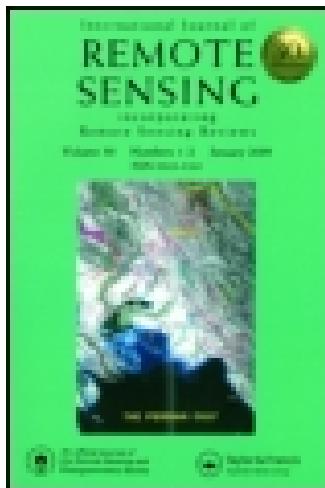


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Localization of aerosol sources in East-European region by back-trajectory statistics

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A concentration-weighted trajectory method for aerosol source localization based on joint statistical analysis of aerosol column volume concentrations and back-trajectory data was used to estimate the spatial distribution of aerosol sources in the East-European region. The aerosol column volume concentration data measured at five AERONET network sites, Belsk, Minsk, Kyiv, Moldova/Kishinev, and Sevastopol, were used. The geographical areas responsible for increased aerosol content at the monitoring sites were mapped separately for coarse-mode and fine-mode aerosol fractions. The investigated area is located between 42° and 62° N in latitude and between 12° and 50° E in longitude.

It was shown that the northeastern territories (in relation to the monitoring stations) give a small contribution to the coarse-mode aerosol content. The events of increased coarse-mode aerosol concentration have been caused by sources in the southeastern regions. On average, the air masses with a large content of coarse-mode aerosol particles were delivered to all stations from regions around Donetsk, Rostov-on-Don, and Kharkiv cities. The fine-mode aerosol fraction originated from areas of Tambov, Voronezh, and Kharkiv cities. The calculated aerosol source regions partly correspond to European Monitoring and Evaluation Programme data for eastern Europe. The cause of difference between calculated regions responsible for increased aerosol content at the monitoring sites and the sources of particle emission according to European Monitoring and Evaluation Programme data are discussed.

1. Introduction

The localization of the aerosol source areas and the sources of emission power estimation are important problems of environment control (Marchuk 1982). Using the aerosol content data and the corresponding meteorological information, it is possible, in principle, to solve the inversion problem and to study the release capacity distribution of aerosol sources (Raputa and Krylova 1995) if the relationship between a source and the aerosol concentration at the monitoring site is known.

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This relationship can be obtained on the basis of aerosol transport modelling. Because the inversion solution is usually an ill-posed problem, the accuracy requirements of the mentioned the relationship must be high enough. Thus, besides high accuracy of the transport model, detailed knowledge of many input parameters is needed, which is usually a difficult task in many practical cases. Therefore, for the investigation of geographical distribution of atmospheric aerosol source areas, the trajectory statistics method proposed by Ashbaugh (1983) and Ashbaugh, Malm, and Sadeh (1985) has been widely used as an alternative to the methods based on the transport model.

Long-term aerosol monitoring data serve as the base for the trajectory statistics method. The back-trajectories of air masses that arrive at the monitoring site strongly deviate from rectilinear directions, and many back-trajectories are intersected under different angles over the source region (Dorling, Davies, and Pierce 1992; Bösenberg et al. 2003). This feature allows determination of the position of the source using monitoring data from a single station (Ferrarese 2002). A large number of measurements at the monitoring site and a set of back-trajectories enable the establishment of a certain statistical connection between the aerosol concentration level at the monitoring site and the territories that the air masses passed over before arrival at the monitoring site. Therefore, this allows localization of the regions where the increased concentrations of aerosol are produced (Ashbaugh 1983; Ashbaugh, Malm, and Sadeh 1985).

Different trajectory statistics methods have been used to reveal the region sources of sulphur oxides and sulphates (Ashbaugh, Malm, and Sadeh 1985; Rúa et al. 1998; Zhao, Hopke, and Zhou 2007; Stohl 1996; Poirot and Wishinski 1986), nitrogen oxides and nitrates (Rúa et al. 1998; Zhao, Hopke, and Zhou 2007), ozone (Wotawa, Kröger, and Stohl 2000), acid precipitations (Charron et al. 2000), aerosol (Seibert et al. 1994; Wang, Zhang, and Arimoto 2006; Chen et al. 2011; Zhou, Hopke, and Liu 2004; Zhao and Hopke 2006; Tositti et al. 2013; Xu et al. 2013; Aher et al. 2014), sources and sinks of CO₂ (Apadula et al. 2003), radioactive Be⁷ (Lee et al. 2004), soot particles (Sharma et al. 2006), and persistent organic pollutants (Du and Rodenburg 2007; Hsu, Holsena, and Hopke 2003; Sofuoğlu et al. 2013) in different areas of the planet.

To reveal the atmospheric aerosol pollution sources in Belarus by the back-trajectory statistics method, the aerosol column volume concentration data at the AERONET site in Minsk (Holben et al. 1998, 2001; Giles et al. 2012) along with local monitoring data of surface air aerosol in the Berezinsky biosphere reserve at a distance of 110 km from Minsk city, Belarus, from the <http://scat.bas-net.by/~lidarteam/Transboundary%20transport-ru/> web site, for 2004–2006 were used in the paper by Kabashnikov et al. (2008). The aerosol measurements at the AERONET Belsk site (50 km southwest of Warsaw, Poland) and the Moldova/Kishinev site (in Kishinev city, Moldova) were used to retrieve validation and comparison. The seasonal features of aerosol behaviour in the East-European region, in particular over the Ukrainian urban-industrial areas and the Minsk site, were also investigated by Milnevsky et al. (2014) with the application of back-trajectory analysis.

The purpose of this work is to identify the main geographical regions that are the sources of particulate matters arriving at the AERONET network sites in Belsk, Kyiv, Moldova/Kishinev, Minsk, and Sevastopol in the East-European region on the basis of respective data from aerosol monitoring.

In contrast to Kabashnikov et al. (2008), where results of the analysis of total (fine-plus coarse-mode) aerosol values were reported, in this paper we separate the sources of fine- and coarse-mode aerosol. In addition, this paper is based on much longer time series data from January 2004 till December 2011. Besides, we use the homogeneous data of aerosol column content.

2. Data and method

We used data obtained by sun photometers at five monitoring sites in the East-European region (Belsk, Minsk, Kyiv, Moldova/Kishinev, and Sevastopol) of the AERONET international network (Holben et al. 1998, 2001; Milinevsky 2013), the locations and details of which provided in Figure 1 and Table 1, respectively.

The aerosol column volume concentration in the atmosphere was estimated on the basis of aerosol optical thickness measurements made by sun photometers CIMEL CE318 (Holben et al. 2001; Giles et al. 2012). Aerosol volume concentration of total VolCon-T, fine-mode VolCon-F (particles with a size $<1\text{ }\mu\text{m}$), and coarse-mode VolCon-C (particles with a size $>1\text{ }\mu\text{m}$) Level 1.5 cloud-screened and outlier-removed data at the five AERONET monitoring sites were obtained from the <http://aeronet.gsfc.nasa.gov/> website, where data are given in $\mu\text{m}^3\text{ }\mu\text{m}^{-2}$, which determine the volume of all aerosol particles in a vertical atmosphere column of $1\text{ }\mu\text{m}^2$ cross-section. In this paper, we used Level 1.5 instead of the better quality Level 2.0 cloud-screened and quality-assured data due to less availability of Level 2.0 data for several sites.



Figure 1. Locations of AERONET sites Belsk, Minsk, Kyiv, Sevastopol, and Moldova/Kishinev (triangles) considered in the paper.

Table 1. AERONET sites in the East-European region selected for analysis.

Monitoring site, country	Latitude	Longitude	Altitude, m a.s.l.
Belsk, Poland	51.84° N	20.79° E	190
Minsk, Belarus	53.92° N	27.60° E	200
Kyiv, Ukraine	50.45° N	30.50° E	200
Moldova/Kishinev, Moldova	47.00° N	28.82° E	205
Sevastopol, Ukraine	44.62° N	33.52° E	80

In our investigation, three-dimensional back-trajectories from the NASA/Goddard <http://aeronet.gsfc.nasa.gov> website (Schoeberl and Newman 1995; Pickering et al. 2001) were extracted for four monitoring sites: Belsk, Moldova/Kishinev, Minsk, and Sevastopol. The trajectory calculations are based on the trajectory code developed from NASA/Goddard Code 614 of NASA Global Modelling Assimilation Office assimilated gridded analysis data over the period 2000–2007 and National Centers for Environmental Prediction analyses over the period 2007–2011. The back-trajectories for the Kyiv site were not available at the NASA/Goddard site and were calculated on the basis of meteorological data supplied by Republican Hydrometeorological Centre of the Republic Belarus (www.hmc.by). All atmospheric trajectories were calculated for two arrival times (00UT and 12UT) and for eight arrival pressure levels between 950 and 200 hPa.

In the method of trajectory statistics, the geographical region surrounding a receptor is divided into cells by a grid. Each cell is marked with (i, j) , where indices i and j are geographical coordinates (longitude and latitude) of the cell centres. We designated $c(l)$ as an air impurity concentration (in our case, it is an aerosol column volume concentration) that can be registered in the monitoring point (receptor) at the moment of l -measurement. Parameter $\tau_{i,j}(l)$ is the time range that the air mass has spent in the grid cell (i, j) moving along back-trajectory, which starts from the monitoring point at the moment of l -measurement. We set to each (i, j) cell the corresponding aerosol volume mean concentration value $P_{i,j}$ measured at the monitoring site under the conditions in which the back-trajectory passes over that cell:

$$P_{i,j} = \left(\sum_{l=1}^L c(l) \tau_{i,j}(l) \right) / \sum_{l=1}^L \tau_{i,j}(l), \quad (1)$$

where L is a total number of measurements. This version of the back-trajectory statistics is known as a concentration-weighted trajectory (CWT) method (Hsu, Holsena, and Hopke 2003).

A parameter $P_{i,j}$ has physical meaning of conditional mean aerosol volume concentration that, on average, appears at the receptor when the air mass passes above the (i, j) cell before arriving at the receptor. The cells with a high concentration $P_{i,j}$ are considered as possible sources of aerosol contamination of the atmosphere over the monitoring site.

A conditional mean aerosol volume concentration can be considered as a parameter that allows estimation of the level of potential influence of different geographical regions on aerosol content measured at the monitoring site. It takes into account the real conditions of the air mass transport and gives a first approximation of the localization pattern for the most affecting sources which are responsible for the highest aerosol concentrations at the monitoring site.

In the case of data from several (M) monitoring sites, the conditional mean aerosol volume concentration was determined as:

$$P_{i,j} = \left(\sum_{m=1}^M \sum_{l=1}^L c^m(l) \tau_{i,j}^m(l) \right) / \sum_{m=1}^M \sum_{l=1}^L \tau_{i,j}^m(l) = \sum_{m=1}^M P_{i,j}^m G_{i,j}^m, \quad (2)$$

where m is the monitoring site index, M is the number of monitoring sites, and $P_{i,j}^m$ and $G_{i,j}^m$ are:

$$P_{i,j}^m = \sum_{l=1}^L c^m(l) \tau_{i,j}^m(l) \Bigg/ \sum_{l=1}^L \tau_{i,j}^m(l), \quad (3)$$

$$G_{i,j}^m = \sum_{l=1}^L \tau_{i,j}^m(l) \Bigg/ \sum_{m=1}^M \sum_{l=1}^L \tau_{i,j}^m(l). \quad (4)$$

It is seen from Equation (2) that the conditional mean aerosol volume concentration value estimated from data of several monitoring sites is the sum of the conditional mean aerosol volume concentration values retrieved from each monitoring site ($P_{i,j}^m$), with the weights ($G_{i,j}^m$) equal to the relations of residence time of back-trajectories, started from a particular site, to the residence time of trajectories started from all monitoring sites.

The back-trajectory data (see Section 2) are available for arrival times 00UT and 12UT only. In cases when the moment of concentration measurement differed from 00UT or 12UT, we used the data obtained at the time nearest to 00UT or 12UT.

The accuracy of the trajectory statistics method can be defined in terms of Spearman's rank correlation between the reconstructed and true source distributions (Wilks 2011). Spearman's rank correlation coefficient is used to reveal the strength of a link between two sets of data. To calculate Spearman's rank correlation coefficient, we have to arrange all conditional mean column aerosol volume concentrations $P_{i,j}$ in increasing order and find a position number $N_{i,j}$ of every $P_{i,j}$ in the rank. Similarly, we calculated ranks of true sources in cells. Spearman's rank correlation coefficient is a Pearson correlation coefficient calculated on these ranks (Wilks 2011).

The accuracy of the trajectory statistics method has been investigated in a number of papers (Wotawa and Kröger 1999; Scheifinger and Kaiser 2007; Kabashnikov et al. 2011). In Kabashnikov et al. (2011), three well-known trajectory statistics methods, namely concentration field, CWT, and potential source contribution function, were tested using known sources and artificially generated data sets to determine the ability of trajectory statistics methods to reproduce spatial distribution of the sources. In real-world experiments, trajectory statistics methods are intended for application to *a priori* unknown sources. Therefore, the accuracy of trajectory statistics methods was tested with all possible spatial distributions of sources. An ensemble of geographical distributions of virtual sources was generated. Spearman's rank order correlation coefficient between spatial distributions of the known virtual and the reconstructed sources was taken to be a quantitative measure of the accuracy. Unfortunately, it is impossible to estimate the accuracy of the particular reconstructed source distribution. Statistical estimates of the mean correlation coefficient and a range of the most probable values of correlation coefficients were obtained. All the trajectory statistics methods that were considered showed similar close results. We can only estimate the mean accuracy averaged over an ensemble of possible true source distributions, which is done in Section 3. As shown in Kabashnikov et al. (2011), the accuracy of the method can reach values of 70–75% on average, when the source distribution is reconstructed within an optimal area with a size equal to the distance over which the air mass passes during the aerosol lifetime in the atmosphere.

From the numerous lidar data on the vertical distribution of aerosol backscatter at different sites, the main amount of aerosol particles within the planetary boundary layer is located mostly between altitudes of 1–2 km (see, e.g. Bösenberg et al. 2003; Balin and Ershov 1999; Matthias and Bösenberg 2002 on this subject). In spite of the fact that all

aerosol particles of the entire atmosphere column contribute into the volume concentration determined from AERONET sun photometer measurements, we assumed that the large-scale transport of the atmospheric aerosol within the boundary layer can be traced by back-trajectories started at 950 hPa (about 450 m a.s.l.) and at 850 hPa (about 1300 m a.s.l.) altitudes. So the back-trajectories start from the monitoring site location in the moment of the l -measurement at these two altitudes.

3. Results

3.1. Spatial distribution of aerosol sources

We use the column aerosol volume concentration values as the monitoring set of data to find spatial distributions of aerosol sources. The value $P_{i,j}$ in Equation (1) is the conditional mean aerosol volume concentration.

See Figures 2 and 3 for results of conditional mean volume concentration value $P_{i,j}$ estimations according to Equation (1) for coarse-mode and fine-mode aerosols on the basis of monitoring data at the Belsk, Minsk, Kyiv, Moldova/Kishinev, and Sevastopol sites (see Figure 1 and Table 1). The size of grid cell is equal to 4° longitude by 2° latitude, which corresponds to a cell of about $270\text{ km} \times 220\text{ km}$ at the Minsk latitude. The area (see Figures 2–4) consists of 100 grid cells and is located approximately between 42° and 62° N in latitude and between 12° and 50° E in longitude.

Aerosol particles are usually removed from the atmosphere by precipitation and due to the sedimentation under the influence of gravity. The lifetime of aerosol particles in the atmosphere depends on their size (r). The lifetime of an aerosol particle in the atmosphere, estimated on the basis of Stokes' law as the time of subsidence from 1 km altitude to the Earth's surface, is equal to 36 h for $10\text{ }\mu\text{m}$ -size particles. For aerosol particles of smaller size, the sedimentation time grows proportionally to r^{-2} . However, the real lifetime of small particles is limited by the removal of precipitation (Dubovik et al. 2008) and is approximately equal to 7 days. Our calculation shows that the time interval for the transport of air masses from the border of the reconstruction area to its central part on average is equal to 80 h. Therefore, the size of reconstruction area shown (Figures 2–4) is close to the optimal one, because the lifetime of particles is comparable to the air mass travel time through the reconstruction area.

The distribution of coarse-mode and fine-mode aerosol conditional mean volume concentrations by data from the Belsk site over the period 2004–2011 is shown in Figure 2. It is seen that the sources which produce the episodes of increased coarse-mode aerosol concentration in Belsk are located in the south of Ukraine, North Caucasus, Romania, and North Balkans. The same areas plus additional regions around Kyiv, Orel, and Moscow cities, and the area to the east of Moscow city are responsible for increased atmospheric pollution by fine-mode aerosol fraction at the Belsk site.

The distribution of aerosol sources retrieved during measurements at the Minsk site by data sets acquired from 2004 to 2011 is shown in Figure 2, and it reveals the increased coarse-mode aerosol volume concentrations that arrived from the territories located eastward of the Azov Sea and in regions near the cities of Donetsk and Rostov-on-Don. The increased fine-mode aerosol volume concentrations in Minsk were produced by sources located to the southeast of the Minsk site in the Krasnodar region, in the south of Romania, and in the Serbia region.

In accordance with the Kyiv site data set, the aerosol sources responsible for coarse-mode aerosol volume concentration increasing (Figure 2) are located to the

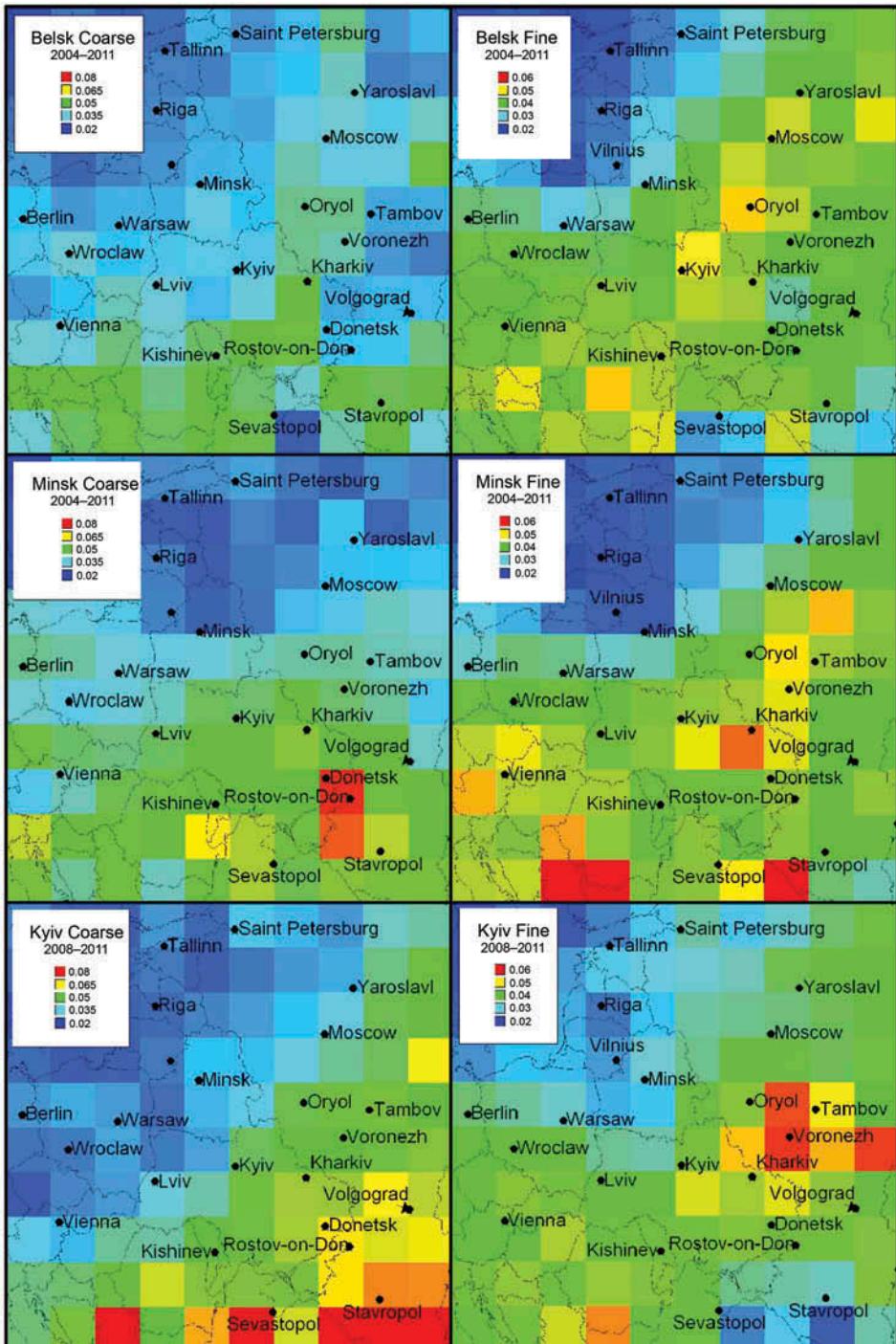


Figure 2. Spatial distribution of the conditional mean aerosol volume concentration in $\mu\text{m}^3 \mu\text{m}^{-2}$ for coarse-mode and fine-mode aerosol fractions from data of the Belsk site and the Minsk site over the period 2004–2011, and of the Kyiv site over the period 2008–2011.

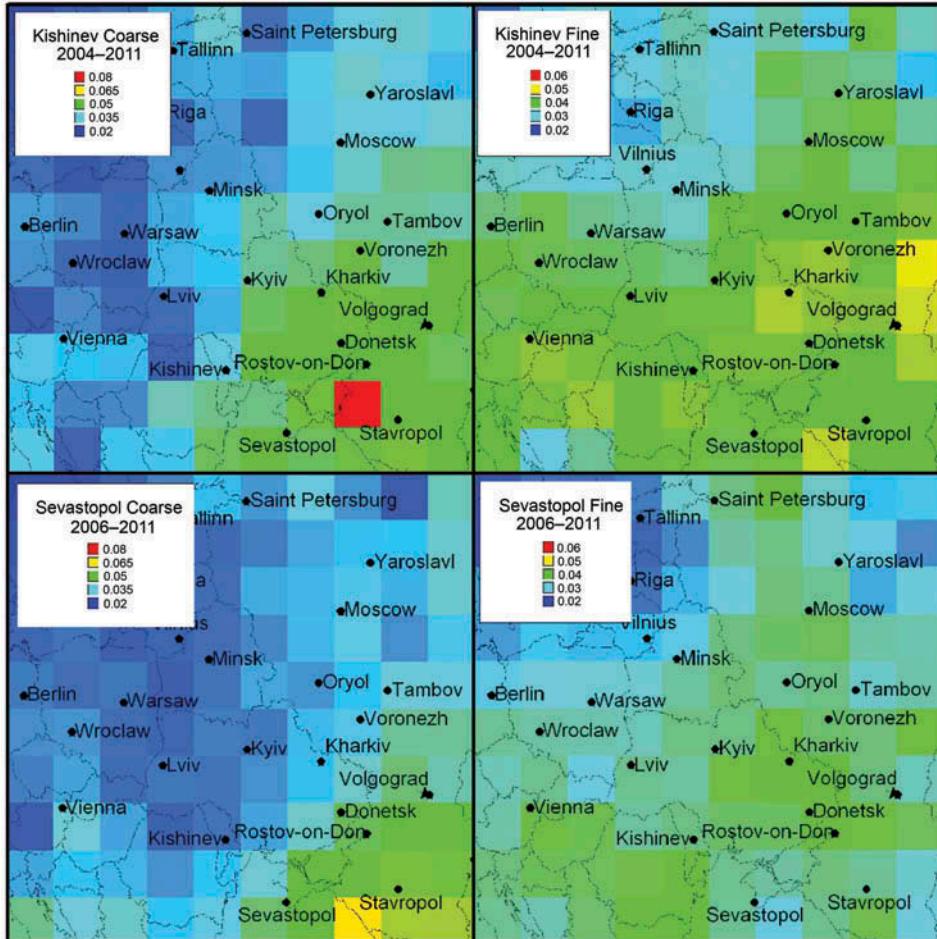


Figure 3. Spatial distribution of the conditional mean aerosol volume concentration in $\mu\text{m}^3 \mu\text{m}^{-2}$ for coarse-mode and fine-mode aerosol fractions from data of the Moldova/Kishinev site over the period 2004–2011, and of the Sevastopol site over the period 2006–2011.

southeast of the Kyiv site. Similar to the Minsk site concentration behaviour (see Figure 2), the largest concentrations appeared at the Kyiv site when the air mass arrived at the measuring site after transportation over the territory of the Krasnodar region. Much smaller but significant coarse-mode volume concentrations took place at the Kyiv site with the winds from the Stavropol region, Serbia, and the Black Sea. Regions responsible for the fine-mode volume concentrations that increased in the Kyiv site are located mainly around Voronezh and Kharkov cities, and to the south of Stavropol city.

Regions responsible for the episodes of increased coarse-mode aerosol concentrations at the Moldova/Kishinev site (see Figure 3) are located in the southeast of Ukraine and in North Caucasus. The area between the Azov Sea and Stavropol city is the most intense aerosol source. The air masses with high levels of fine-mode aerosol concentrations arrived at the Moldova/Kishinev site from the regions of Kharkiv, Voronezh, Volgograd, and Saratov cities, and also from the Crimean Peninsula and Azov Sea.

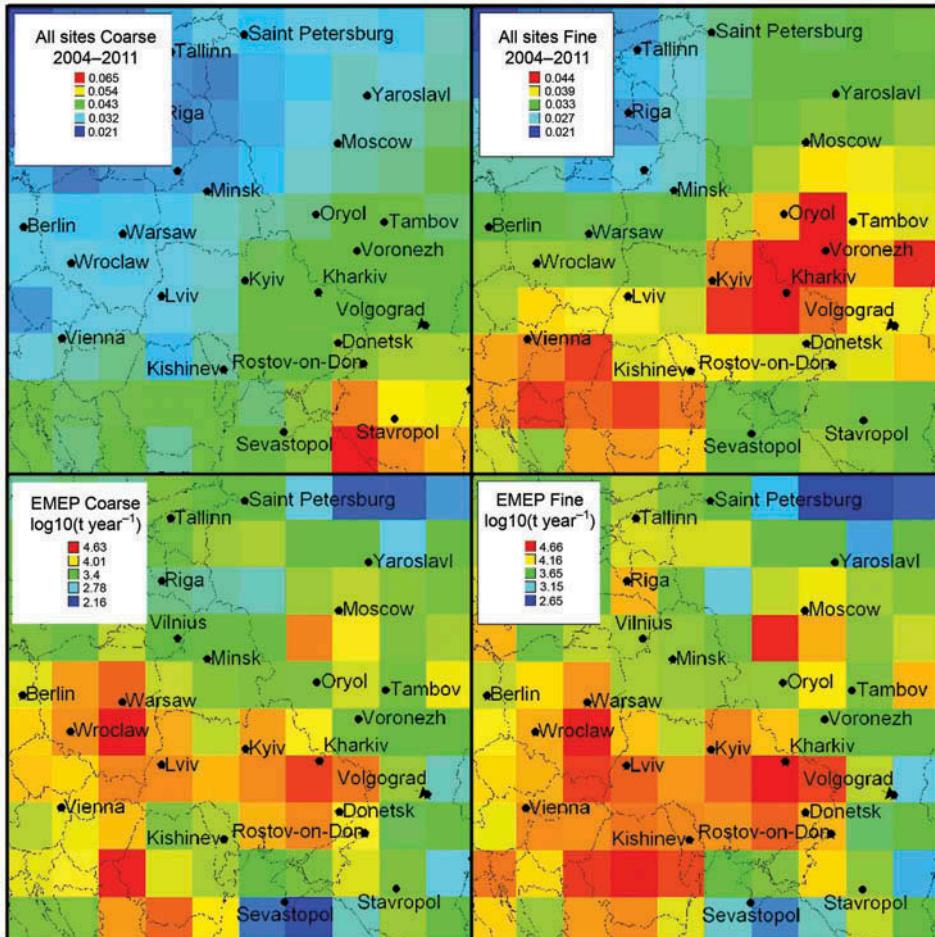


Figure 4. The conditional mean aerosol volume concentration ($\mu\text{m}^3 \mu\text{m}^{-2}$) of coarse-mode and fine-mode particle distribution showing the region sources that produced the aerosol contamination over the Belsk, Minsk, Kyiv, Moldova/Kishinev, and Sevastopol AERONET sites (top figures ‘All sites’), and spatial distribution of the aerosol coarse-mode and fine-mode fraction sources in 2008 according EMEP data.

Main sources that produced the increased coarse-mode aerosol concentrations in the Sevastopol site (Figure 3) are again situated in the Krasnodar region and North Caucasus. The increased fine-mode aerosol concentrations are mainly due to the sources located in the east of Ukraine and in the south of Romania.

3.2. Comparison with the EMEP data

It is well known that the main part of aerosol contamination has a natural origin (Dubovik et al. 2008; Kondratyev et al. 2006). However, quantitative data on spatial distribution of the power of aerosol sources of natural origin are still not well known. Therefore, it is interesting to compare the results of the averaged data from all sites shown in Figure 4 with the distribution of anthropogenic sources for coarse-mode and fine-mode particles

taken from the EMEP data (EMEP 2013). For comparison, we used gridded ($0.5^\circ \times 0.5^\circ$) PM2.5 and PM coarse emission data related to the year 2008 (updated in 2012) as used in EMEP models. The EMEP data (see Figure 4) are adjusted to the grid of 4° longitude by 2° latitude. The anthropogenic source powers vary in values $(0.14\text{--}42.66) \times 10^3 \text{ t year}^{-1}$ in the case of the coarse-mode fraction and $(0.45\text{--}45.71) \times 10^3 \text{ t year}^{-1}$ for the fine-mode fraction.

From the data presented, the territories responsible for increased coarse-mode aerosol volume concentrations do not exactly correspond with the coarse-mode particle source distribution from the EMEP data (Figure 4). Much better accordance can be found between the eastern groups of source areas (see Figure 4), responsible for increased fine-mode aerosol volume concentration release in the southeast of Ukraine from the EMEP data. The sources of fine-mode aerosol fraction in Romania, Hungary, and Balkan countries by EMEP data (see Figure 4) partly have correspondence to conditional mean volume concentrations above the same territories (see Figure 4, top row). The significant discrepancy between calculated sources and EMEP data is seen in the western part of the map (see Figure 4). The sources in Poland are not revealed in our calculations, but they are presented according to the EMEP data.

4. Discussion and conclusions

The regions responsible for cases of increased fine-mode and coarse-mode aerosol concentrations at five monitoring sites of eastern Europe were identified by the CWT method using the data of column aerosol concentrations retrieved from measurements by sun photometers. For each part of the area around the monitoring site, we calculated the volume mean aerosol concentration, which is the value of averaged concentration over the receptor (the monitoring site) using the condition when an air mass passes over the particular element of territory before measurement. The areas of high conditional mean aerosol volume concentration are sometimes identified as potential sources. In these cases, the source spatial distribution calculated by the data from different monitoring sites must be the same or similar.

Our analysis shows that the similarity of the aerosol sources and geographical distribution is responsible for increased aerosol concentrations in the East-European monitoring sites (see Figures 2 and 3). The distribution difference can be explained by the low accuracy of the back-trajectory statistics method, measurement errors, and by variations in meteorological conditions at the monitoring sites. A number of processes impact on the transfer of the contaminated air mass from the aerosol source to the receptor: turbulent diffusion, dry and moist sedimentation, and secondary aerosol formation. The aerosol volume concentration measured at the monitoring site varies due to these processes. Spearman's correlation coefficient between the different spatial source distributions is equal on average to 0.69 for the aerosol source distributions reconstructed by the data at the Belsk, Minsk, Kyiv, Moldova/Kishinev, and Sevastopol sites (Table 2). The generalized field of conditional mean aerosol volume concentrations created on the basis of data from the five monitoring sites is shown in Figure 4. The main territories responsible for the increased concentrations of coarse-mode particles are located in North Caucasus (Figure 4). This result can be explained by intensive soil erosion, which produced the dust sources in this region (Larionov, Skidmore, and Kiryukhina 1997). The main source areas responsible for episodes of increased concentrations of fine-mode aerosol at all monitoring sites are located in the regions around Kharkiv, Orel, Voronezh, Saratov cities, and also in Romania, Hungary, and Croatia.

Table 2. Spearman correlation coefficients of the aerosol source spatial distributions (shown in Figures 2, 3, and 4) retrieved from the data at the East-European monitoring sites.

Monitoring site	Coarse mode	Fine mode
Minsk – Belsk	0.702	0.716
Minsk – Moldova/Kishinev	0.649	0.723
Minsk – Sevastopol	0.578	0.624
Minsk – Kyiv	0.732	0.750
Belsk – Moldova/Kishinev	0.588	0.609
Belsk – Sevastopol	0.506	0.626
Belsk – Kyiv	0.649	0.708
Moldova/Kishinev – Sevastopol	0.790	0.736
Moldova/Kishinev – Kyiv	0.878	0.745
Sevastopol – Kyiv	0.818	0.669
Average	0.689	0.691

Our results show that the cases of maximum coarse mode of aerosol appeared when wind blows from south to east in spite of prevailing westerlies. The fine-mode particle pattern exhibits similar behaviour. We determined two big areas of fine-mode aerosol sources in east and northeast of Ukraine (areas around Kharkov and Donetsk) and south of the European part of Russia (areas around Voronezh and Orel cities). We also found that the northeastern areas (see the fragment of map shown in Figures 2 and 3), on average, do not produce the coarse-mode aerosol concentration increase.

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