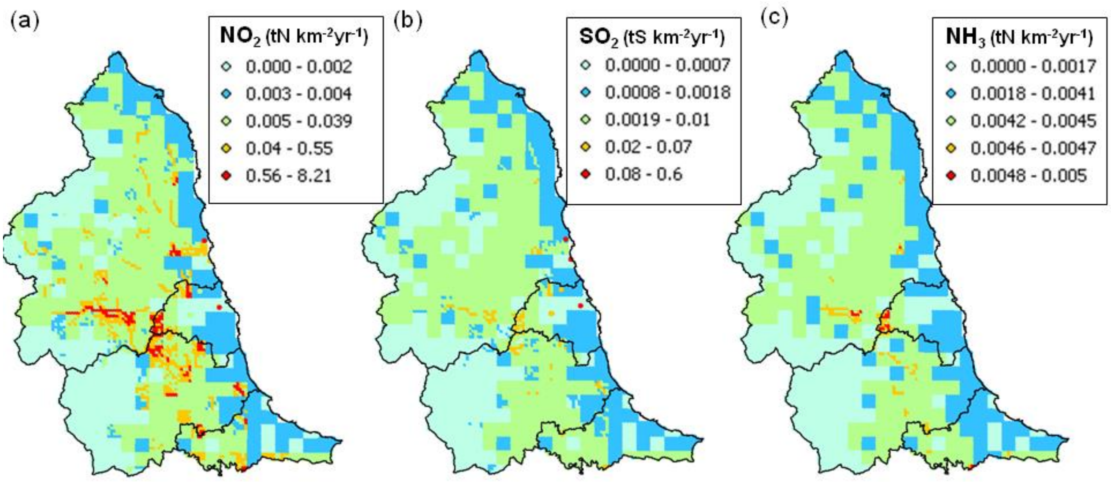
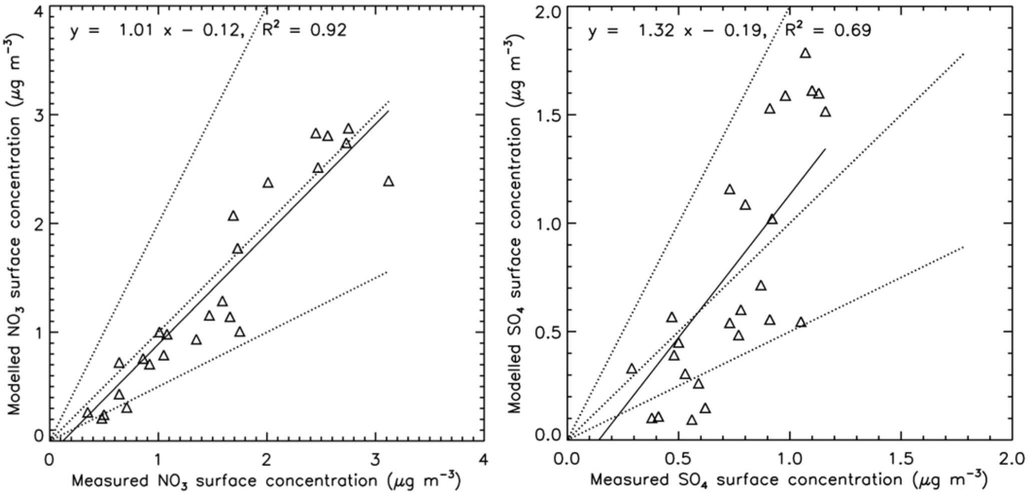


Fig 6



I. NO_3^{1-} II. SO_4^{2-}

Fig 7

I. Baseline_2020 II. Aggressive_2020

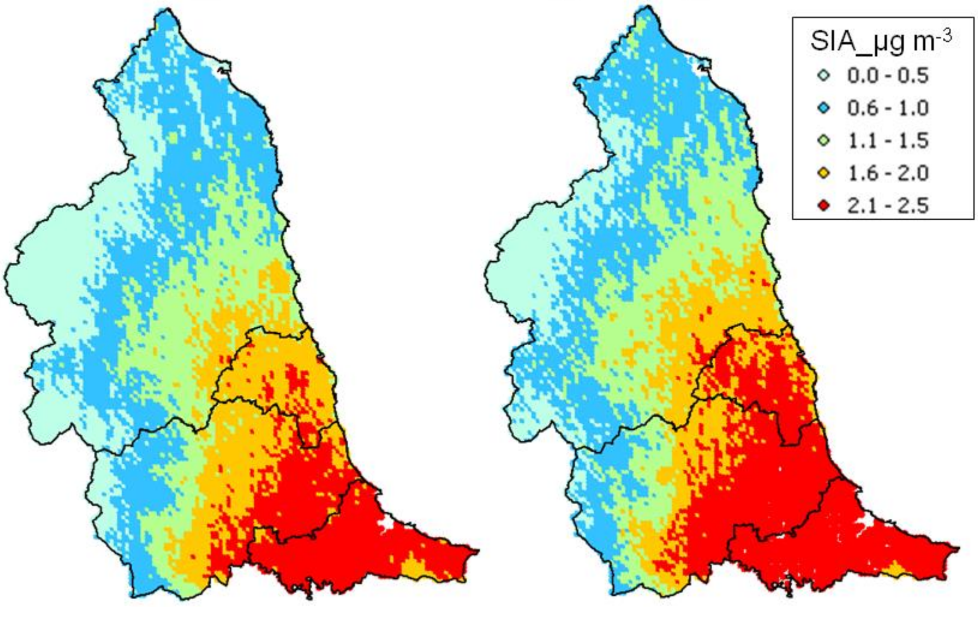


Fig 8

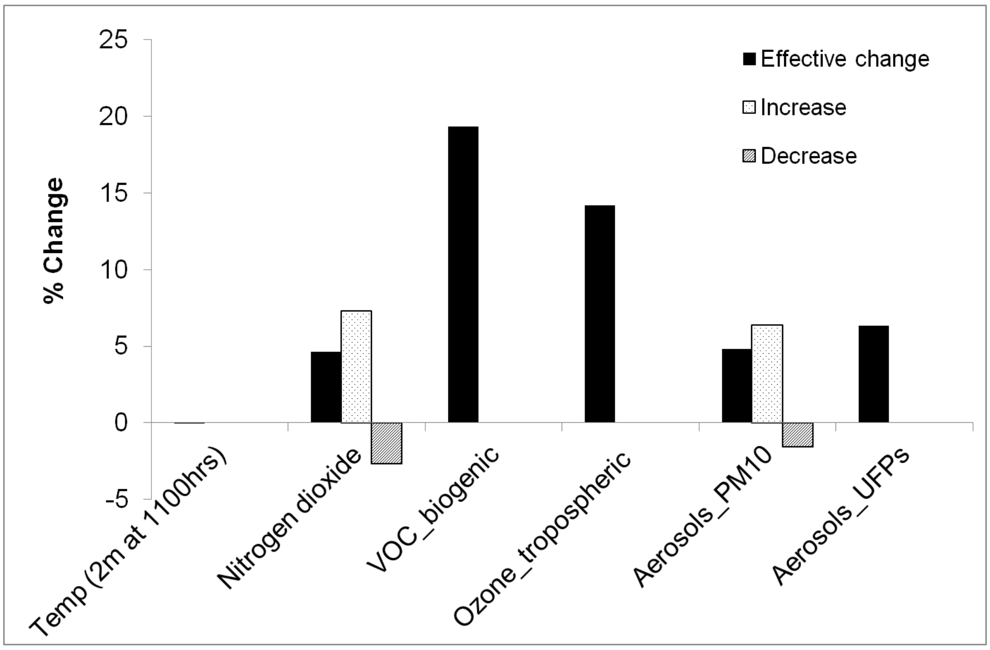


Figure 9

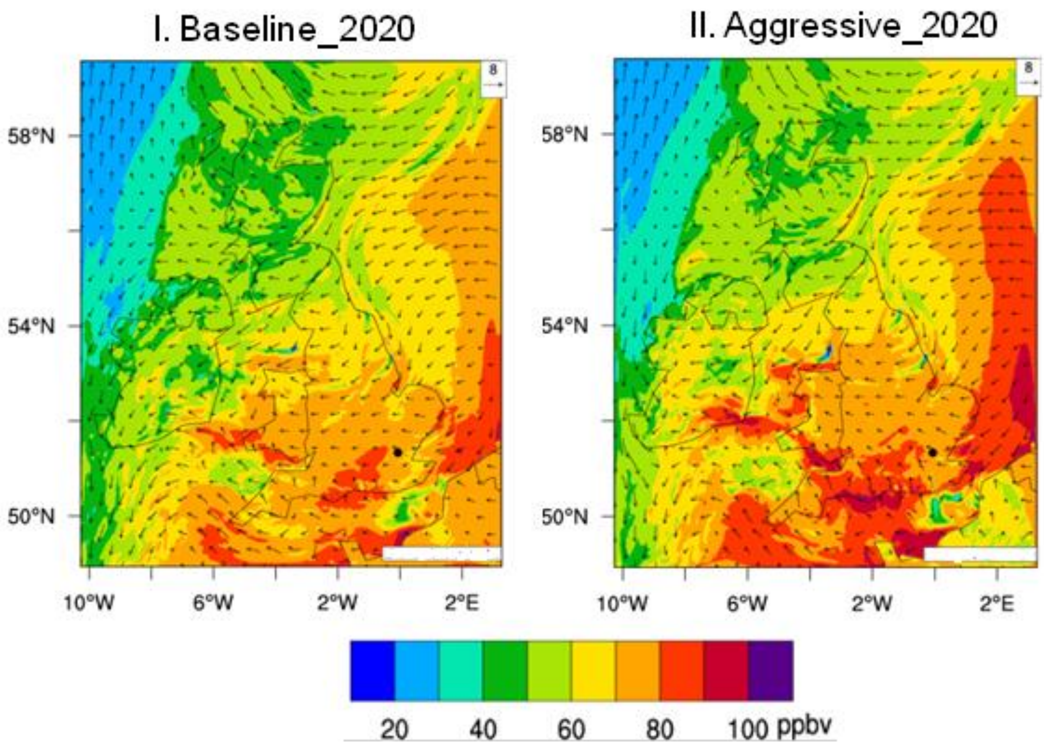
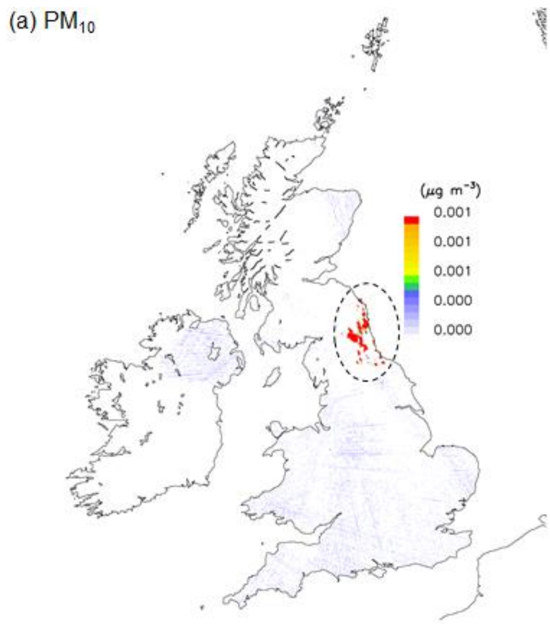


Fig 10


(a) PM_{10}
($\mu g\ m^{-3}$)
0.001
0.001
0.001
0.000
0.000
(b) NH_3
($\mu g\ m^{-3}$)
0.013
0.010
0.006
0.003
0.000