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Title: Differences in the spatial distribution and chemical composition of PM₁₀ between the **United Kingdom and Poland**

Authors: Małgorzata Werner^{a*}, Maciej Kryza^a, Anthony J. Dore^b

^a Department of Climatology and Atmosphere Protection, University of Wrocław,

Kosiby 8, Wrocław 51-670, Poland

^bCentre for Ecology and Hydrology,

Penicuik, Midlothian EH26 9HF, UK

*) corresponding author:

Małgorzata Werner, email: malgorzata.werner@uni.wroc.pl

Tel. +48 71-348-54-41

Fax. +48 71-372-94-98

Abstract

The Fine Resolution Atmospheric Multi-pollutant Exchange Model was used to calculate the

spatial distribution and chemical composition of PM₁₀ concentrations for two geographically

remote countries in Europe – the United Kingdom (UK) and Poland – for the year 2007.

These countries are diverse in terms of pollutant emissions as well as climate conditions.

Information on the contribution of natural and anthropogenic as well as national and

imported particles in total PM₁₀ concentrations in both countries is presented. The paper

shows that the modelled national annual average PM₁₀ concentrations, calculated for the

entire country area, is similar for the UK and Poland and close to 12 μg m⁻³. Secondary

inorganic aerosols dominate the total PM₁₀ concentrations in Poland. Primary particulate

matter has the greatest contribution to total PM₁₀ in the UK, with large contribution of base

cations. Anthropogenic sources predominate (81%) in total PM₁₀ concentrations in Poland,

whereas natural prevail in the UK – hence, the future reduction of PM_{10} air concentrations

by emissions reduction could be more difficult in the UK than in Poland.

Keywords: particulate matter, air concentration, chemical composition, United Kingdom, Poland

1. Introduction

Studies of human health have indicated that there are significant correlations between particulate matter levels (PM) and increased respiratory and cardiovascular diseases, and mortality (Pope et al. 2002, Pérez et al. 2008). An in depth understanding of the nature and origin of atmospheric particles is important in order to identify the relative contribution of emission sources, to investigate the associations between specific particle components and health, and also for policy makers to introduce suitable legislation for its control (Yin et al. 2010).

The PM composition and toxicity depend especially on a local characteristic of emission and prevailing wind directions. PM and PM precursors may be transported in the upper layer of the atmosphere over long distances, so the problem of this pollution must be considered transboundary. Atmospheric PM is a complex mixture of elemental and organic carbon, ammonium, nitrates, sulphates, mineral dust, heavy metals, PAHs, base cations and other trace elements, as well as water (Hueglin et al. 2005). It may be of natural (i.e. sea salt, mineral dust, biogenic organic) or anthropogenic origin (i.e. nitrates and sulphates, anthropogenic organic) and can be emitted directly into the atmosphere (primary particles), or formed in the atmosphere from gaseous precursors (secondary particles; Aldabe et al. 2011). So far there is no conclusive evidence which component is the most adverse to human health (Hoek et al. 2002). Some researchers suggest it could be soot but heavy metals (HM) are also considered to be very dangerous for live forms. An increase of HM concentration in the atmosphere is given as one of the reasons for the increased incidences

of cancer, cardiovascular or respiratory diseases (Raghunath et al. 1999, Nel 2005, Feng et al. 2009, Hernández-Soriano et al. 2011, Ru-Zhong et al. 2011).

The concentrations and chemical composition of PM can be influenced by processes on a regional scale as well as by long-range pollution transport. In some European regions in addition to anthropogenic sources, the ambient aerosol has an important contribution from natural sources (Korcz et al. 2009). Natural dust, due to local emissions from bare soil, and an influence of episodic African dust transport outbreaks is important for southern Europe. For the coastal areas, sea salt aerosol (SSA) plays a large part in total PM, especially in situations when the prevailing wind directions are from the sea region (Artíñano et al. 2001). It is estimated that natural particles globally contribute 80% of total PM emission(Korcz et al. 2009). According to Pueschel et al. (1995), 90% of a natural PM comes from the seas, oceans and deserts. Globally, most of the coarse PM fraction (PM_{coarse}, diameter in range from 2.5µm to 10µm) in the atmosphere origins from natural processes, whereas fine fraction (PM_{2.5}) origins from combustion processes and chemical condensation in the atmosphere (van Loon and Duffy 2008). The highest PM concentrations concern coastal and arid areas, where annual average values can reach 500 μg m⁻³, and also in urban areas where concentrations can exceed 200 µg m⁻³. The highest PM₁₀ concentrations in Europe are measured in the Benelux countries, the Po Valley, Slovakia and in large urban agglomerations (Yttri et al. 2009). The lowest PM₁₀ and PM_{2.5} concentrations in Europe concern the north and north-west region of the continent.

Together with particulate matter sampling and chemical characterisation, mathematical models have been shown to be extremely useful to interpret and understand the geographical distribution, temporal evolution and origin of the pollutants (Aldabe et al. 2011). Here, we use the Fine Resolution Atmospheric Multi-pollutant Exchange model

(FRAME) to calculate the spatial distribution and chemical composition of PM₁₀ concentrations for two geographically remote countries in Europe – the UK and Poland – for the common year 2007. These countries are diverse in terms of pollutant emissions as well as climate conditions. The UK is an island, exposed predominantly to maritime air masses from the Atlantic Ocean whereas Poland is a continental country in the centre of the European continent with a short coastline along the Baltic Sea but exposed to pollution transported from neighbouring European countries. The results will allow us to answer the following questions:

- What is the contribution of natural and anthropogenic particles to total PM₁₀
 concentrations in the UK and Poland?
- What is the contribution of transboundary transport and national emission in total pollutants concentration for the UK and Poland?
- What is the difference in chemical composition of PM₁₀ between the UK and Poland?
- What is the spatial distribution of PM₁₀ components' (secondary inorganic aerosols SIA, primary particulate matter PPM, secondary organic aerosols SOA) contribution for the UK and Poland?

Answers to these questions will also provide quantitative information to policymakers regarding to the possibility of PM_{10} concentrations reduction based on national emission abatements.

2. Data and methods

2.1. FRAME model description

The Fine Resolution Atmospheric Multi-pollutant Exchange (FRAME) model is used here to assess the annual mean concentrations and deposition of air pollutant. FRAME is a Lagrangian statistical trajectory model that is used for the United Kingdom and Poland to

support environmental management and protection. The fundamentals of the model are described by Singles et al. (1998), Fournier et al. (2004), Dore et al. (2007), Vieno et al. (2010) and, for FRAME model for Poland, by Kryza et al. 2012. FRAME is used here with a grid resolution of 5 km x 5 km and grid dimensions of 172 x 244 cells for the UK and 160 x 160 cells for Poland. The import of particles from outside of both domains is calculated with FRAME-Europe – a similar model to FRAME, which runs for the entire Europe on the EMEP grid at 50 km × 50 km resolution and grid dimension of 123 × 119 cells. Originally, FRAME was developed to represent the transport and deposition of nitrogen and sulphur compounds and assess the exceedance of critical loads for acid and nitrogen deposition (Matejko et al. 2009). An important step to achieve the total concentrations of PM₁₀ was the development of FRAME to include the emission and transport of sea salt aerosol, which is described in detail by Werner et al. (2011).

Dry deposition of sulphur and nitrogen compounds is calculated by determining vegetation dependent velocities (V_d) for each chemical species derived from a dry deposition model (Smith et al. 2000). The model derives maps of deposition velocity taking into account surface properties and geographical and altitudinal variation of wind speed. Dry removal rates of primary particulate matters are calculated separately for fine ($PM_{2.5}$) and coarse (PM_{co}) particles using the geometric mean size. For sea salt aerosol four groups of particle diameters were used, with the midpoints at 1.25 μ m, 3.75 μ m, 6.25 μ m and 8.75 μ m. A higher deposition velocity is applied to larger particles for which gravitational settling is more efficient. This is set according to the values proposed by Ruijgrok et al. (1995, 1997), Zhang et al. (2001) and Nho-Kim et al. (2004).

Wet deposition is calculated with a scavenging coefficient and constant drizzle approach, using precipitations rates calculated from a measurement-based map of average annual precipitation. The amount of material removed in a time period (Δt) is given by:

$$\Delta c_i = c_i (1 - e^{\lambda_i \Delta_t})$$

 Δc_i – decrease in concentration of species i due to removal by precipitation

 λ_i – scavenging coefficient

c_i – concentration of pollutants

The wet deposition flux to the surface is the sum of wet removal from all volume elements aloft, assuming that scavenged material comes down as precipitation. There is no separation between in-cloud and below-cloud processes and an averaged value of the scavenging ratio (Δ_i) is used in the model. To produce scavenging coefficient λ_i , Δ_i is combined with the precipitation rate and the depth of the mixing layer ΔH_{mix} :

$$\lambda_i = (\Delta_i P)/H_{mix}$$

An increased washout rate is assumed over hill areas due to the seeder-feeder effect. It is assumed that the washout rate for the orographic component of rainfall is twice that used for the non-orographic components (Dore et al. 1992).

To obtain the total concentrations of PM_{10} , the FRAME model was run for:

- Secondary inorganic aerosols (SIA): SO₄²⁻, NO₃-, NH₄+.
- Primary particulate matter (PPM): PPM_{2.5} and PPM_{co.} Additional simulations were done to show individual contribution of heavy metals (HM) and base cations (BC).
- Sea salt aerosol (SSA).

Due to the highly complex chemical reactions associated with SOA formation, FRAME is not able to calculate SOA concentrations. SOA concentration map for Europe was derived from the EMEP-Unified model (Simpson et al. 2012). The chemical scheme employed as well as validation of modelled organic aerosol concentrations with filter pack measurements and aerosol mass spectrometry is described in Bergström et al (2012).

To calculate the import of pollutants from other countries into the UK and Poland, additional simulations, which excluded national emissions were run. Import was understood as the influx of pollutants into the administrative boundaries of the UK and Poland (not within the domain). Similarly, export was calculated for pollutants emitted within the administrative borders of the analysed countries (the UK and Poland).

2.2. Emissions data

According to the EMEP estimates for the year 2007 Poland holds the second position in Europe for total national primary PM_{10} emission (after France) and is third in the case of $PM_{2.5}$ (after France and Italy). The UK is in the sixth and seventh place for PM_{10} and $PM_{2.5}$ emissions respectively.

To calculate the total PM_{10} concentrations the emissions data are required for primary particulate matter (PPM), together with emissions of gaseous compounds which are precursors of secondary aerosols. Input information of PPM is divided into two classes – $PM_{2.5}$ and PM_{co} (coarse particulates) in the case of anthropogenic emission and land natural emission, whereas for SSA, emission it is divided into four classes. The emission from anthropogenic point sources was inserted into the model according to the stack height using a plume rise model (Vieno et al. 2010). The SSA and wind blown dust emissions were injected in the surface layer (first layer of the FRAME model).

Area and point sources anthropogenic emissions of SO₂, NO_x, PPM_{2.5} and PPM_{co} for the UK were taken directly from the National Atmospheric Emissions Inventory (NAEI, www.naei.org.uk). Ammonia emissions were estimated for each grid square using the AENEID model (Atmospheric Emissions for National Environmental Impacts Determination) that combines data on farm animal numbers with land cover information, as well as fertiliser application, crops and non-agricultural emissions (Dragosits et al. 1998). Emissions of SO₂, NO_x, and PPM₁₀ from shipping were provided by AMEC and covered an area extending to 200 nautical miles from the coast of the UK (Whall et al. 2010).

For Poland, point source emissions were provided by the Institute of Environmental Protection. 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. (2008) and, in a spatial form suitable for modelling from Kryza et al. (2010, 2011). The summary of the anthropogenic input emission data is provided in Table 1. Emission data for remaining areas of the model domain (neighbouring countries) was taken from the EMEP inventory.

Table 1 Summary of anthropogenic land emission for the UK and Poland in year 2007 [Gg].

Country	SO ₂	NO _x	NH ₃	PPM _{2.5}	PPM _{co}
UK	585	1413	301	80	54
Poland	1131	885	291	134	136

Table 2 SNAP'97 sectors.

Short name	Full name
S01	Combustion In Energy And Transformation Industries
S02	Non-Industrial Combustion Plants
S03	Combustion In Manufacturing Industry

S04	Production Processes
S05	Extraction And Distribution Of Fossil Fuels And Geothermal Energy
S06	Solvent And Other Product Use
S07	Road Transport
S08	Other Mobile Sources And Machinery
S09	Waste Treatment And Disposal
S10	Agriculture
S11	Other Sources And Sinks

S01 (Table 2) emission sector contributes over 60% of total SO_2 emission in both the UK and Poland. Simultaneously, a significantly higher contribution of Non-Industrial Combustion Plants (residential combustion) appears in Poland (18%) than in the UK (7%). Moreover, Poland has one of the highest SO_2 emission per capita in Europe if S02 is considered. This stems from the widespread usage of coal in residential combustion due to economic reasons. In other European countries, including the UK, gas is the main fuel for residential heating, which has a large positive ecological impact. In the case of NO_{x_0} , for both countries more than 60% of emission is from two sectors - S07 and S01. The main source of ammonia emission is agriculture, respectively 89 and 98% of the total for the UK and Poland.

For Poland the main source of $PPM_{2.5}$ and PPM_{co} is S02, which contribute 50 and 41%, respectively. For both $PPM_{2.5}$ and PPM_{co} over 60% of mass is emitted from the first three SNAP sectors. In contrast, road transport and agriculture contribute the most of the UK emission of $PPM_{2.5}$ and PPM_{co} , respectively.

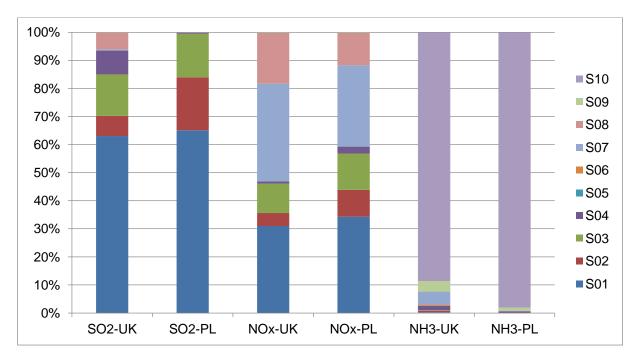


Figure 1 Contribution of SNAP'97 sectors to total SO_2 , NO_x and NH_3 emissions in the UK and Poland for the year 2007. List of SNAP sectors is in Table 1.

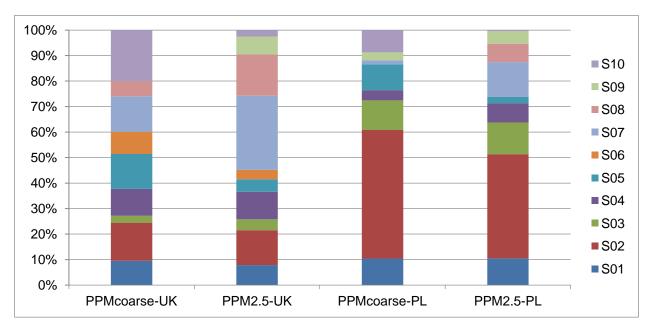


Figure 2 Contribution of SNAP'97 sectors to total PPM_{coarse} and $PPM_{2.5}$ emission in the UK and Poland for the year 2007. List of SNAP sectors is in Table 1.

Details on SSA emission calculations are described in Werner et al. (2011) for the UK and in Werner et al. (2012) for Poland. Here, only the main assumptions are described. Sea salt aerosol emission was calculated using three parameterisations dependent on particles size. For the particles radius at relative humidity 80% below 0.8 μ m (r_{80} <0.8) the parameterisation

proposed by Monahan et al. (1986) and modified by Gong (2003) was applied. For larger particles (0.8< r_{80} <4.0) the sea salt generation function is taken from Monahan et al. (1986). For particles larger than 4.0 μ m radius, the Smith and Harrison (1998) parameterization was used.

Natural land emission of PM₁₀ was taken from the NatAir project (Korcz et al. 2009; natair.ier.uni-stuttgart.de). NatAir provides data with spatial resolution of 10 km×10 km and temporal resolution of one hour. The data was prepared for four years: 1997, 2000, 2001 and 2003. The spatial pattern of the emission for different years is similar, and changes are mainly due to meteorological condition. Because there is no information about emission for year 2007 an average value from all available years were used. A scaling factor provided by Korcz et al. (2009) was used to calculate PPM_{2.5} and PPM_{co} emission from land natural sources:

PPM_{2.5}=PPM₁₀*0.11

Total emission of natural land PPM₁₀ emission amounts to 56.10 Gg for the UK and 35.10 Gg for Poland. Average emission is 2.25 kg ha⁻¹year⁻¹ for the UK and 0.70 kg ha⁻¹year⁻¹ for Poland. For the UK the highest values are along the east coastline, which exceed 30 kg ha⁻¹year⁻¹. In Poland the largest emission are located in Żułway Wiślane (the region around the delta of the Wistula River) and do not exceed 10 kg ha⁻¹year⁻¹

2.3. Meteorological data for the FRAME model

FRAME requires annual average meteorological information on wind and precipitation conditions. Wind speed and direction data is required in 24 sectors (each with a 15 degree resolution). The information was calculated for the selected year 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 selection criteria were data completeness and geographical representation of northern, southern, western and eastern extent of the British Isles. The selected stations were: Aberporth, Camborne, Herstmonceaux West End, Larkhill, Lerwick, Nottingham Watnall and Shoeburyness Landwick. For the FRAME runs for Poland, radiosonde data from stations Wrocław, Łeba, Warszawa (all located in Poland), Greifswald, Lindenberg (Germany), Prague (Czech Republic), Poprad (Slovakia), and Kiev (Ukraine) were used to calculate the wind roses. The wind roses for the UK and Poland for 2007 are shown in Figure 3.

In the UK, the circulation pattern for the year 2007 illustrates the predominant wind direction from the SW-W, and low frequency of the NE-SE sector. For Poland, the average westerly direction frequencies were approximately twice those of easterlies. The harmonic average wind speed for the year 2007 was 6.5 m s⁻¹ for the UK and 5.9 m s⁻¹ for Poland. The highest wind speeds coincided with the wind direction of the highest frequencies.

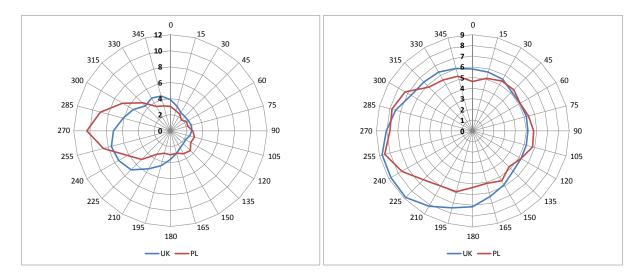


Figure 3 Wind direction frequency (left side, %) and wind speed (right side, m s⁻¹) rose for the UK and Poland in year 2007.

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. For Poland precipitation data was 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). The average annual rainfall was 1200mm for the UK and 710 mm for Poland. These values were 5% higher for the UK and 15% higher in Poland in comparison to the long term average precipitation from the period 1986-2007.

2.4. Model evaluation

Measured PM_{10} concentrations for the UK was derived from the Particulate Matter Network and available through the website: http://uk-air.defra.gov.uk. Measurements for Poland are conducted by the National Environmental Monitoring and the results are available through the AirBase database (www.eea.europa.eu). Comparison of the modelled versus measured values of secondary organic aerosols concentrations was presented in details in Simpson et al. (2007) and Bergström *et al.* (2012). The validation was made at selected CARBOSOL and EMEP sites in Europe.

According to the CAFE directive, for the evaluation of the regional model the rural stations are the most appropriate. Because most of the stations both in the UK and Poland are urban background, in model-measurement comparison both rural and urban background were used. Total number of 82 stations for the UK and 34 stations for Poland were used in this study. All the stations met the criterion of data completeness (data completeness > 90%). The reference measurement method of the PM_{10} 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 contribute to 20-35% of particle mass (Tsyro 2005, Stedman et al. 2007). If other than reference method is used, an appropriate scaling factor must be used to assure equivalence. This procedure is used in the UK before data are made available (AQEG 2005). In this case the FRAME model concentrations (dry particles) for the UK were recalculated by applying the scaling factor of 1.3 to include water content before modelmeasurement comparison. In Poland measurements are conducted both – using the reference gravimetric method as well as automatic methods. The studies concerning the relation of operational measurements to the reference method have not been done yet but are planned in the near future (Chief Inspectorate of Environmental Protection, oral communication). In such situation we decided to use the Mean Bias statistic for modelled values suggested by Łobocki (2003), Monteiro et al. (2011), Borrego et al. (2011). There are several techniques by which bias correction can be applied, such as mean subtraction, multiplicative ratio adjustment (Mc Keen at al. 2005), hybrid forecast and Kalman filter (Kang et al. 2008). The bias-correction does not try to gain additional insight into model deficiencies or performance or to correct them artificially, but aims to remove potential errors e.g. intrinsic to each model formulation or input data (Borrego et al. 2011). In this study a multiplicative ratio correction is used to correct the bias of the modelled data. The corrected concentrations are estimated based on the application of a correction factor to the raw modelled concentrations. The correction factor is calculated as the quotient between the additions of observed and modelled concentrations.

The scaling factor for the UK as well as bias-correction for Poland were only used in the evaluation process. Remaining presented results are shown without correction. It was important to keep data for compounds analysis without correction. Water contribution is

not the same for particular components, so including correction would give an unreliable picture of the components contribution.

The model-measurements comparison was summarized using five error statistics, including mean bias (MB), normalized mean bias (NMB) root mean squared error (RMSE), factor of two (FAC2) and correlation coefficient (R). All the statistics are summarized in Table 3 and 4. For Poland results are provided for two cases –before and after BIAS correction. In order to provide more complete picture of modelled results, additionally the statistics are provided for secondary inorganic aerosols (SIA, SO_4^{2-} , NO_3^- , NH_4^+) and base cations (BC), which are the components of PM_{10} . Measurements of inorganic aerosol components in the UK were taken from 30 sites in the Acid Gases and Aerosols monitoring network which is part of the UKEAP (UK Eutrophying and Acidifying Pollutants) national network (http://pollutantdeposition.defra.gov.uk/ukeap).

FRAME model evaluation indicates a tendency for underestimation of measured concentrations of PM₁₀. NMB indicates that, including bias correction for Poland, a larger underestimation appears for the UK than Poland. For Poland all stations are between reference lines 1:2 and 2:1, while for the UK one station is outside this range. The correlation coefficient is 0.56 for the UK and 0.43 for Poland.

Table 3 Error statistics for PM₁₀, SIA and BC for the UK

	PM ₁₀	SO ₄ ²⁻	NO ₃	NH_4^{+}	Na⁺	Mg ²⁺
МВ	-4.098	0.472	0.031	-0.187	-0.093	0.003
RMSE	5.793	0.717	0.546	0.250	0.511	0.043
NMB	-0.197	0.505	0.019	0.247	-0.116	0.052
FAC2	0.97	0.77	0.89	0.79	0.65	0.50
R	0.56	0.79	0.95	0.94	0.43	0.49

Table 4. Error statistics for PM₁₀, SIA and BC for Poland

	PM ₁₀	PM10 BIAS	SO ₄ ² -	NO ₃	NH ₄ ⁺	Na⁺
MB	-4.098	-1.262	-0.559	0.136	-0.337	-0.277
RMSE	5.793	7.771	0.961	0.584	0.362	5.106
NMB	-0.197	-0.044	-0.220	-0.085	0.300	-0.057
FAC2	0.97	1.00	0.90	0.72	0.83	0.64
R	0.43	0.43	0.76	0.64	0.80	0.70

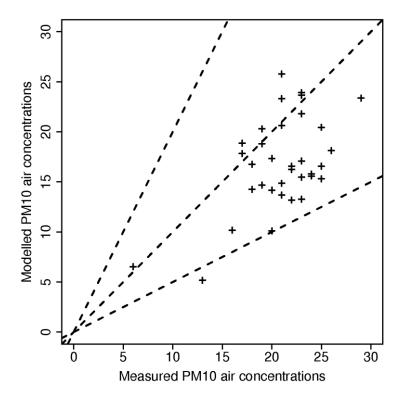


Figure 4 Scatter plot for PM_{10} concentration in the UK for year 2007. Unit: $\mu g \; m^{\text{-}3}$

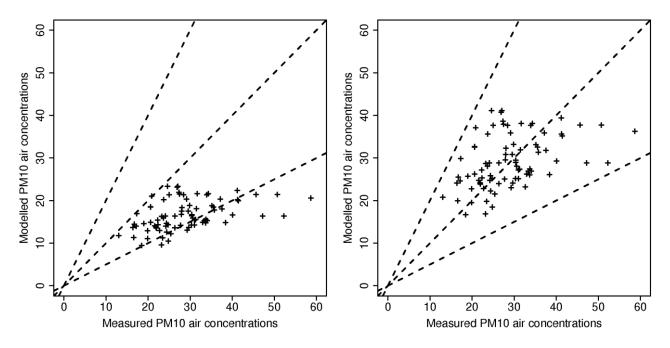


Figure 5 Scatter plot for PM $_{10}$ concentration in Poland: without BIAS correction (left), with BIAS correction (right) for year 2007. Unit: $\mu g m^{-3}$

3. Results

The results are organised as follows: first, the average concentrations of PM_{10} and contribution of their compounds for the UK and Poland are presented. Afterwards, the contribution of natural and anthropogenic particles for both countries is described. The spatial distribution of PM_{10} concentrations and spatial distribution of compounds of PM_{10} are then presented. Finally the role of transboundary transport in PM_{10} concentrations is shown for both countries.

The modelled national annual average PM_{10} concentration, calculated for the entire country area, is similar for the UK and Poland and close to 12 μg m⁻³. Secondary inorganic aerosols dominate the total PM_{10} concentration in Poland (Table 4). PPM (including SSA) has the greatest contribution to total PM_{10} in the UK, with a large contribution of base cations (28%). The contribution of heavy metals (As, Cd, Pb) is marginal both for the UK and Poland but environmentally important because of their adverse impact on an environment, including human health. The average concentration of SOA is about four times lower for the UK than

for Poland and respectively accounts for 4 and 18% of total PM_{10} concentration. The difference is especially related with concentrations of biogenic SOA. Average concentration (for country) of biogenic SOA is more than 5 times higher in Poland than in the UK and equal 0.84 and 0.16 μ g m⁻³, respectively. Average concentration of anthropogenic SOA is 0.25 μ g m⁻³ in Poland and 0.09 μ g m⁻³ in the UK.

Anthropogenic sources predominate (81%) in total PM_{10} concentration in Poland, whereas natural sources prevail in the UK. Among the anthropogenic particles, in both countries, the highest contribution is for SIA, respectively 70 and 57% for the UK and Poland. Among natural particles, SSA contributes 95% for the UK and 61% for Poland.

Table 4 The average (for all country grids) concentration of PM_{10} and their components for the UK and Poland in year 2007. Unit: $\mu g \ m^{-3}$

Component	Species	UK	Poland
	NO ₃	1.87	2.06
CIA	NH_4^+	0.65	1.10
SIA	SO ₄ ²⁻	1.53	2.41
	NO ₃ 1.87 NH ₄ ⁺ 0.65	5.57	
	Na⁺	1.81	0.12
		0.16	1.10
	Ca ²⁺	0.70	0.03
PPM+SSA	K ⁺	0.60	0.03
FFIVITSSA	As	0.00004	0.00032
	Cd	0.00003	0.00020
	Pb	0.00334	0.00300
	other	4.10	2.96
	total	7.37	4.24
SOA	total	0.52	2.18
PM ₁₀ (SIA+PPM+SSA+SOA)	total	11.94	11.99

The character of the spatial distribution is different for both countries. For the UK, increased values concern a few kilometres area around the coastline. PM_{10} concentrations amount to 18-20 $\mu g \ m^{-3}$ on the west coast and locally on the south-east can exceed 30 $\mu g \ m^{-3}$. In addition to these regions, increased values in comparison to the average concentration,

appear in the south-east with a local maximum close to London and equal 18 μ g m⁻³. In northern Scotland, except for the coastline areas, concentrations are significantly lower and in the range of 4.0-5.0 μ g m⁻³. For Poland, increased values of the PM₁₀ concentrations appear in central and southern part of the country and in the few kilometres-wide area along the coastline of the Baltic Sea. Increased values, in comparison to the country average, appear also in large urban areas (e.g. Warszawa, Łódź, Wrocław, Poznań, Kraków) and in the Upper Silesia region, where concentrations exceed 24 μ g m⁻³. Close to the Baltic Sea concentrations reaches 20 μ g m⁻³.

The spatial distribution of the primary particles, SIA and SOA contribution to the total PM_{10} concentrations is shown in Figs. 8 and 9. The PPM_{10} contribution in PM_{10} amounts to 30-40% in the centre of the UK and increases to 90% close to the coastline. In Poland the PPM_{10} contribution varies from 20 to 80% and the highest is related to urban agglomerations, but for the majority of Poland amounts to 20-30%. The SIA contributes to about 10-20% of the total PM_{10} concentrations close to the UK coastline and increases to 60-70% in central England. For Poland the lowest contribution of SIA concerns the coast of the Baltic Sea (< 20%) and also centres of urban agglomerations (emission source areas for gaseous precursors of particulates). The average contribution of SIA in central Poland amounts to 50-60%.

Similarly to SIA, the lowest contribution of SOA in total PM_{10} was calculated along the coast and in urban areas (< 5% for the UK and Poland), whereas maxima reaches 20% in the northern Scotland and 20-25% in north – and south – east Poland.

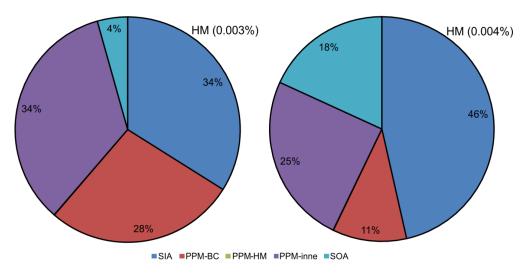


Figure 6 Contribution of PM_{10} components (SIA, PPM-BC, PPM-HM, PPM-other, SOA) for the UK (left) and Poland (right). PPM concentration contains SSA

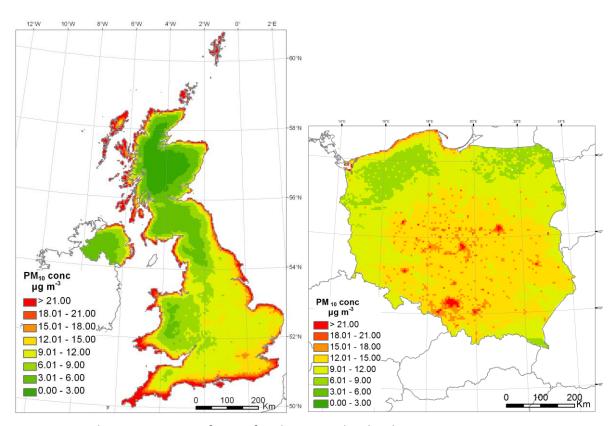


Figure 7 Total concentration of PM_{10} for the UK and Poland in year 2007

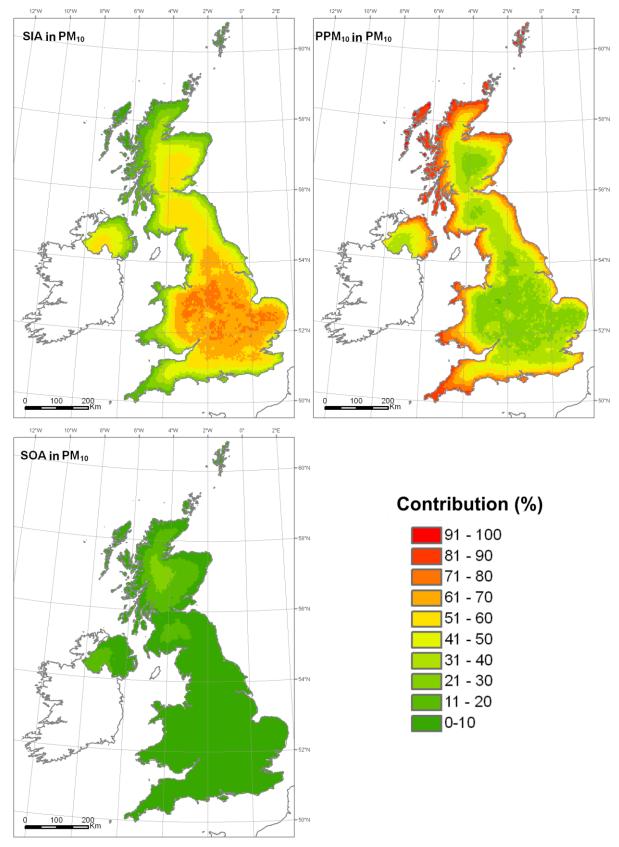


Figure 8 The contribution of SIA, PPM (including SSA) and SOA in total PM_{10} concentrations for the UK in year 2007

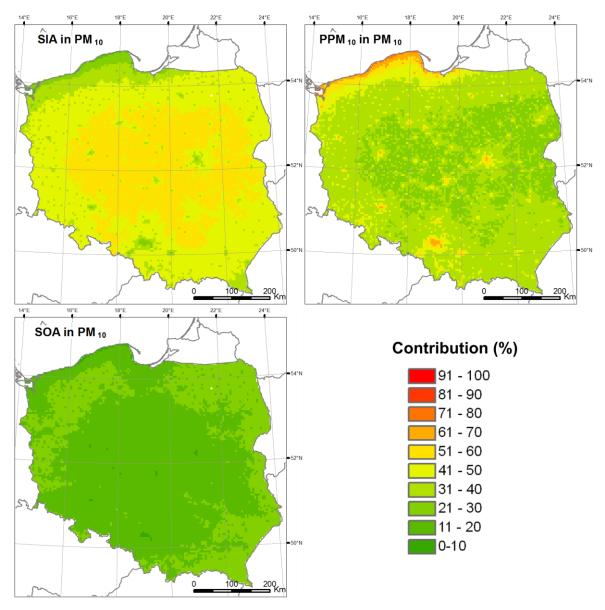


Figure 9 The contribution of SIA, PPM (including SSA) and SOA in total PM_{10} concentrations for Poland in year 2007

The role of trans-boundary transport is shown individually for SIA components and PPM $_{10}$. The absolute mass of import is higher for Poland than the UK in the case of pollutants for which the sea surface is not the main source of emission (e.g. N-NOx, N-NHx). For sulphur and nitrogen compounds the largest import is for N-NOx both for the UK and Poland. For PPM $_{10}$ the role of import was calculated for two cases – including and excluding SSA. When we exclude SSA, PPM $_{10}$ import is higher for Poland than for the UK. In this case import exceeds the country PPM $_{10}$ emission (1.1 times) in Poland but is below the country emission

in the UK (by about 30%). Export is higher than import by 13% in the UK. Including SSA, import is higher for the UK than for Poland and for both countries import is higher than country emission.

Lower absolute import to the UK than to Poland for nitrogen compounds or PPM (excluding the role of SSA) results from the larger distance of the UK from sources of emission (e.g. the continental Europe). During the transport, some pollutants are deposited and do not reach the UK. Simultaneously the predominant westerly circulation results in the more frequent import of clean air from oceanic regions than from continental areas. The same factors influence the domination of export over import in the UK. Poland, situated in the centre of Europe, is surrounded by a number of major emission sources and receives a significant input of pollutants emitted outside the country.

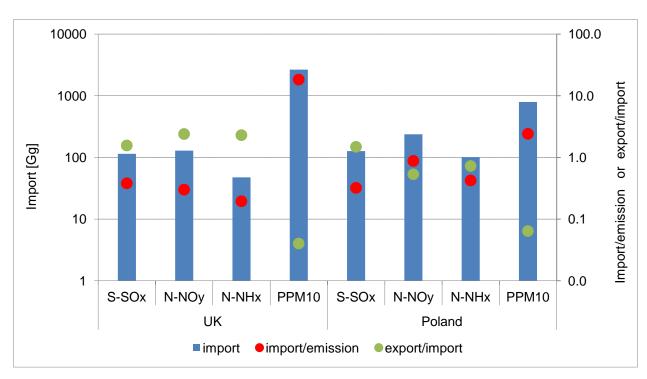


Figure 10 Absolute import of sulphur and nitrogen compounds and PPM $_{10}$ for the UK and Poland in year 2007 and ratios of import/emissions and export/import

4. Summary and conclusions

In this study, two geographically remote countries in Europe – the UK and Poland were compared in terms of spatial distribution and chemical composition of PM₁₀. The results address a number of important questions for both countries: what is the contribution of natural and anthropogenic particle in total PM₁₀ concentrations?; what is the contribution of transboundary transport? and what is the chemical composition of PM_{10} ? Similar average concentration of PM₁₀ for the UK and Poland, considering significant differences in national total emissions of anthropogenic primary particulates and gaseous precursors of atmospheric aerosols, results from high values of SSA along the coastline in the UK. Decrease of SSA concentrations by 90% in comparison to the values close to the coast is at a distance of 50-60 km from the coastline, both in the UK and Poland. Similar results (90% decrease after 58 km) were obtained by Delalieux (2006) for the area of Galicia in Spain. Somewhat further inland increased concentrations of SSA appear in Cornish and Pembroke Peninsula (West Wales), where the 90% decline is after 80 km. Increased concentrations of SSA as well as PM₁₀ on the west cost of the UK in comparison to the east are a result of more frequent westerly atmospheric circulation as well as increased emission of atmospheric aerosol related to the increased wind speed in the open ocean, compared with the North Sea.

The highest PM₁₀ concentrations in Poland concern the central and southern part of the country, especially Upper Silesia and urban agglomerations.

The highest contribution to total PM_{10} concentrations is SIA for Poland and PPM (with SSA) for the UK. The quantitative contribution of heavy metals is marginal both for the UK and Poland, however important with regard to impact on the environment. The results confirm studies conducted in continental European countries by Hueglin et al. (2005) and Putaud et

al. (2004), which indicated the dominance of sulphate and nitrate aerosol in total PM_{10} concentrations.

The lowest contribution of SIA in PM_{10} concentrations in north-west Poland results due to the absence of significant sources of emission in this area, as well as the inflow of relatively clean air from the NW sector, from the Atlantic Ocean. Similarly, the lowest contribution of SIA in the northern and western part of the UK results from a smaller number of emitters as well as the transport of pollutants to the east in prevailing westerly circulation.

For Poland, anthropogenic particles dominate the total PM_{10} concentrations (81%), with high contribution of secondary inorganic aerosols. For the UK, natural particles dominate (54%) PM_{10} , among which 90% is due to sea salt aerosol.

When SSA aerosol is not included, the role of import in total PM₁₀ concentrations is lower for the UK than Poland. This is related to the greater distance between the UK and continental emission sources, as well as the prevailing westerly circulation in which imports clean air from oceanic regions. During the transport, part of the aerosol mass is deposited and does not reach the UK. The same factors are responsible for the dominance of the export over import in the UK. Poland, located in the centre of Europe and surrounded by many emission sources in neighbouring countries, receives a major contribution from pollutants emitted outside the country.

After inclusion of SSA, import is higher in the UK than Poland and in both countries import is higher than national emission.

Based on the results obtained from this study, the following conclusions can be drawn:

Because natural particles dominate the average PM₁₀ concentrations in the UK,
 whereas anthropogenic particles dominate in Poland, the future reduction of PM₁₀ air

concentrations by emissions reductions policies could be more difficult in the UK than in Poland.

- High concentrations of PM₁₀ in the UK are related to a narrow area along the coast. From this, it follows that, although there is a similar average concentration of particulate matter in the UK and Poland, the amount of people exposed to high values of particulate matter concentrations could be lower in the UK, than in Poland where high concentrations are related to urban areas.
- The analysis of trans-boundary transport indicated that the import was higher for Poland in the case of particles for which sea salt aerosol was not the main compound. The air pollution predictions, especially in Poland, must take into consideration not only the emission trends in the country being studied, but also emission changes in neighbouring countries.

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