

Spatial analysis of air masses backward trajectories in order to identify distant sources of fine particulate matter emission

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Abstract: The paper presents a method of identifying distant emission sources of fine particulate matter $PM_{2.5}$ affecting significantly $PM_{2.5}$ concentrations at a given location. The method involves spatial analysis of aggregate information about $PM_{2.5}$ concentrations measured at the location and air masses backward trajectories calculated by HYSPLIT model. The method was examined for three locations of $PM_{2.5}$ measurement stations (Diabla Góra, Gdańsk, and Katowice) which represented different environmental conditions. The backward trajectories were calculated starting from different heights (30, 50, 100 and 150 m a. g. l.). All points of a single backward trajectory were assigned to the $PM_{2.5}$ concentration corresponding to the date and the site of the beginning of trajectory calculation. Daily average concentrations of $PM_{2.5}$ were used, and in the case of Gdańsk also hourly ones. It enabled to assess the effectiveness of the presented method using daily averages if hourly ones were not available. Locations of distant sources of fine particulate matter emission were determined by assigning to each grid node a mean value of $PM_{2.5}$ concentrations associated with the trajectories points located within the so-called search ellipse. Nearby sources of fine particulate matter emission were eliminated by filtering the trajectories points located close to each other (so-called duplicates). The analyses covered the period of January–March 2010. The results indicated the different origin of air masses in the northern and southern Poland. In Diabla Góra and Gdańsk the distant sources of fine particulate matter emission are identified in Belarus and Russia. In Katowice the impact of the Belarusian $PM_{2.5}$ emission sources was also noted but as the most important fine particulate matter emission sources were considered those located in the area of Romania, Hungary, Slovakia and Ukraine.

Introduction

Assessment of distant emission sources impact on air pollution in a certain area is one of main problems in air pollution monitoring and forecasting. It is a very complex issue because the influence of a distant emission source on air pollution measured at a given receptor depends on a location of the source as well as atmospheric circulation and wind conditions. The distant emission source is understood to be able to affect a given receptor located at a distance of several hundred kilometres from the source through cross-border transport of air pollutants. Air mass source regions may be recognised by backward trajectory analysis. Backward trajectory is obtained by solving a trajectory differential equation (Stohl 1996, Stohl 1998, Stohl et al. 1995) using wind data. In principle, backward trajectories can be calculated directly from surface and radiosonde wind observations by interpolating data from available measurement sites. It is a proper approach if a dense network of wind measurement exists but its applicability seems to be restricted to a homogenous terrain. In practice, however, due to sparse radiosonde measurements and complex terrain,

calculations of trajectories are mostly based on gridded output from numerical models.

Previously, backward trajectories were mainly used in climatological studies (Miller 1981a, Miller 1981b). Recently, due to dynamic development of computational tools, several modern methods of determining distant sources of air pollution emission have been introduced (Stohl 1998) including different methods of backward trajectories analysis. Initially, these methods have ignored inhomogeneity of emission sources, indicating only potential contribution of air masses from different regions in air pollution occurring near receptor (White et al. 1994, Moody et al. 1995). Analysis of air particles residence time in polluted areas turned out more useful in solving air pollution issues. Such analysis is based on either estimating probability of finding an air particle in a given position during a given time period (Ashbaugh 1983, Ashbaugh et al. 1985) or calculating logarithmic mean pollutant concentration for each grid cell of trajectories domain (Seibert et al. 1994). However, these methods underestimate spatial changeability of emission since all sections of trajectories are treated in the same way. A different approach

to the matter is the method proposed by Stohl (Stohl 1996). It is based on redistribution of pollutant concentration fields. Out of all trajectories passing through model grid nodes, the subset resulting in low level of pollutant concentration in receptor is chosen. Hence, it can be concluded that there are no significant emission sources within selected grid cells. Then, out of all trajectories related with high level of pollutant concentration in receptor, only grid cells not belonging to the earlier mentioned subset are taken into account. Stohl's idea supported by spatial analysis has been developed in this paper. Spatial analysis limits uncertainty of results caused by trajectories errors, and it could overcome difficulty of separating local and distant emission sources.

There are several sources of errors in determination of trajectory. Some of them are connected with calculations using discrete spatio-temporal data and involving data on land use, terrain and meteorological conditions. In practice, land use and terrain data characterise each grid cell as a whole, while meteorological data are inherently discrete. Furthermore, some processes, especially those concerning planetary boundary layer (PBL), are not truly resolved but only parameterised. It results in errors related to imperfection of physical parameterisations. Ultimately, trajectory calculations are based on rough gridded output of numerical models. Another type of errors are so-called truncation errors which occur because of neglecting higher-order terms in Taylor series during approximate solving of the differential trajectory equation. Many authors tried to estimate errors deriving from different sources. Kahl and Samson (Kahl and Samson 1986) found out that mean error of 3–4 m/s caused by spatial interpolation of horizontal wind component at an altitude of 1000 m results in mean trajectory position error of 400 km after 72 h travel and of 100 km after 24 h travel. During highly convective conditions mean wind interpolation error rises to 5 m/s causing mean trajectory position error of 500 km after 72 h travel (Kahl and Samson 1988). The truncation errors of position trajectories after 42 h increase from 100 km for constant acceleration to 300 km for zero acceleration (Walmsley and Mailhot 1983). The mentioned errors change each trajectory into 3-dimensional shape similar to cone with a vertex in a point where the trajectory starts. The calculated trajectory is of maximal probability of the real one. However, probability of computing correctly the real backward trajectory decreases significantly in time together with increasing an area where the trajectory may be found. The paper presents a methodology of identifying distant emission sources of fine particulate matter $PM_{2.5}$ involving spatial analysis of aggregate information on $PM_{2.5}$ concentrations and air masses backward trajectories.

Methodology

The backward trajectories of air masses were computed using the HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model version 4 (Draxler and Hess 1997, Draxler and Hess 1998, Draxler 1999) accessed via web-based Real-time Environmental Applications and Display sYstem (READY) developed by NOAA's Air Resources Laboratory (ARL) (Draxler and Rolph 2013, Rolph 2013). The HYSPLIT model is a complete system for computing simple air parcel trajectories to complex dispersion and deposition simulations. There is a possibility of using few input meteorological data,

e.g. GDAS, REANALYSIS, etc. Depending on data applied there will be differences in calculated trajectories. In the study, the calculations were done basing on meteorological data from the Global Data Assimilation System (GDAS). The GDAS is run operationally 4 times a day (at 00, 06, 12, and 18 UTC) by the NOAA's National Centers for Environmental Prediction (NCEP). Model output is for the analysis time and 3, 6, and 9-hour forecasts. NCEP post-processing of the GDAS converts the data from spectral coefficient form to 1 degree latitude-longitude (360 by 181) grids and from sigma levels to mandatory pressure levels. NOAA's Air Resources Laboratory (ARL) saves the successive analyses and 3-hour forecast, 4 times each day to produce a continuous data archive. Some fields such as precipitation and surface fluxes are not available at the analysis time, therefore these are taken from the 6-hour forecast files. A new three-dimensional variational (3D-Var) data assimilation analysis system called the Gridpoint Statistical Interpolation (GSI) is incorporated into the GDAS package. The GSI improves performance of meteorological analysis (Kleist et al. 2009). The GDAS archive is set up as default input meteorological data in HYSPLIT model. The decision to use these data was caused by enhanced data assimilation methods as well as the highest horizontal, vertical and temporal resolution.

The 48 h backward trajectories were calculated every 1 h for the period of January–March 2010 starting in the selected $PM_{2.5}$ measurement locations at heights of 30, 50, 100 and 150 m a.g.l. The selection of the period and the locations was associated with a research experiment conducted to analyse the state of PM_{10} and $PM_{2.5}$ air pollution taking into account the chemical composition of dust and natural sources (Klejnowski et al. 2011).

It was decided to start the trajectories at several heights because pollutants inside PBL are subjected to homogenisation processes by convective or mechanical mixing. In winter there are observed similar conditions for processes within PBL, especially those related to day length and Sun altitude at noon. This season is characterised by long-lasting ground-based inversions (also during daytime) and a weak convective mixing mainly around the midday. Because of the weak air mixing it is assumed that only backward trajectories low-lying near starting points affect air quality at receptors. It is supposed that emission of pollutants influences their measured concentrations near surface only when backward trajectory starts within mixing layer or ground inversion layer. Therefore, the heights of trajectories starting points are situated in the bottom part of PBL. Meteorological data used in HYSPLIT model are of low spatial resolution. Thus, potential modifying effect of local phenomena on air flow trajectory may be not taken into account, especially when backward trajectory starts low above the ground. Errors related to this problem were attempted to be reduced by analyzing backward trajectories starting at different heights a.g.l., deleting selected parts of trajectories, and averaging $PM_{2.5}$ concentrations spatially.

A concentration of $PM_{2.5}$ measured at a certain time and a certain location was assigned to each point of the backward trajectory starting at respective time and location. Only those sections of the trajectories from the starting point to a point with a height above 10 m a.g.l. and/or precipitation lower than 0.2 mm/h were left. It was assumed that below given height dry deposition would significantly increase and precipitation

could totally remove $PM_{2.5}$ from the atmosphere. As a result there would be no connection between the emission in regions through which a deleted section of the trajectory passed and $PM_{2.5}$ concentration measured at the receptor.

This study attempted to assess contribution of distant emission sources to winter $PM_{2.5}$ air pollution measured at three monitoring stations: Diabla Góra, Gdańsk and Katowice. Table 1 presents geographic location and type of the selected stations as well as averaging times of $PM_{2.5}$ measurement taken at the stations. The Diabla Góra station represents rural background conditions. Diabla Góra is a small village located in the Borecka Forest on the north-east of Poland. Two other stations Gdańsk and Katowice represent urban background conditions. Gdańsk is a coastal city and seaport located in northern Poland, forming together with Gdynia and Sopot the Tricity agglomeration with a population of approximately 740 000. Katowice is a city located on upland area in southern Poland, which is a centre of the high-industrialised Upper-Silesian agglomeration of 14 cities and towns with a population approximately 2 000 000.

Each point of a single backward trajectory was assigned to the $PM_{2.5}$ concentration measured at a certain time and location. In the case of using hourly $PM_{2.5}$ concentration data the particular value was assigned to all points of the trajectory starting at the certain hour. In the case of using daily mean $PM_{2.5}$ concentrations the one value was assigned to all points of all trajectories for that day. In this study the analysis was performed mainly on the basis of daily $PM_{2.5}$ measurement data, because such kind of data was available for all selected stations. Moreover, for the Gdańsk station, where hourly averages of $PM_{2.5}$ concentration were measured, a comparison of results obtained using hourly and daily $PM_{2.5}$ data was performed.

The presented method of identifying distant emission sources of fine particulate matter is based on aggregation of information about the backward trajectories and the measured $PM_{2.5}$ concentrations. For each starting location and height all the data from considered winter months of 2010 were analysed together. If in some areas there can be found only backward trajectories assigned to high $PM_{2.5}$ concentrations measured at a starting point, there is high probability of existing $PM_{2.5}$ emission source in those areas. On the other hand, if trajectories passing through some areas are assigned to large $PM_{2.5}$ concentrations only occasionally, it may be assumed that there are no emission sources in those areas. By analysing $PM_{2.5}$ data in the vicinity of a point, it can be determined if there is an emission source in this point.

Locations of distant sources of fine particulate matter emission were determined using a data metrics gridding method. A mean value of $PM_{2.5}$ concentrations (Z mean value) associated with the trajectories points located within the so-called search ellipse was assigned to each grid node. Instead of Z mean value, lower and upper quartiles as well as median values can be used (Godłowska 2010). The search ellipse defines the local neighbourhood of each grid node. In this study the grid node spacing 0.05° in longitude and 0.025° in latitude were used, and radii of the search ellipse 0.56° and 0.33° respectively were determined by experiments. Regarding the radii of the search ellipse an effect of the trajectory position error should be considered. Thus, a maximal number of points taken to the calculation of Z mean value in the search ellipse was limited causing gradual decrease of the ellipse radii towards the backward trajectory starting location. Furthermore, in order to ensure statistical significance of results a minimal number of points taken to the calculation of Z mean value in the search ellipse was set to 30. For the elimination of nearby sources of fine particulate matter emission, before calculating the mean value of $PM_{2.5}$ concentrations, the trajectories points were filtered according to the criterion of so-called duplicates, i.e. when the distance between two points is lower than 3.6 km. Such distance corresponds to the trajectory position error achieved after 1 h and caused by the wind speed error of 1 m/s. The duplicate point associated with higher $PM_{2.5}$ concentration is then removed.

Results

The number of the 48 h backward trajectories points used in finding distant emission sources depends on atmospheric conditions occurring in the area through which a particular trajectory passes. A low height of air particle and greater precipitation characterising a trajectory point determine deleting a section of this trajectory following for that point. Filtering the duplicate points further reduces the number of the backward trajectories points useful in the spatial analysis (Table 2). The higher the trajectory starting height, the more the remaining points after subsequent stages of data post-processing.

Before filtering the trajectories duplicate points and removing the points with higher $PM_{2.5}$ concentrations, the backward trajectories points with the assigned adequate $PM_{2.5}$ concentrations were plotted on a map of national borders. In the paper the plots of the backward trajectories started at a height of 150 m a.g.l. in Diabla Góra (Fig. 1), Gdańsk

Table 1. Characterisation of the air pollution monitoring stations

Monitoring station	Latitude	Longitude	Altitude [m a.s.l.]	Station type	$PM_{2.5}$ measurement averaging time
Diabla Góra (the Borecka Forest)	54°07'42"	22°02'17"	157	rural background	24 h
Gdańsk (Leczkowa st.)	54°22'49"	18°37'13"	40	urban background	24 h
					1 h
Katowice (Kossutha st.)	50°15'56"	18°58'40"	274	urban background	24 h

(Fig. 2) and Katowice (Fig. 3) are presented. The trajectories points are grouped with respect to the $PM_{2.5}$ concentration assigned to them. Similar patterns were observed for the other starting heights. In the cases of Diabla Góra and Gdańsk the trajectories assigned to the highest $PM_{2.5}$ concentrations mostly followed backward to eastern Europe. In the case of Katowice most trajectories followed backward to south-eastern Europe.

The number of the trajectories points for Katowice taken into account in the spatial analysis was about two times less than those for Diabla Góra or Gdańsk. It was due to the fact that the trajectories frequently descended below 10 m a.g.l. or passed through regions with precipitation.

Gridding of the backward trajectories points associated with the $PM_{2.5}$ concentrations, together with blanking part of

Table 2. Numbers of the backward trajectories points (N) calculated for January–March 2010, after subsequent stages of data post-processing

Monitoring stations	$PM_{2.5}$ measurement averaging time	Starting height [m]	N after including $PM_{2.5}$ data	N after deleting trajectories sections with low height and precipitation	N after filtering duplicate points
Diabla Góra (the Borecka Forest)	24 h	30	102312	52863	17675
		50	102312	58964	22342
		100	102312	62449	28197
		150	102312	63234	30970
Gdańsk (Leczkowa st.)	24 h	30	102312	62005	24644
		50	102312	66920	27799
		100	102312	71180	31441
		150	102312	71686	33291
	1 h	30	102312	62005	24602
		50	102312	66920	28439
		100	102312	71180	33880
		150	102312	71686	36047
Katowice (Kossutha st.)	24 h	30	99960	25903	9319
		50	99960	28860	10702
		100	99960	35357	14030
		150	99960	39694	16770

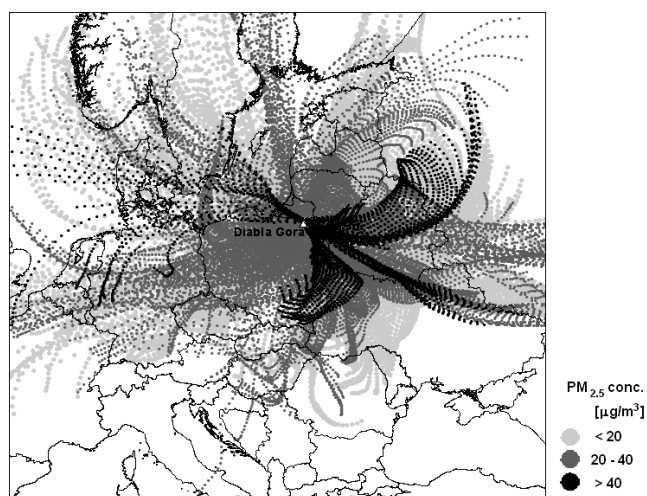


Fig. 1. The backward trajectories from January–March 2010 started in Diabla Góra at a height of 150 m a.g.l., after deleting their sections with the low height and the precipitation, grouped with respect to the $PM_{2.5}$ concentration assigned to the trajectories points

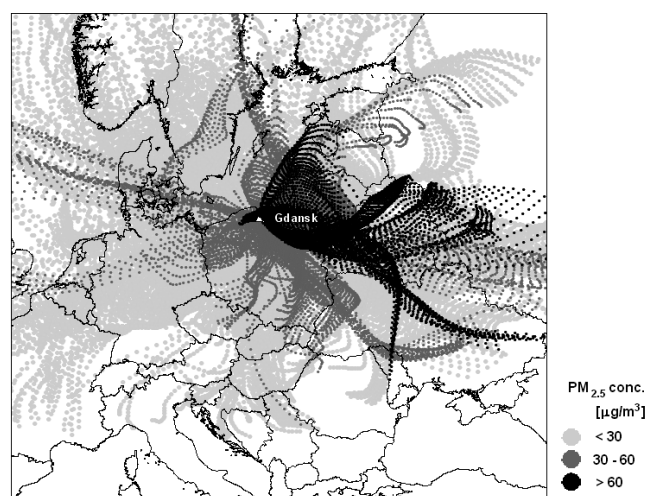


Fig. 2. The backward trajectories from January–March 2010 started in Gdańsk at a height of 150 m a.g.l., after deleting their sections with the low height and the precipitation, grouped with respect to the $PM_{2.5}$ concentration assigned to the trajectories points

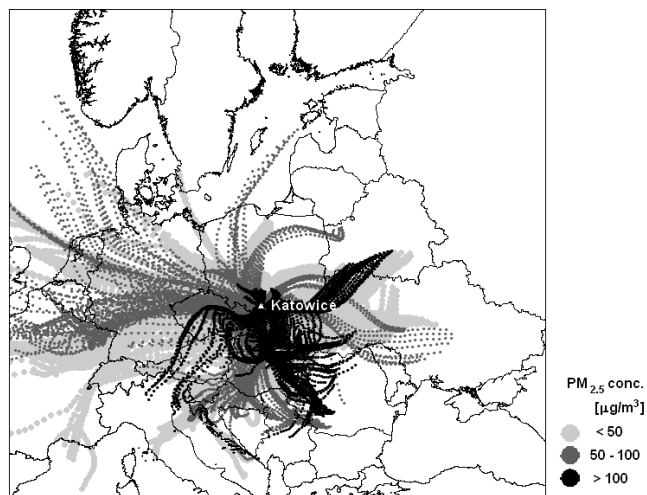


Fig. 3. The backward trajectories from January–March 2010 started in Katowice at a height of 150 m a.g.l., after deleting their sections with the low height and the precipitation, grouped with respect to the PM_{2.5} concentration assigned to the trajectories points

the data to eliminate nearby sources of fine particulate matter emission, resulted in the spatial distribution of the mean PM_{2.5} concentration which was plotted on the national borders map separately for the considered locations and the trajectories starting heights. By means of the plotted spatial distributions, distant sources of fine particulate matter emission influencing the PM_{2.5} concentrations measured in Diabla Góra, Gdańsk and Katowice were identified. In the paper the results for the trajectories starting heights of 50 and 150 m a.g.l. are presented (Figs 4–7).

Distant PM_{2.5} emission sources affecting Diabla Góra were identified mainly in northern Belarus, western Russia near Russian-Belarusian and Russian-Estonian borders, as well as in south-eastern part of Poland near Polish-Slovakian border (Fig. 4). Gdańsk was affected by distant sources of fine particulate matter emission mostly similar to those for Diabla Góra (Fig. 5–6), i.e. located in northern Belarus and western

Russia near Russian-Belarusian border. Additionally, the emission sources near Polish-Ukrainian border and in Ukraine were identified. The results obtained for Gdańsk on the basis of 1 h and 24 h PM_{2.5} concentrations are comparable. In the case of the 1 h data, regions of emission sources are better separated from the other ones. The results of identification of distant PM_{2.5} emission sources influencing Katowice (Fig. 7) are partly similar to those obtained earlier for the winter months at the turn of 2005 and 2006 (Godłowska 2010). The emission sources located in Romania and near Romanian-Hungarian border were identified in both cases. Moreover, in this study the sources situated in Slovakia, Ukraine and near Belarusian-Ukrainian border were identified.

Discussion

It was noticed that distant PM_{2.5} emission sources located in different regions affected southern and northern parts of Poland in the winter months of 2010. The PM_{2.5} concentrations in Gdańsk and Diabla Góra are only marginally affected by the emission sources situated in the heavily industrialised southern part of Poland. On the other hand, the PM_{2.5} concentrations in Katowice are affected mainly by the sources located in countries located south of Poland. However, the fine particulate matter emission sources in Belarus seem to be among the most important for all analysed places. The accuracy of identifying distant emission sources depends on the number of backward trajectories points used in the analysis. The further from the trajectories starting point, the lower the points density, and hence the increasing uncertainty in the results. The setting of minimal number of trajectories points taken to the calculation of PM_{2.5} mean concentration in the search ellipse ensures statistical significance but eliminates additional points from spatial analysis resulting in no-data grid nodes. The use of 1 h data is recommended for identifying distant PM_{2.5} emission sources. However, the results obtained for Gdańsk suggest that 24 h data equally with 1 h data can be used. Through blanking part of the data to eliminate nearby sources of fine particulate matter emission the methodology presented is suitable not only for rural background measurement stations but also for urban ones.

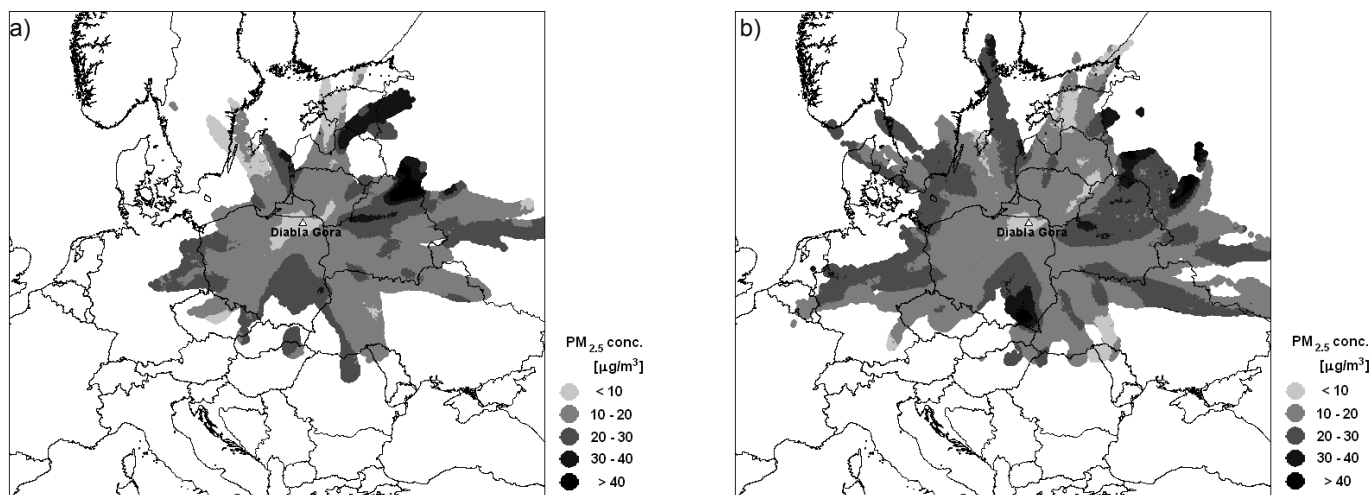


Fig. 4. Spatial distribution of the mean PM_{2.5} concentration calculated with the use of the backward trajectories from January–March 2010 started at the heights of 50 m a.g.l. (a) and 150 m a.g.l. (b), and the 24 h PM_{2.5} concentrations measured in Diabla Góra assigned to the trajectories points

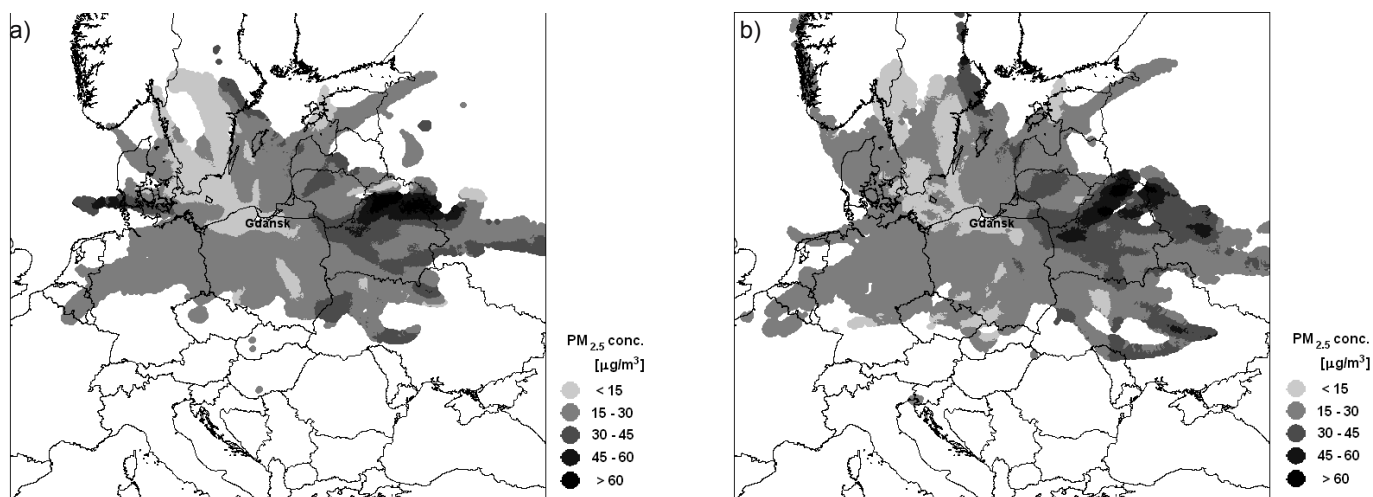


Fig. 5. Spatial distribution of the mean $PM_{2.5}$ concentration calculated with the use of the backward trajectories from January–March 2010 started at the heights of 50 m a.g.l. (a) and 150 m a.g.l. (b), and the 24 h $PM_{2.5}$ concentrations measured in Gdansk assigned to the trajectories points

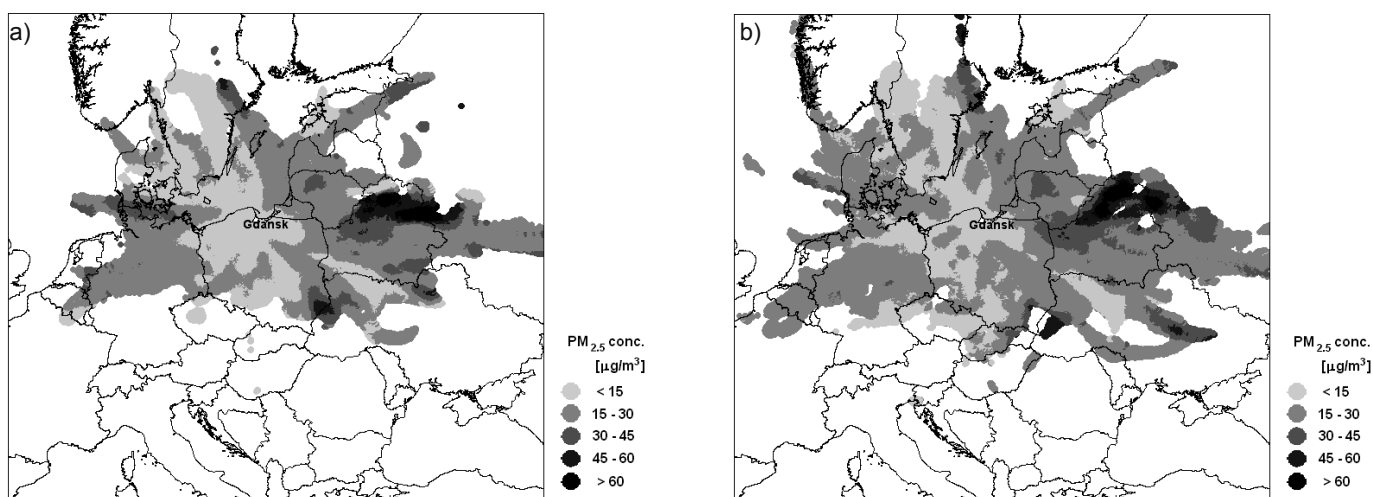


Fig. 6. Spatial distribution of the mean $PM_{2.5}$ concentration calculated with the use of the backward trajectories from January–March 2010 started at the heights of 50 m a.g.l. (a) and 150 m a.g.l. (b), and the 1 h $PM_{2.5}$ concentrations measured in Gdansk assigned to the trajectories points

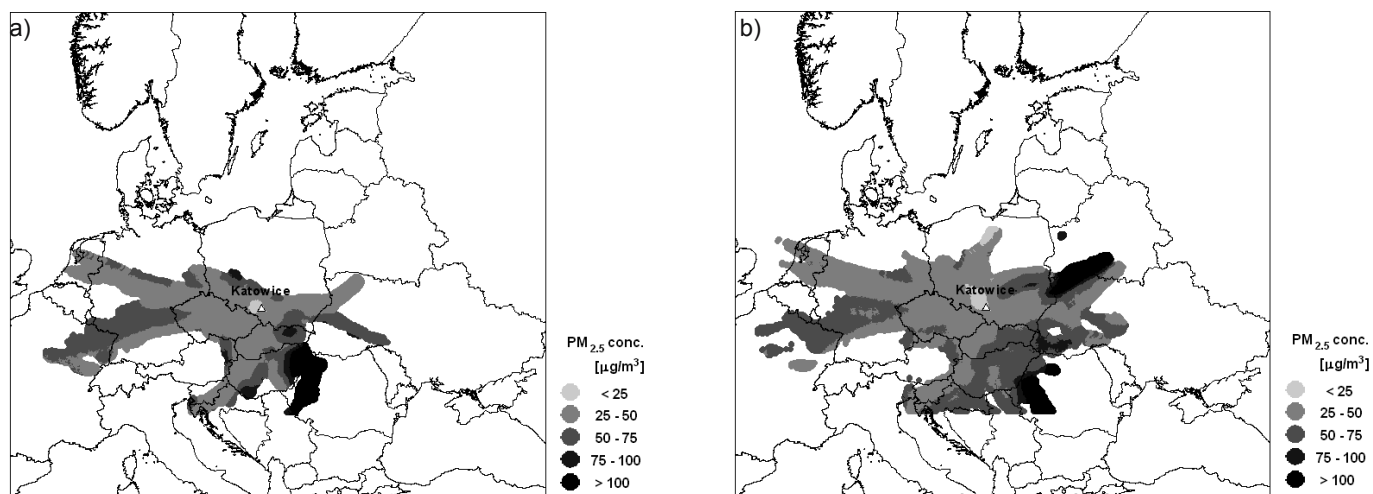


Fig. 7. Spatial distribution of the mean $PM_{2.5}$ concentration calculated with the use of the backward trajectories from January–March 2010 started at the heights of 50 m a.g.l. (a) and 150 m a.g.l. (b), and the 24 h $PM_{2.5}$ concentrations measured in Katowice assigned to the trajectories points

The results shown in Figs 4–7 were compared to the gap-filled and gridded PM_{2.5} emission data from 2010 (Fig. 8) accessed via the Centre on Emission Inventories and Projections (CEIP) Website (CEIP Website 2014). The particulate matter emission data for Europe are collected and processed in the frame of Co-operative Programme for Monitoring and Evaluation of Long Range Transmission of Air Pollutants, shortly called European Monitoring and Evaluation Programme (EMEP). Fig. 8 presents the PM_{2.5} emission data summed for all types of sources (e.g. industrial, traffic, agricultural) in a 0.5° × 0.5° grid.

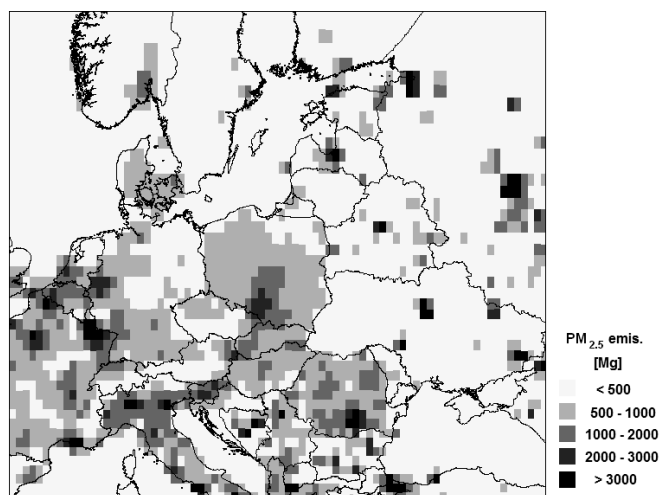


Fig. 8. The gap-filled and gridded PM_{2.5} emission data from 2010 by EMEP-CEIP

The comparison of the results to the EMEP-CEIP emission spatial data approximately confirmed the distant PM_{2.5} emission sources identified for Diabla Góra, Gdańsk and Katowice, especially those located in Belarus, Romania and Ukraine. Sources of the greatest emission were not identified

precisely probably due to insufficient trajectories points within their locations. It should be also noted that EMEP-CEIP PM_{2.5} inventoried emission was derived from different (i.e. point, mobile, area) sources. Furthermore, there was no sufficient information on PM_{2.5} emission from countries not associated in the European Union.

The earlier obtained results of identifying the distant PM₁₀ emission sources for Małopolska (Lesser Poland) and Górny Śląsk (Upper Silesia) in the 2005/2006 winter months (Godłowska 2010) indicated that using the 72 h backward trajectories started in many neighbouring measurement stations could provide more precise locations of distant particulate matter emission sources. However, using the 48 h backward trajectories the trajectory position errors are smaller. Accuracy of the method may be also improved by taking into account longer period of time, e.g. several winters. The number of useful trajectories points will raise if the starting height is greater.

Summary and conclusions

The presented methodology of identifying distant sources of fine particulate matter emission enables the determination of regions potentially affecting a given PM_{2.5} measurement location. The method involves spatial analysis of aggregate data on measured PM_{2.5} concentrations and air masses backward trajectories calculated by HYSPLIT model. The key stages of the data post-processing are: 1) deleting trajectories sections following points low-lying or located within precipitation; 2) filtering duplicate data and removing those with higher air pollutant concentration; 3) gridding XYZ data with use of Z mean value calculated in the search ellipse with properly defined latitude and longitude radii.

The presented method first of all may be applied in assessment of air pollution long-range transport affecting measured concentrations of pollutant like PM_{2.5} regardless of their levels.

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SURFER software version 9 by Golden Software, Inc. was used in data gridding.

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Analiza przestrzenna wstecznych trajektorii mas powietrza w celu rozpoznania odległych źródeł emisji pyłu drobnego

W pracy zaprezentowano metodę rozpoznawania odległych źródeł emisji pyłu drobnego PM_{2.5}, polegającą na przestrzennej analizie łącznej informacji o imisji PM_{2.5} oraz o wstecznych trajektoriach mas powietrza, obliczonych za pomocą modelu HYSPLIT. Trajektorie wsteczne obliczono startując z wysokości 30, 50, 100 i 150 m n.p.g., dla trzech lokalizacji stacji pomiarowych stężeń PM_{2.5} (Diabla Góra, Gdańsk, Katowice), reprezentujących różne warunki środowiskowe. Wszystkim punktom pojedynczej trajektorii wstecznej przyporządkowano stężenie PM_{2.5} odpowiadające dacie startu obliczeń tej trajektorii. Użyto dobowych średnich stężeń PM_{2.5}, a w przypadku Gdańska dodatkowo także godzinnych średnich, co umożliwiło ocenę skuteczności przedstawionej metody. Położenie odległych źródeł emisji pyłu drobnego zostało określone poprzez interpolację danych punktowych trajektorii do regularnej siatki przy zastosowaniu metody metryki danych. Każdemu węzłowi siatki przypisano wartość średnią obliczoną ze stężeń PM_{2.5} przyporządkowanych punktom trajektorii znajdujących się w obrębie tzw. elipsy wyszukiwania. Przed obliczeniem wartości średniej ukryto część danych, eliminując w ten sposób bliskie źródła emisji pyłu drobnego. Analizy objęły okres styczeń–marzec 2010 roku. Wyniki wskazały na odmienne pochodzenie mas powietrza w północnej i południowej Polsce. W Diabli Górze i Gdańsku odległe źródła emisji pyłu drobnego rozpoznano głównie w Białorusi i Rosji. W Katowicach również zaznaczył się wpływ źródeł białoruskich, ale jako najbardziej istotne odległe źródła emisji PM_{2.5} uznano te zlokalizowane na obszarze Rumunii, Węgier, Słowacji i Ukrainy.