SPATIAL AND CHEMICAL PATTERNS OF PM$_{2.5}$ – A CASE STUDY FOR THE UNITED KINGDOM AND POLAND

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Abstract: The Fine Resolution Atmospheric Multi-pollutant Exchange model was used to calculate the mean annual concentration of PM$_{2.5}$ at a resolution of 5km x 5km for the UK and Poland for the year 2007. Area and point source anthropogenic emissions for the UK were taken from the NAEI database. For Poland point source emissions, including information about stack parameters, was delivered by the National Administration of the Emission Trading Scheme (KASHUE). Area source emissions were calculated using a top-down approach, based on information on national total emissions delivered by KASHUE, which was disaggregated spatially using different types of proxy data. Natural wind blown dust emission for both countries was taken from the NATAIR project.
Modelled average PM$_{2.5}$ concentration is higher for Poland than the UK and amounts to 9.2 µg m^{-3} and 5.6 µg m^{-3}, respectively. The highest concentrations concern London and coastal areas for the UK and urban agglomerations in the case of Poland. Maximum values occurring close to the UK coastline can reach 18 µg m^{-3} and are related to high levels of sea salt aerosol (SSA). The average contribution of natural particles amounts to 34% and 20% of total PM$_{2.5}$ concentration, respectively for the UK and Poland. Among anthropogenic particles for both countries the highest contribution falls on secondary inorganic aerosols (SIA) and the lowest contribution is for secondary organic aerosols (SOA). In the case of natural particles there is a higher difference in compounds contribution between the UK and Poland. For the UK, primary particulate matter dominates in natural PM$_{2.5}$ concentration (63%). SSA contributes 84% of natural PM$_{2.5}$ for the UK and only 3% in Poland. In total (anthropogenic+natural) concentration of PM$_{2.5}$ dominates SIA (about 50%) for both countries.

Key words: PM$_{2.5}$, concentrations, United Kingdom, Poland

INTRODUCTION

Inhalable particulate matters can penetrate into the human body and cause serious health problems. Most of the particles with diameter above 5.0 µm are filtrated in the nasal cavity, whilst the most dangerous are the fine fraction with diameter below 2.5 µm (Pope et al., 1995). Both anthropogenic (e.g. transport sector, industrial processes, power generation) and natural emissions (e.g. sea spray, wind blow dust) as well as meteorological or climatic factors contribute to the occurrence of high values of particulate matter (PM) concentrations (Basart et al., 2012, Kryza et al., 2012). It is essential to know what is the contribution of different emission sources because when PM concentrations exceed the limit values specified by Air Quality Directive 2008/50/EC and are related with natural events, Member States are obligated to inform the Commission, providing the necessary justification that such exceedances are due to natural events (European Commission, 2011). The concentration and chemical composition of PM can be influenced by processes on a regional scale as well as by long-range pollution transport. Application of atmospheric chemical transport models enables better understanding of the spatial and temporal distribution of particles. Here we use the Fine Resolution Atmospheric Multi-pollutant Exchange model (FRAME) to provide spatial distribution and chemical composition of PM$_{2.5}$ concentrations for two countries in Europe (the United Kingdom and Poland), which are diverse in terms of emission structure as well as climate.

DATA AND METHODS
FRAME model
The statistical trajectory model FRAME is used here to assess the annual mean concentrations of PM$_{2.5}$ for the UK and Poland for the selected year 2007. The fundamentals of the model are described by Fournier et al. (2004), Dore et al. (2007), Vieno et al. (2010) and, for the FRAME model for Poland, by Kryza et al. (2012). FRAME is used here with a grid resolution of 5km x 5km and grid dimensions of 172 x 244 cells for the UK and 160 x 160 cells for Poland. The import of pollutants from outside of both domains (the UK and Poland) is calculated with FRAME-Europe – a similar model to FRAME, which runs for the entire Europe on the EMEP grid at a 50km x 50km resolution. To get the total concentration of PM$_{2.5}$, the FRAME model was run for: secondary inorganic aerosols (SIA: SO$_4^{2-}$, NO$_3^-$, NH$_4^+$), primary particulate matter (PPM$_{2.5}$), sea salt aerosol (SSA, particles below 2.5 um). The contribution of particles at a diameter below 2.5 µm for SIA was taken from the study of Stedman et al. (2007) and the following values were used: 0.45, 0.94, 0.97, respectively for NO$_3^-$, SO$_4^{2-}$ and NH$_4^+$. To show the contribution of anthropogenic sources, additional simulations were run including: only anthropogenic emission, anthropogenic and land natural emission and final - including sea aerosol. A concentrations map of secondary organic aerosols (SOA) for Europe was derived from the EMEP-Unified model

(Simpson et al., 2012). The chemical scheme employed for SOA EMEP-Unified model runs, as well as evaluation of modelled organic aerosol concentrations is described in Bergstrom et al (2012).

Input data
Emissions data
To get the total $PM_{2.5}$ concentrations the emissions data are required for primary particulate matter, together with emissions of gaseous compounds which are precursors of secondary aerosols. Anthropogenic emissions of SO_2, NO_x, $PPM_{2.5}$ for the UK were taken from the National Atmospheric Emissions Inventory (NAEI, www.naei.org.uk). Ammonia emissions were estimated using the AENEID (Atmospheric Emissions for National Environmental Impacts Determination) model that combines data on farm animal numbes with land cover information, fertiliser application, crops and non-agricultural emissions (Dragosits et al., 1998). Emissions of SO_2, NO_x and PPM from shipping were provided by AMEC (Whall 2010). For Poland point source emissions were provided by the National Administration of the Emission Trading Scheme (KASHUE). For the remaining emission sources, the national emissions inventory for the year 2007, organized by the SNAP sectors, including area, line and point sources, was taken from Dębski et al. (2009) and, in a spatial form suitable for modelling from Kryza et al. (2010). Land anthropogenic emission of PPM2.5 amounted to 80 Gg in the UK and 134 Gg in Poland in 2007. The main emission sources of PM2.5 in Poland are Non-Industrial Combustion Plants SNAP sector (about 40%). More than 60% mass is produced from the first three SNAP sectors (Combustion in Energy and Transformation Industries, Non-Industrial Combustion Plants and Combustion in Manufacturing Industry). In the UK the largest contribution is Road Transport sector (about 30%). Details on SSA emission calculations are described in Werner et al. (2011). Natural land emission of PPM was taken from the NatAir project (Korcz et al., 2009). Emission data for remaining areas of the model domain (FRAME-Europe) was taken from the EMEP inventory.

Meteorological data
Wind speed and direction data are required for FRAME with 24 sectors (each with a 15 degree resolution). The information was calculated using radiosonde data for the altitude 500-1000 m above sea level, according to the methodology proposed by Dore et al. (2006). For the UK, data was taken from seven different geographical locations and the station criteria were data completeness and geographical representation of northern, southern, western and eastern extend of the British Isles. For Poland in addition to three national stations there were also included two stations from Germany, and one station from the Czech Republic, Slovakia and Ukraine. Precipitation data for the UK was generated by the interpolation of measurements from the tipping bucket rain gauges gathered at the Meteorological Office national network at approximately 5000 stations. Precipitation data for Poland were developed using measurements from about 200 weather stations and spatially interpolated with the residual kriging procedure supported by a high resolution map of the long-term precipitation (Kryza 2008, Wałaszek et al., 2013).

Model evaluation
FRAME model results for SIA (SO_4^{2-}, NO_3^-, NH_4^+) and sea salt aerosol concentrations were earlier evaluated for both countries and were found to be in good agreement with measurement. The details are provided by Dore et al. (2007), Kryza et al. (2010, 2012), Werner et al. (2011, 2012). There are only four stations of $PM_{2.5}$ concentrations available for the year 2007 in the UK. The modelled and measured values are shown in Table 1.

Table 1. Measured and modelled concentrations of $PM_{2.5}$ in the UK in 2007. Unit: [$\mu g\ m^{-3}$].

Nr	Type of station	Measured	Modelled
1	rural background	4.0	4.1
2	rural background	12.0	8.8
3	urban background	14.0	16.2
4	urban traffic	22.0	15.6
5	rural background	11.0	11.3

RESULTS
Concentration of $PPM_{2.5}$
On average concentration of anthropogenic $PPM_{2.5}$ is about 2.5 times higher in Poland than in the UK (Table 2). When we include natural land emissions the increase of $PPM_{2.5}$ concentration for both countries is very small – about 1%. After including SSA the average concentration of $PPM_{2.5}$ in the UK increases significantly giving the similar values to those in Poland.

Table 2. Average concentration of PPM$_{2.5}$ for the UK and Poland in 2007 depending on the emission type included (anthr = anthropogenic, land nat = land natural, SSA = sea salt aerosol), Unit: [μg m^{-3}].

Emission included	UK PPM$_{2.5}$	Poland PPM$_{2.5}$
anthr	0.98	2.61
anthr + land nat	0.99	2.65
anthr + land nat + SSA	2.72	2.72

Concentration of PM$_{2.5}$

The average, for the total country area, concentration of PM$_{2.5}$ is higher in Poland than in the UK – 9.15 and 5.60 μg m^{-3}, respectively. The highest values concern London and coastline in the case of the UK and agglomerations in Poland (Figure 1). Local maxima in the UK can exceed 18 μg m^{-3} - for the areas, where a grid is divided between the sea and land. Around the London maxima concentrations come to 11 μg m^{-3}, whereas in Polish urban agglomerations values reach 16-18 μg m^{-3}.
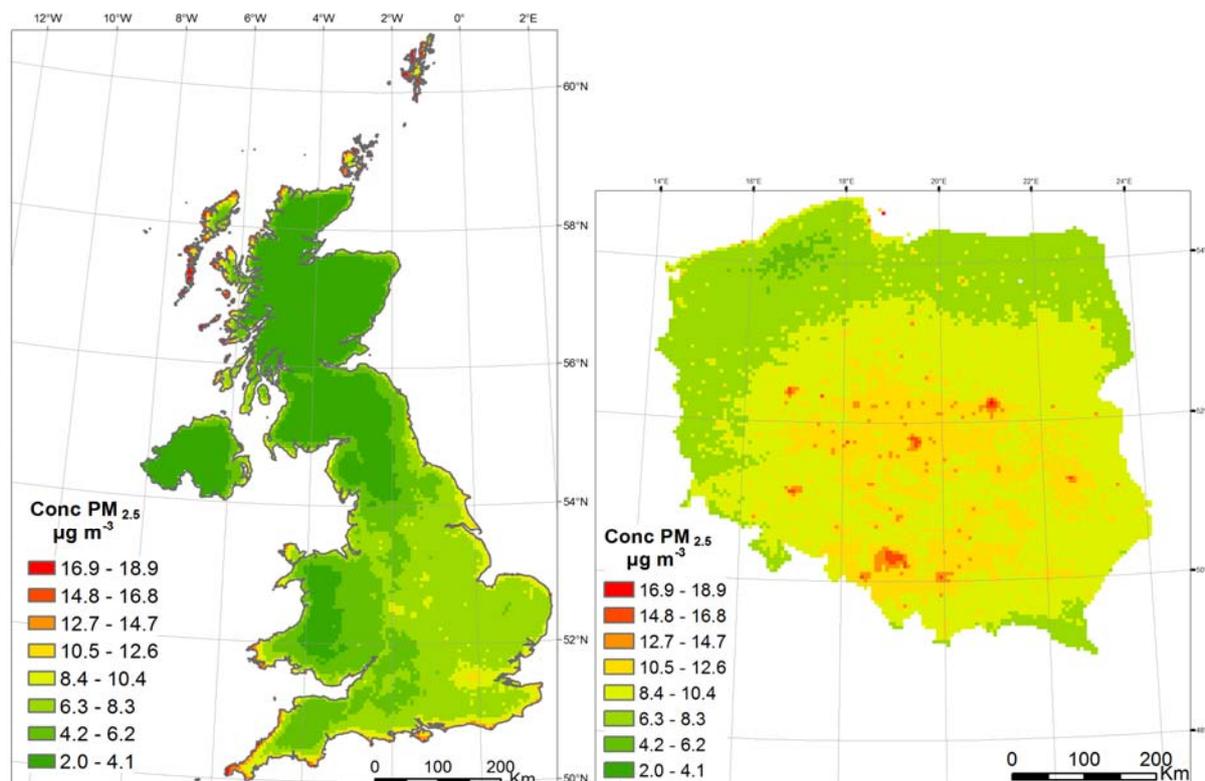
Figure 1. Total concentration of PM$_{2.5}$ in the UK and Poland in 2007 .

Natural particles contribute 34% of average PM$_{2.5}$ concentration in the UK and 20% in Poland. Among anthropogenic particles the highest contribution is from secondary inorganic aerosols – 71% and 58%, respectively for the UK and Poland. The second is primary particulate matter, and the lowest contribution is for secondary organic aerosols (Figure 2). In the case of natural particles there is a higher variability in the contribution of different compounds between the UK and Poland than for anthropogenic – PPM dominate in the UK (63%) and SOA in Poland. In total PM$_{2.5}$ concentration (anthropogenic + natural particles) dominate SIA (about 50%) in both countries, whereas the lowest contribution is for SOA.

Spatially, the highest contribution of PPM (including SSA) in total PM$_{2.5}$ concentration is along the coast (60-70% in the UK, 45-50% in Poland) and in agglomerations (40-50% in the UK, 55% in Poland). The highest SIA contribution is in the central part of both countries and reaches 70%.

The role of import and export of PPM$_{2.5}$ was calculated for two cases – including and excluding SSA. When we exclude SSA, the absolute mass of import is higher for Poland than for the UK. In this case import is higher than national emission in Poland and lower than the national emission in the UK (by about 30%). Spatially, in the UK, the highest contribution of imported particles is in the north of Scotland (>60%) and the lowest is in agglomerations. For Poland, the highest contribution is on the west and east parts of the country, where the fraction can exceed 70%. When we include SSA - import is higher than national emission in both countries and

the relation between export and import (export divided by import) amounts to 0.2 in the UK and 0.3 in Poland. Spatially, for both countries, the highest contribution of import is close to the coastline.
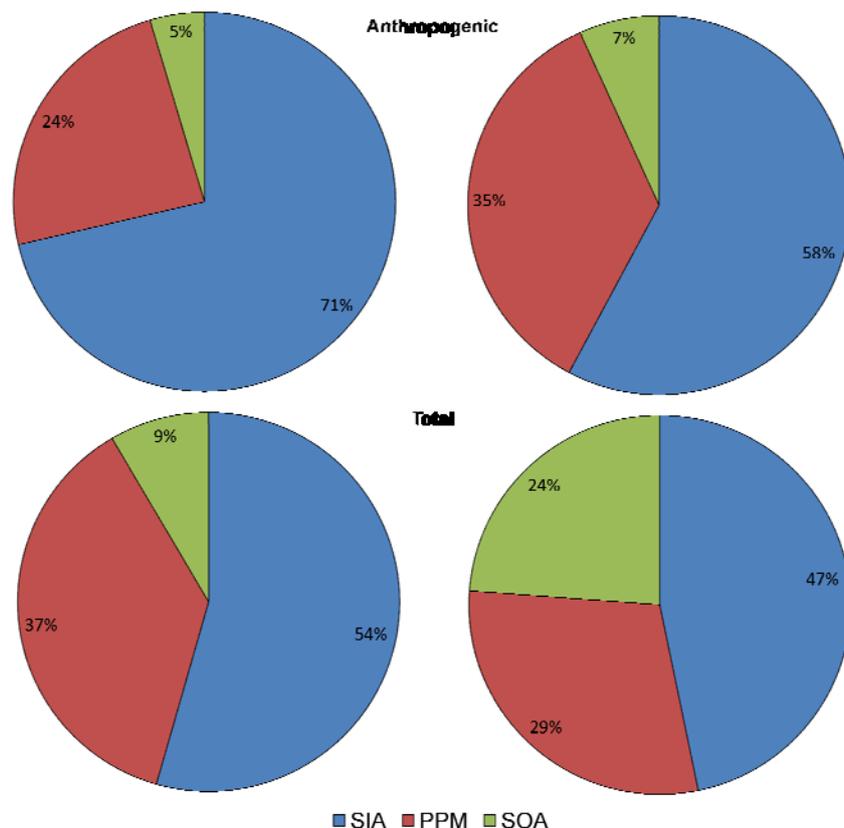
Figure 2. Contribution of $PM_{2.5}$ components for the UK (left) and Poland (right). PPM concentrations contain SSA.

SUMMARY AND CONCLUSIONS

The main aim of this study was to provide information about differences in spatial distribution and chemical composition of $PM_{2.5}$ between two countries which are diverse in terms of geographical location (inland, island), emission structure and climate conditions. The study can be summed up and concluded in the following statements:

- For both countries, high concentrations of $PM_{2.5}$ are related with anthropogenic emissions in agglomerations (e.g. road transport, combustion processes). Additionally, for the UK, high values appear along the coast, which are generated by the influence of sea salt aerosol.
- Particles of natural emission origin dominate in average $PM_{2.5}$ concentrations in the UK whereas the anthropogenic particles dominate in Poland. The highest contribution of natural particle in the UK comes from SSA.
- $PM_{2.5}$ concentrations reach higher values in Polish agglomerations than in the UK, thus the negative impacts of high particulate matter concentrations could influence more people in Poland than in the UK
- When we exclude the influence of sea salt aerosol, import is lower in the UK than in Poland. This is the result of the larger distance between the country and potential emission sources (e.g. from the European continent).

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