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## SPATIAL AND CHEMICAL PATTERNS OF PM<sub>2.5</sub> - DIFFERENCES BETWEEN A MARITIME AND AN INLAND COUNTRY

### PRZESTRZENNA I CHEMICZNA KOMPOZYCJA PM<sub>2.5</sub> - RÓŻNICE POMIĘDZY OBSZAREM Z DOMINUJĄCYM WPLYWEM MORZA ORAZ ŁĄDU

**Abstract:** The Fine Resolution Atmospheric Multi-pollutant Exchange model was used to calculate the mean annual concentration of PM<sub>2.5</sub> at a resolution of 5 km x 5 km for the United Kingdom (UK) and Poland for the year 2007. The modelled average PM<sub>2.5</sub> concentration is higher for Poland than the UK and amounts to 9.2  $\mu\text{g} \cdot \text{m}^{-3}$  and 5.6  $\mu\text{g} \cdot \text{m}^{-3}$ , respectively. The highest concentrations concern London and coastal areas (due to the sea salt contribution) for the UK and urban agglomerations in the case of Poland. Maximum values occurring close to the UK coastline can reach 18  $\mu\text{g} \cdot \text{m}^{-3}$ . The average contribution of natural particles amounts to 34 and 20% of total PM<sub>2.5</sub> concentration, respectively for the UK and Poland. Among anthropogenic particles for both countries the highest contribution falls on secondary inorganic aerosols and the lowest contribution is for secondary organic aerosols.

**Keywords:** PM<sub>2.5</sub>, concentrations, FRAME, United Kingdom, Poland

## Introduction

Particulate matter (PM) consists of complex and varying mixtures of particles suspended in the air, which vary in size and composition and are produced by a variety of natural and anthropogenic activities [1, 2]. The size of the particles determines where in the respiratory track they will deposit. Most of the PM<sub>10</sub> particles with a diameter above 5.0  $\mu\text{m}$  are deposited mainly in the upper respiratory track, while fine (PM<sub>2.5</sub>) and ultrafine particles are able to reach lungs [3]. The current interest in atmospheric PM is mainly due to its effect on human health [4-6] and on climate [7, 8]. A constant finding is that air pollutants contribute to increased mortality and hospital admissions [9].

Both anthropogenic (*eg* transport sector, industrial processes, power generation) and natural emissions (*eg* sea spray, wind blow dust) as well as meteorological or climatic factors contribute to the occurrence of high values of particulate matter (PM) concentrations [10]. Globally, high contribution of coarse particles comes from natural processes, whereas fine particles originate mainly from combustion processes and chemical

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condensation in air [11, 12]. Airborne particulate matter has both a primary component, which is emitted directly from pollution sources, and a secondary component, which is formed in the atmosphere by chemical reactions of gases, most notably sulphur dioxide, oxides of nitrogen and volatile organic compounds [13]. Turnbull and Harrison [14] studied the physical and chemical characteristics of PM at four sites across the UK. They found out that the secondary particles contributed 28-35% of sit-mean, sodium chloride 11-34%, and other, mainly crustal particles, accounted of 3-21%.

Mass contribution of  $PM_{2.5}$  in  $PM_{10}$  in the particulate matter emitted from different SNAP sectors is diverse and depends on *eg* the technology and characteristics of the source and air protection devices used [15, 16].

Detailed investigation of the chemical characteristics of atmospheric PM are important for both, elucidating the particles toxicity and its role in climate change [2, 15]. Moreover, The contribution of different emission sources is important 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 [17] Application of atmospheric chemical transport models allows for better understanding of the spatial and temporal distribution of particles.

In this study, the Fine Resolution Atmospheric Multi-pollutant Exchange model (FRAME) was used to provide spatial distribution and chemical characteristic 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. The differences in chemical composition of particles, distribution of emission sources and spatial distribution of concentrations were presented and discussed in this paper.

## 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 [18] and, for the FRAME model for Poland, by Kryza et al [19]. FRAME is used here with a grid resolution of 5 km x 5 km for both domains 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 the 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 50 km x 50 km 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 ( $PPM_{2.5}$  without sea salt aerosol) and sea salt aerosol (SSA, particles below 2.5  $\mu m$ ). The contribution of particles at a diameter below 2.5  $\mu m$  for SIA was taken from the study of Stedman et al [20] and the following factors 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, following simulations were run: 1) including only anthropogenic emission, 2) including anthropogenic and land natural emission and final 3) including anthropogenic, land natural and sea aerosol emission. To calculate the import of particles from other countries, additional simulations, which excluded national emissions were run. Import was understood as the influx of pollutants into the administrative boundaries of the UK or Poland (no within the model domain). Due to the highly complex chemical reactions

associated with secondary organic aerosols (SOA) formation, FRAME is not able to calculate SOA concentrations. SOA concentration map for Europe was derived from the EMEP-Unified model [21]. 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 [22].

### Input data

To get the total PM<sub>2.5</sub> concentrations the emissions data are required for primary particulate matter (PPM<sub>2.5</sub>), together with emissions of gaseous compounds which are precursors of secondary aerosols. Anthropogenic emissions of SO<sub>2</sub>, NO<sub>x</sub>, PPM<sub>2.5</sub> for the UK were taken from the National Atmospheric Emissions Inventory (NAEI, [www.naei.org.uk](http://www.naei.org.uk)). Ammonia emissions were estimated using the AENEID model (Atmospheric Emissions for National Environmental Impacts Determination [23]). For Poland point source emissions were provided by the National Administration of the Emission Trading Scheme. 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 Debski et al [24] and, in a spatial form suitable for modelling from Kryza et al [19]. Land anthropogenic emission of PPM<sub>2.5</sub> amounted to 80 Gg in the UK and 134 Gg in Poland in 2007. The main emission sources of PM<sub>2.5</sub> in Poland are Non-Industrial Combustion Plants (SNAP sector 02, about 40%). More than 60% mass is produced from the 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 from Road Transport sector (about 30%). The contribution of point sources in total land emission of PPM<sub>2.5</sub> is slightly higher for Poland than for the UK (24 and 18% respectively). The natural emissions included sea salt aerosols (SSA) and wind blown dust particles [25]. Emission data for remaining areas of the model domain (FRAME-Europe) was taken from the EMEP inventory.

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. 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. The selected stations included: Aberporth, Camborne, Herstmonceaux West End, Larkhill, Lerwick, Nottingham Watnall and Shoeburyness Landwick. For the FRAME runs for Poland, radiosonde data from stations Wroclaw, Leba, Warszawa (all located in Poland), Greifswald, Lindenberg (Germany), Prague (Czech Republic), Poprad (Slovakia), and Kiev (Ukraine) were used to calculate the wind roses. 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.

### Model evaluation

FRAME model results for SIA (SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, NH<sub>4</sub><sup>+</sup>) and sea salt aerosol concentrations were earlier evaluated for both countries and were found to be in good agreement with measurement, *eg* [19]. There are only five stations of PM<sub>2.5</sub> concentrations available for the

year 2007 in the UK and no stations available for this year for Poland. The reference measurement method of the PM concentrations is defined in the CAFE directive and is based on gravimetric measurements defined as the EN 1234:1999 standard. CAFE allows using of other methods but underlines that it has to be indicated that they are equivalent with the reference one. It is estimated that using the gravimetric method, an average water content contributes to 20-35% of particle mass [26]. In this case the FRAME model concentrations (dry particles) were recalculated by applying the scaling factor of 1.3 to include water content before model-measurement comparison. The modelled and measured values are shown in Table 1. For available stations there is a good accordance between modelled and measured concentrations. The highest underestimation of measured value is for urban traffic station.

Table 1

Measured and modelled concentrations of PM<sub>2.5</sub> in the UK in 2007

No.	Type of station	Measured [ $\mu\text{g} \cdot \text{m}^{-3}$ ]	Modelled [ $\mu\text{g} \cdot \text{m}^{-3}$ ]
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<sub>2.5</sub>

On average, for the total country area, concentration of anthropogenic primary particulate matter (PPM<sub>2.5</sub>) is about 2.5 times higher in Poland than in the UK (Table 2). When we include natural land emissions the increase of PPM<sub>2.5</sub> concentration for both countries is very small - about 1%. After including SSA the average concentration of PPM<sub>2.5</sub> in the UK increases significantly (2.7 times) giving the same value as in Poland. Increased PPM<sub>2.5</sub> concentrations (anthropogenic and land natural emission included) in the UK and Poland concern especially agglomerations. Clear modification in spatial distribution of PPM<sub>2.5</sub> is observed after including of sea salt aerosol into the simulation - then concentrations of PPM<sub>2.5</sub> close to the coastline reach 6.0-8.0  $\mu\text{g} \cdot \text{m}^{-3}$  in the UK and 3.0-4.0  $\mu\text{g} \cdot \text{m}^{-3}$  in Poland.

Table 2

Average concentration of PPM<sub>2.5</sub> for the UK and Poland in 2007 depending on the emission type

Emission included	UK	Poland
	PPM <sub>2.5</sub> [ $\mu\text{g} \cdot \text{m}^{-3}$ ]	
anthropogenic	0.98	2.61
anthropogenic + land natural	0.99	2.65
anthropogenic + land natural + SSA	2.72	2.72

### Concentration of PM<sub>2.5</sub>

Average, for the total country area, concentration of PM<sub>2.5</sub> is higher in Poland than in the UK - 9.2 and 5.6  $\mu\text{g} \cdot \text{m}^{-3}$ , respectively. The highest values concern London and coastline in the case of the UK and urban agglomerations in Poland (Fig. 1).

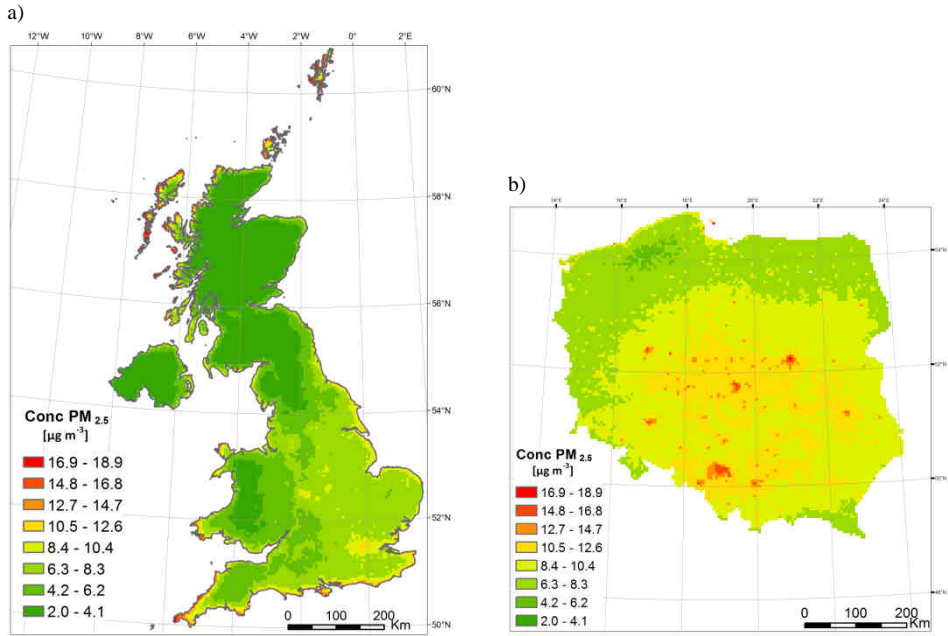


Fig. 1. Total concentration of PM<sub>2.5</sub> in: a) the UK and b) Poland in 2007

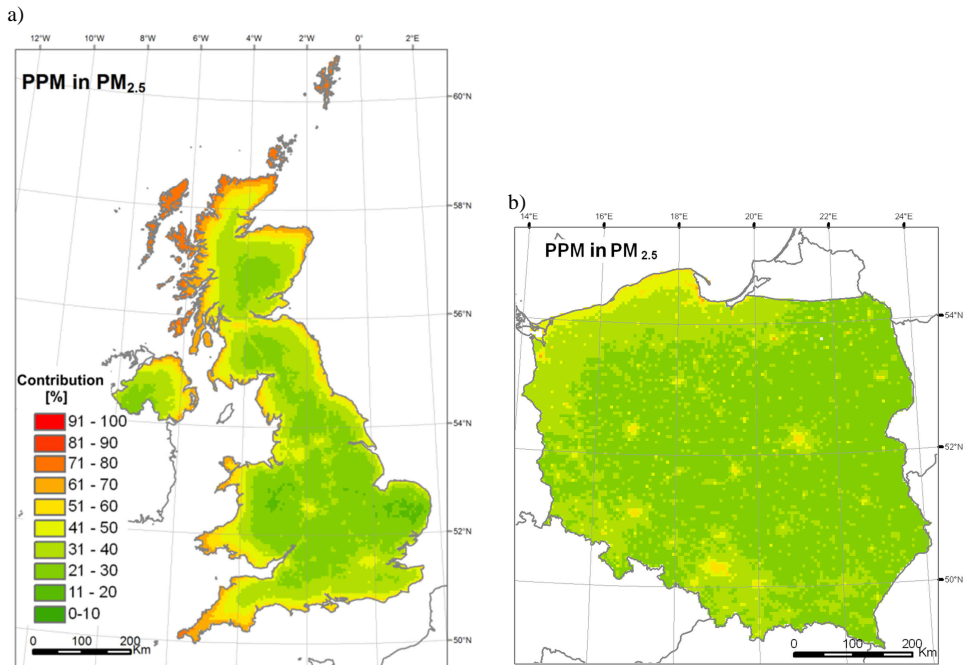


Fig. 2. Percentage contribution of primary particulate matter in PM<sub>2.5</sub> in: a) the UK and b) Poland in 2007 (the same legend for both figures)

For London maximum concentrations are equal  $11.0 \mu\text{g} \cdot \text{m}^{-3}$ , whereas in Polish urban agglomerations reach  $16.0\text{-}18.0 \mu\text{g} \cdot \text{m}^{-3}$ . Local maximum in the UK, where grids are divided between the sea and land, can exceed  $18.0 \mu\text{g} \cdot \text{m}^{-3}$ . High values are related there with high contribution of SSA. Spatially, the highest contribution of PPM (including SSA) in total  $\text{PM}_{2.5}$  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, Fig. 2). The highest SIA contribution is in the central part of both countries and reaches 70%.

Natural particles contribute 34% of average  $\text{PM}_{2.5}$  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 (Fig. 3).

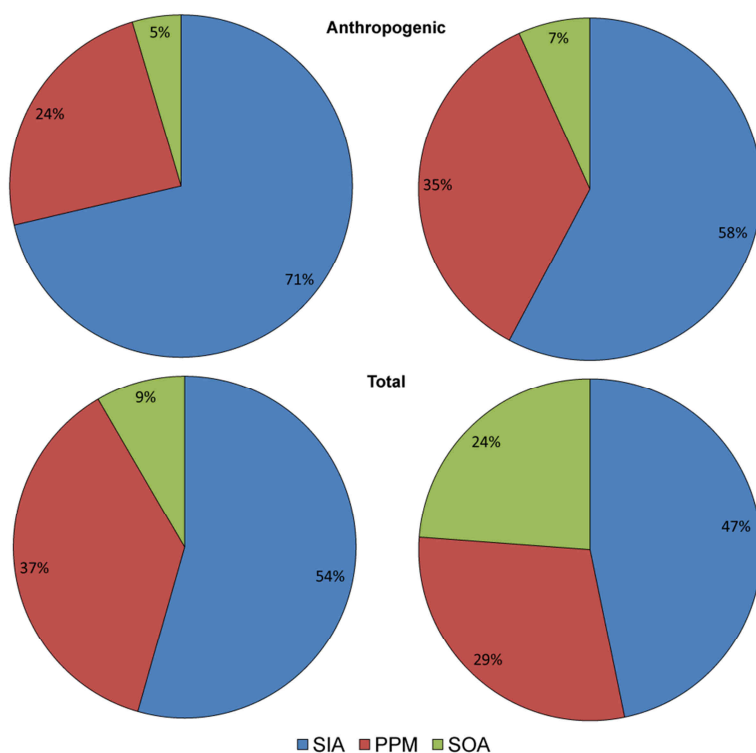


Fig. 3. Contribution of  $\text{PM}_{2.5}$  components for the UK (left) and Poland (right). PPM concentrations contain SSA

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 -  $\text{PPM}_{2.5}$  dominate in the UK (63%) and SOA in Poland. In total  $\text{PM}_{2.5}$  concentration (anthropogenic + natural particles) dominate SIA (about 50%) in both countries, whereas the lowest contribution is for SOA.

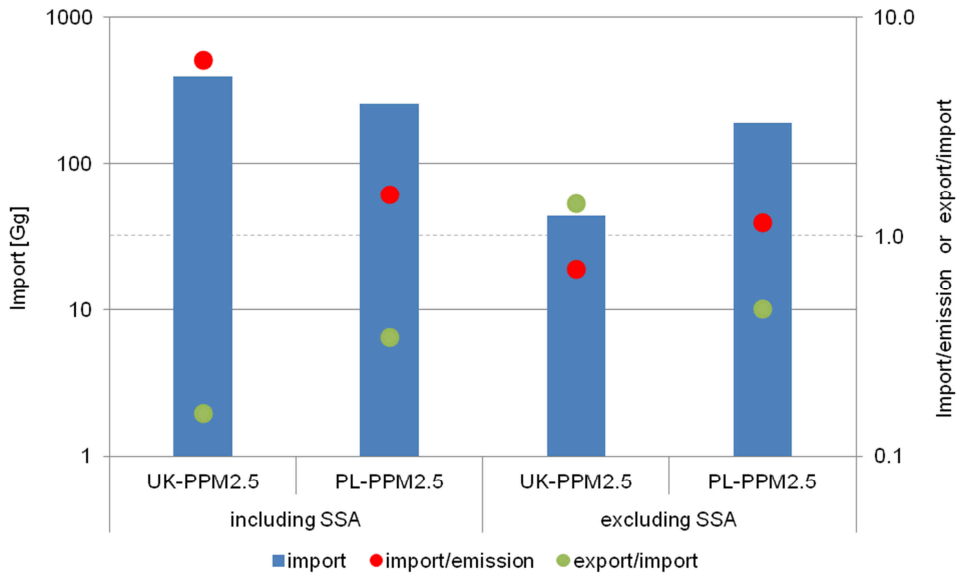


Fig. 4. The role of import and export of PPM<sub>2.5</sub> for two cases - including and excluding of SSA for the UK and Poland

The role of import and export of PPM<sub>2.5</sub> was calculated for two cases - including and excluding of SSA (Fig. 4). 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.

## Summary and conclusions

In this study, two countries which are diverse in terms of geographical location, emission structure and climate conditions, were compared in terms of spatial distribution and composition of PM<sub>2.5</sub>. The study can be summed up and concluded in the following statements:

- Average, for the total country area, concentration of PM<sub>2.5</sub> is higher in Poland than in the UK - 9.15 and 5.60  $\mu\text{g} \cdot \text{m}^{-3}$ , respectively.
- For both countries, high concentrations of PM<sub>2.5</sub> are related with anthropogenic emissions in urban agglomerations (*eg* road transport, combustion processes). It is more evident for Poland (cities in central and southern regions of the country) than for the UK (mainly London). Additionally, for the UK, high values appear along the coast, which are generated by the influence of sea salt aerosol. The contribution of SSA in total PM<sub>2.5</sub> concentration is not so high like calculated for PM<sub>10</sub> [27].

- The contribution of natural particles in total PM<sub>2.5</sub> (34% for the UK and 20% for Poland) is lower than calculated for PM<sub>10</sub> [27]. The highest contribution of natural particle in the UK is from SSA. Taking into account total concentration of PM<sub>2.5</sub> (natural and anthropogenic particles) secondary inorganic aerosols dominate for both countries.
- PM<sub>2.5</sub> concentrations reach higher values in Polish agglomerations in comparison to the UK, thus the negative impacts of high particulate matter concentrations could influence more people in Poland than in the UK.
- Absolute mass of import (excluding SSA) is higher for Poland than for the UK. This is the result of the larger distance between the UK and potential emission sources (*eg* from the European continent).

## Acknowledgements

This work was supported by the Polish National Science Centre grant number UMO-2012/05/B/ST10/00446. Preparation of the meteorological data for calculation of the SSA emissions for Poland has been carried out in the Wrocław Centre for Networking and Supercomputing (<http://www.wcss.wroc.pl>), Grant No. 170.

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