Advances in Astronomy and Space Physics, **3**, 102-108 (2013)

The spatial variability of $PM_{2.5}$ over Europe using satellite POLDER-3/PARASOL data

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The paper presents the results of the monthly mean $PM_{2.5}$ analysis in the period from 2005 to 2013 over the Europe based on the connection between daily fine particle concentrations ($PM_{2.5}$) by surface in-situ measurements in AIRBASE network and column aerosol optical thickness (AOT) derived from POLDER-3/PARASOL satellite sensor. The regression function between $PM_{2.5}$ and AOT was derived from measurements done over Europe in the period from April to October 2007. Considering 749 match-up data points over 20 fine particle monitoring sites, we found that the POLDER-3/PARASOL derived AOT at 865 nm is correlated with collocated $PM_{2.5}$ measurements with a correlation coefficient 0.62 (RMS = 3.26). According to the obtained linear regression $PM_{2.5} = 73.4 \times AOT_{865} + 9.6$, a significant offset caused an introduction of the threshold of 0.01 in monthly mean AOT for assessment of $PM_{2.5}$ based on satellite data. Therefore, only $PM_{2.5}$ values larger than 10.3 $\mu g/m^3$ can be obtained using this method. According to results the monthly mean $PM_{2.5}$ in the period from 2005 to 2013 over the Europe is usually characterised by values less than 12 $\mu g/m^3$ (classified as "good" by Air Quality Categories, AQC), but values ranging from 12 to 18 $\mu g/m^3$ (classified as "moderate") are found in the densely populated and industrial areas, such as the Netherlands, Belgium, the Ruhr and Danube area, Northern Italy, Poland, Romania and Eastern Ukraine. Additionally, the maximum values of $PM_{2.5}$ over Eastern Europe are observed during forest, peat and agricultural wildfires in May 2006 (15–21 $\mu g/m^3$), April 2009 (14–18 $\mu g/m^3$) and August 2010 (35–55 $\mu g/m^3$, classified as "unhealthy for sensitive groups"). An extended set of aerosol parameters including particle size distribution, complex refractive index, as well as parameters characterising aerosol particle shape and vertical distribution will be analysed in the future work.

Key words: aerosols, $PM_{2.5}$, atmosphere, AOT, pollution

INTRODUCTION

Atmospheric aerosols are conventionally defined as particles suspended in air having radius in the range from 0.001 μm to 100 μm [7]. These particles have a significant negative effects on human health [23], including lung cancer, asthmatic symptom, pulmonary inflammation and cardiopulmonary mortality. The solution of many practical problems in the field of environmental and human health protection is connected with needs for high quality information about pollution level and the physicochemical properties of atmospheric aerosols.

The particulate matter (PM) mass, also known as air pollution, is a generally accepted means to quantify the amount of aerosols in the atmosphere and is used as a standard to evaluate air quality by United States Environmental Protection Agency[1] (EPA). Fine fraction of particulate matter ($PM_{2.5}$, $\mu g/m^3$) is the integrated mass of aerosols with diameter up to 2.5 μm that is generally resulted from combustion, including motor vehicles, power plants, forest wildfires, and agricultural burning. The EPA has established that the 24-hour averaged $PM_{2.5}$ concentration must be less than 35.4 $\mu g/m^3$ for healthy conditions concerning the whole population. Most of monitoring stations are located close to major urban regions leaving large areas without operational observations. Furthermore, it is widely recognised that it is problematic to measure the absolute level of $PM_{2.5}$ on a routine basis. In routine measurement processes, heating of the air sample is necessary, which partially increase volatilisation of semi-volatile aerosol components. This in turn leads to systematic measurement errors depending on measurement technique and aerosol composition. The limited spatial representativeness and different systematic errors of national or regional air quality networks make it impossible to achieve an air quality overview across Europe by surface in-situ measurements only.

In addition to surface in-situ measurements of $PM_{2.5}$, the monitoring of aerosols and trace gases from space has been widely used [16, 20, 26]. Primary aerosol quantity derived from spaceborne remote sensors operated in the visual and IR wave-

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[1] `http://www.epa.gov`

length bands is the aerosol optical thickness (AOT). The relationship between column AOT derived from satellite and $PM_{2.5}$ surface in-situ measurements has already been explored over the United States [1, 27], Italy [8] and some populated and industrial regions in Asia [18] using retrievals from the MODIS radiometer (MODerate Imaging Spectroradiometer), and over France [15] based on data from the POLDER-2 (Polarization and Directionality of Earth's Reflectances) satellite instrument.

Currently, the comparison studies between new satellite radiometer POLDER-3 and $PM_{2.5}$ surface in-situ measurements over Europe have not been conducted yet. In the present paper we use the relation obtained for 2007, being the year when $PM_{2.5}$ data were available online. We present results on the spatial distributions of monthly mean $PM_{2.5}$ values estimations based on POLDER-3 data over Europe. Last section gives an overview of special events and $PM_{2.5}$ characteristics in the period from 2005 to 2013 over investigated area.

SURFACE IN-SITU AIRBASE DATA

In this paper, the $PM_{2.5}$ data from the AIRBASE database[2] were used for determine the relationship between $PM_{2.5}$ values and AOT retrieved from POLDER-3. AIRBASE is a public database system operated by the European Environmental Agency (EEA). It provides air quality monitoring data and associated information submitted by more than 30 participating countries within Europe [9]. Measurement sites are classified according to the type of surroundings (urban, suburban, or rural) and dominant local source (traffic, industrial, or background). The reported precision of the measurements is satisfactory (random errors typically less than 15% for daily averages), and the absolute systematic uncertainty in these data is estimated to be 19%. The gravimetric methods based on the determination of the $PM_{2.5}$ mass fraction of particulate matter collected on a filter under ambient conditions was used to give typically 24-hour averages, while automatic instrumental methods TEOM (Tapered Element Oscillating Microbalance) and Beta Absorption provided hourly data [21]. In the Beta Absorption and TEOM methods, the sampled air is heated before collecting the aerosols on a filter to avoid humidification of the filter. This procedure is known to lead to loss of semi-volatile aerosol components. The extent to which volatilisation occurs depends on time and space dependent chemical composition of the aerosols, and the measurement technique. To correct for systematic errors, correction factors are generally applied to the measured data, but these differ by 30% or more between countries [6]. Consequently, systematic differences between territories of measurements may be present in the data due to use of different methods. However, the data from AIRBASE were used in the form that submitted by the countries. In the comparison of spatial distribution of AOT and $PM_{2.5}$, 17 rural and 3 suburban background stations have been analysed. The stations were selected in Austria, Belgium, Cyprus, Czech Republic, Finland, France, Germany, Italy, Latvia, Netherlands, Slovenia, Spain, Sweden, Switzerland and the United Kingdom to present different underlying surfaces (need for satellite retrieval) and aerosol loading. According to standard data from stations, the 24-hour averaged $PM_{2.5}$ values were used in analysis.

SATELLITE POLDER-3/PARASOL DATA

A unique set of observations on the global aerosol distribution has been acquired by the POLDER-3 instrument that was installed aboard French microsatellite PARASOL in December 2004 and collected data in the period from March 2005 to September 2013. The instrument carried out multispectral (443, 490, 565, 670, 763, 765, 865, 910 and 1020 nm), multi-directional (as many as 16 directions within the scope of 100° approximately along the ground trace) measurements of intensity and linear polarization degree of the back-scattered solar radiation [25]. The current standard aerosol inversion strategy detailed in [10] is based on the look-up tables approach, where the reflected radiances are simulated for 10 aerosol models with log-normal size distributions of particles with effective radius from 0.075 to 0.225 μm and a complex refractive index of $1.47–0.01i$. This inversion can be used to characterize the fine mode aerosols with particle size $< 0.3\,\mu$m [2, 5, 11]. According to the cloud-screening algorithm [4] the cloud-free pixels were processed only. The surface contribution to the polarized reflectance was based on a priori values (as a function of observation geometry and surface type) derived from statistical analysis of POLDER-1 data [22]. The aerosol parameters were adjusted to give the best agreement between the measured and simulated multidirectional polarized radiances at 490, 670 and 865 nm wavelengths.

The spectroradiometer POLDER-3 data were processed and described at the project web-page[3]. The spatial resolution of POLDER-3 instrument was 16 km × 18 km in the AOT. The aerosol characteristics accuracy has been estimated over ocean [12, 13] and land [5, 11], where correlation with AERONET (AErosol RObotic NETwork) [14] data is equal to 0.77–0.95 depending on location of measurement and aerosol type. For example, the correlation coefficients for the POLDER-3 and AERONET AOT retrieval comparisons are equal: 0.78 for Moscow site,

[2] http://acm.eionet.europa.eu/databases/airbase/

[3] http://www.icare.univ-lille1.fr/parasol

0.76 – Minsk, 0.86 – Belsk, 0.81 – Moldova, 0.93 – Kyiv and 0.63 for Sevastopol sites [2].

COMPARISON BETWEEN AOT AND $PM_{2.5}$

The satellite overpass time for the investigated territory was 10:00–11:30 UT depending on the orbit and geometry of measurements. The POLDER-3 derived AOT values at 865 nm were selected from each pixel (16 km × 18 km) covered the $PM_{2.5}$ surface in-situ stations. It should be noted that the amount of data increased for the sites at lower latitudes and larger number of sunshine days. For latitudes above 50° N, data were not available in winter, because the solar elevation is too low at these latitudes to allow AOT retrieval. The underlying surface in the investigated territory varies highly in spring and autumn due to variable vegetative cover. Moreover, the surface impact on polarization is relatively greater when the amount of aerosol in the atmosphere is small.

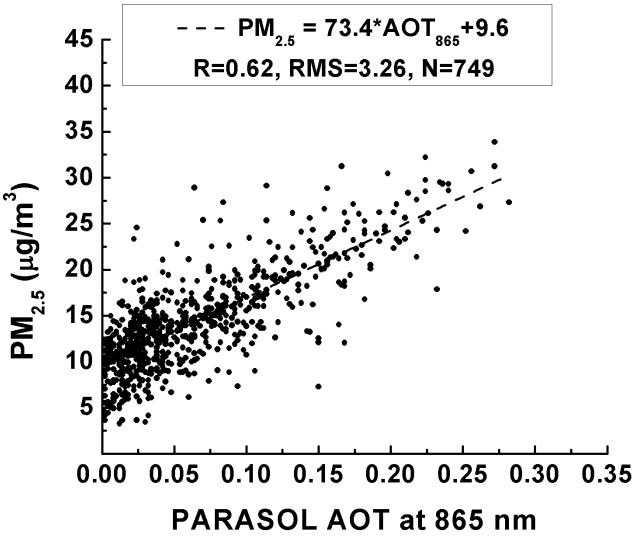

Fig. 1: Regression line between POLDER-3/PARASOL derived AOT at 865 nm and $PM_{2.5}$ measured at the surface over 20 stations in Europe from April to October 2007.

For deriving the regression function $PM_{2.5}$ versus AOT the 24-hour averaged $PM_{2.5}$ values were selected from 20 stations in Europe with more than 75% of valid measurements in the period from April to October 2007. Than we excluded AOT zero values obtained by POLDER-3 (lack of phase angles, glint or impossibility of retrievals) from analysis as well. Taking into account the above requirements the comparison was performed between POLDER-3 derived AOT and $PM_{2.5}$ values and presented in Fig. 1. The overall match-up database corresponds to 749 comparison points. A significant offset 9.6 $\mu g/m^3$ indicates that the satellite has a limited capacity for monitoring small amounts of aerosols. The slope of the regression contributes to 73.4 $\mu g/m^3$ per AOT unit (RMS = 3.26). The correlation coefficient is estimated as 0.62. The deviation could be explained by the rough spatial resolution of the POLDER-3 instrument comparing to the local station. Besides, the relationship between $PM_{2.5}$ and AOT strongly depends on vertical profile and aerosols type with similar optical properties that will be evaluated in the future study.

The comparison of relation between POLDER-3 AOT and $PM_{2.5}$ with those previously established using MODIS is complicated by the influence of several factors, like using PM_{10} instead of $PM_{2.5}$ [8, 18], different wavelengths and spatial resolutions for the satellite AOT [27], different time periods, and different sensitivity of the sensors to fine particles [5]. For example, the local study over Alabama [27] demonstrates that the slope is equal to 71 $\mu g/m^3$ per AOT unit using $PM_{2.5}$ based on MODIS data that is in a good agreement with the results presented here.

Since the $PM_{2.5}$ values lie in the range from 3.2 to 33.8 $\mu g/m^3$, it can be argued that no specific events occurred in the days of comparison. $PM_{2.5}$ values larger than 35.4 $\mu g/m^3$ are not presented, which characterise this match-up data by two first categories according to the EPA Air Quality Categories (AQC) [27], i. e. below 12 $\mu g/m^3$ (classified as "good") and between 12.1 $\mu g/m^3$ and 35.4 $\mu g/m^3$ (classified as "moderate"). Besides, the AOT at 870 nm over Europe during 2003–2011 years was characterized by values ranging from 0.002 to 0.2, except the period of July–August 2010 with strong forest and peat wildfires when the AOT typical values ranged from 0.3 to 0.5 [2].

SPATIAL DISTRIBUTIONS OF MONTHLY MEAN $PM_{2.5}$

Based on analysis presented in previous section, it is possible to estimate the spatial distributions of monthly mean $PM_{2.5}$ using POLDER-3/PARASOL AOT data during 2005–2013 over Europe. Nevertheless, a significant offset 9.6 $\mu g/m^3$ indicates that atmosphere with small amount of aerosols cannot be represented on the maps. Thus, in order to decrease the uncertainties in determining $PM_{2.5}$ the threshold of 0.01 was applied for monthly mean AOT values. This threshold was chosen due to increasing uncertainties in determining AOT when the amount of aerosol in the atmosphere is small. According to linear regression (Fig. 1) only $PM_{2.5}$ values larger than 10.3 $\mu g/m^3$ can be obtained using this method.

The monthly mean $PM_{2.5}$ values for August month through 2005–2013 except 2010 are presented in Fig. 2. The territory is usually characterised by $PM_{2.5}$ values less 12 $\mu g/m^3$. In general, there are some changes in distribution of aerosol pollution in the same month from year to year due to regional emissions, variable transboundary transfers and dif-

ferent quantity of sunshine days for averaging. In addition, areas with moderate $PM_{2.5}$ values (12–18 $\mu g/m^3$) were found in the densely populated and industrial regions of Europe. It should be noted that analysis of aerosol pollution over these areas was based on POLDER-3 data retrieved during the period from April 2005 to September 2013.

According to [24] the MODIS instrument observed emissions from agricultural fires in the Baltic countries, Belarus, Ukraine and Russia in April and May 2006. The $PM_{2.5}$ shows the high values ranging in 15–21 $\mu g/m^3$ in May 2006 over Belarus, Northern Ukraine and west of the European part of Russia seen in Fig. 3a, which was caused by biomass burning particles appeared during this event. Moreover, values 18–21 $\mu g/m^3$ were observed over Finland which can be explained by transportation of these particles from wildfires that caused the most severe air pollution episode over the recorded period at research stations in Svalbard [19]. Furthermore, the aerosol transboundary transportation from forest and brushwood fires in the European part of Russia in April 2009 produced increased $PM_{2.5}$ values of 14–18 $\mu g/m^3$ over Ukraine, Romania, Moldova and Eastern Belarus (Fig. 3b). As one can seen from Fig. 3a and Fig. 3b the $PM_{2.5}$ values in the range of 17–19 $\mu g/m^3$ were observed over Belgium, Netherlands, and Germany Ruhr area, that equally characterizes these industrial area in the spring of 2006 and 2009.

The maximum $PM_{2.5}$ values were revealed in August 2010 (Fig. 3c) that corresponds to wildfires in the centre of the European part of Russia [17]. Monthly mean values are ranged from 35 to 55 $\mu g/m^3$ over Moscow, Nizhniy Novgorod, Ryazan, Tula and Vladimir regions, and according to the AQC criteria this period can be classified as "unhealthy for sensitive groups". Moreover, the biomass burning aerosols transboundary transportation to Ukraine [3], Belarus, Lithuania, Latvia and Estonia was observed with $PM_{2.5}$ values from 20 to 35 $\mu g/m^3$ (Fig. 3c). For comparison to the presented events the monthly average $PM_{2.5}$ values for April 2013 are given in Fig. 3d, which are probably characterised by presence of anthropogenic aerosols over all Europe, particularly over East Germany (17–19 $\mu g/m^3$), Northern Italy (15–18 $\mu g/m^3$), Central and Northern France (13–16 $\mu g/m^3$), Central Ukraine and Moldova (12–15 $\mu g/m^3$). The short time events (hours, days) were not presented in this study due to monthly averaging data.

CONCLUSIONS

The AOT values retrieved from POLDER-3 data during 2005–2013 over Europe have been used to characterize aerosol pollution near the surface. POLDER-3 AOT retrieval algorithm over land is based on the measurement of the linear polarization of the light backscattered to space. It should be emphasised that standard algorithm is particularly sensitive to fine mode aerosols that polarize the signal. The relationship between column aerosol optical thickness derived from satellite POLDER-3/PARASOL and observed $PM_{2.5}$ by surface in-situ measurements is not straightforward and depends on the vertical distribution of the particles and their optical and microphysical properties. The uncertainties of both types of measurements should be taken into consideration as well. In the case of the satellite inversion algorithm the major influence of the surface and the spatial resolution produce those uncertainties. However, the POLDER-3/PARASOL derived AOT at 865 nm is fairly well correlated with collocated daily $PM_{2.5}$ measurements ($PM_{2.5} = 73.4 \times AOT_{865} + 9.6$). The correlation coefficient 0.62 and slope of the regression 73.4 $\mu g/m^3$ per AOT unit (RMS = 3.26) are derived based on AIRBASE data from 20 stations in Europe considered in the present study. Consequently, a significant offset caused an introduction of the threshold of 0.01 in monthly mean POLDER-3/PARASOL AOT for assessment of $PM_{2.5}$ based on satellite data.

With the aim to provide possible explanations for the general agreement and differences between aerosol properties retrieved from satellites and those observed at surface in-situ stations, in future work we will analyse an extended set of aerosol parameters including particle size distribution, complex refractive index, as well as parameters characterizing aerosol particle shape and vertical distribution. It is planned to use GEOS-Chem model for retrieving the distribution of aerosol mass and AOT with a transport time step of 15 min to calculate $PM_{2.5}$ with better precision.

The spatial distributions of monthly mean $PM_{2.5}$ using POLDER-3/PARASOL AOT data have been firstly estimated over Europe in 2005–2013. The Europe territory is usually characterised by $PM_{2.5}$ up to 12 $\mu g/m^3$, which is lower than in industrial regions of East Asia and India. The POLDER-3/PARASOL measurements show the major aerosol source regions in the Netherlands, Belgium, Germany, Northern Italy, Eastern Poland and Great Britain, Romania, Hungary, Moldova, Eastern Ukraine and Belarus, as well as individual large cities and industrial valleys (Moscow, London, Rhone, and Danube). The $PM_{2.5}$ values are ranged from 12 to 18 $\mu g/m^3$ over these industrial regions in the period from April 2005 to September 2013. In addition, the maximum values of $PM_{2.5}$ (15–21 $\mu g/m^3$) over Eastern Europe have been observed during forest, peat and agricultural wildfires in May 2006 over Belarus, Finland and Northern Ukraine, in April 2009 – 14–18 $\mu g/m^3$ over European part of Russia, Ukraine, Romania, Moldova and Eastern Belarus, and in August 2010 – 35–55 $\mu g/m^3$

over Moscow, Nizhniy Novgorod, Ryazan, Tula and Vladimir region in Russia. Aerosol amount during August 2010 is classified as "unhealthy for sensitive groups" according to AQC criteria.

ACKNOWLEDGEMENT

This study was supported by Award No. UKG2-2969-KV-09 of the U.S. Civilian Research & Development Foundation (CRDF) and by the projects M/115-2012, and F41/106-2012 of Derzhinformnauka, and PICS 2013-2015 of CNRS and NASU, and the project 11BF051-01-12 of the Taras Shevchenko National University of Kyiv. The author thanks the ICARE Data and Services Center team for providing access to the POLDER-3 data and for general assistance and development support. The author acknowledges all network operators that submitted their data to the AIRBASE database. The author is grateful to G. Milinevsky, V. Danylevsky, V. Bovchaliuk, P. Goloub, O. Dubovik, F. Ducos, M. Sosonkin and anonymous referee for detailed comments and useful suggestions.

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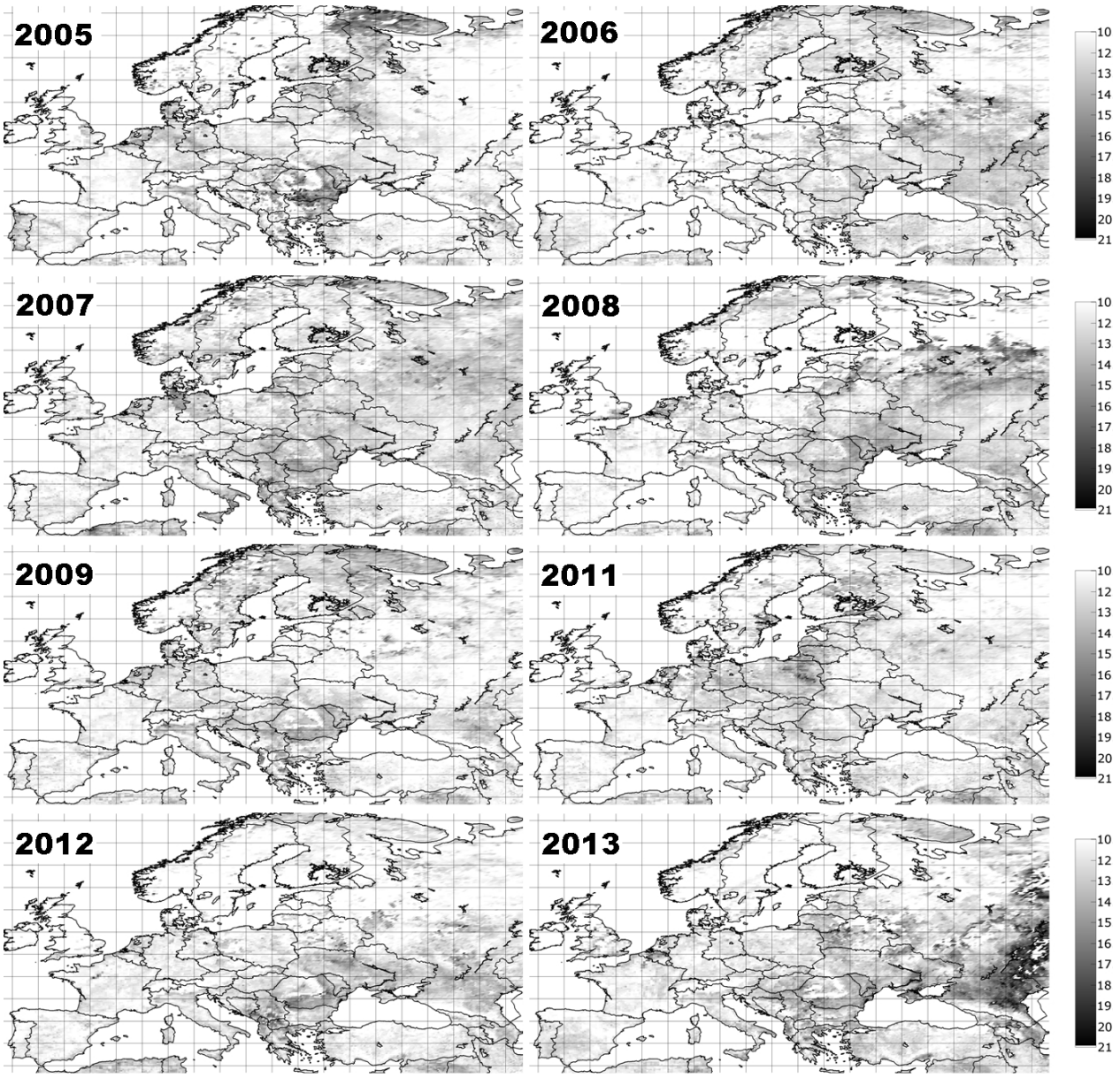

Fig. 2: Spatial distributions of monthly mean $PM_{2.5}$ for August month through 2005–2013 except 2010. White: missing data or ocean.

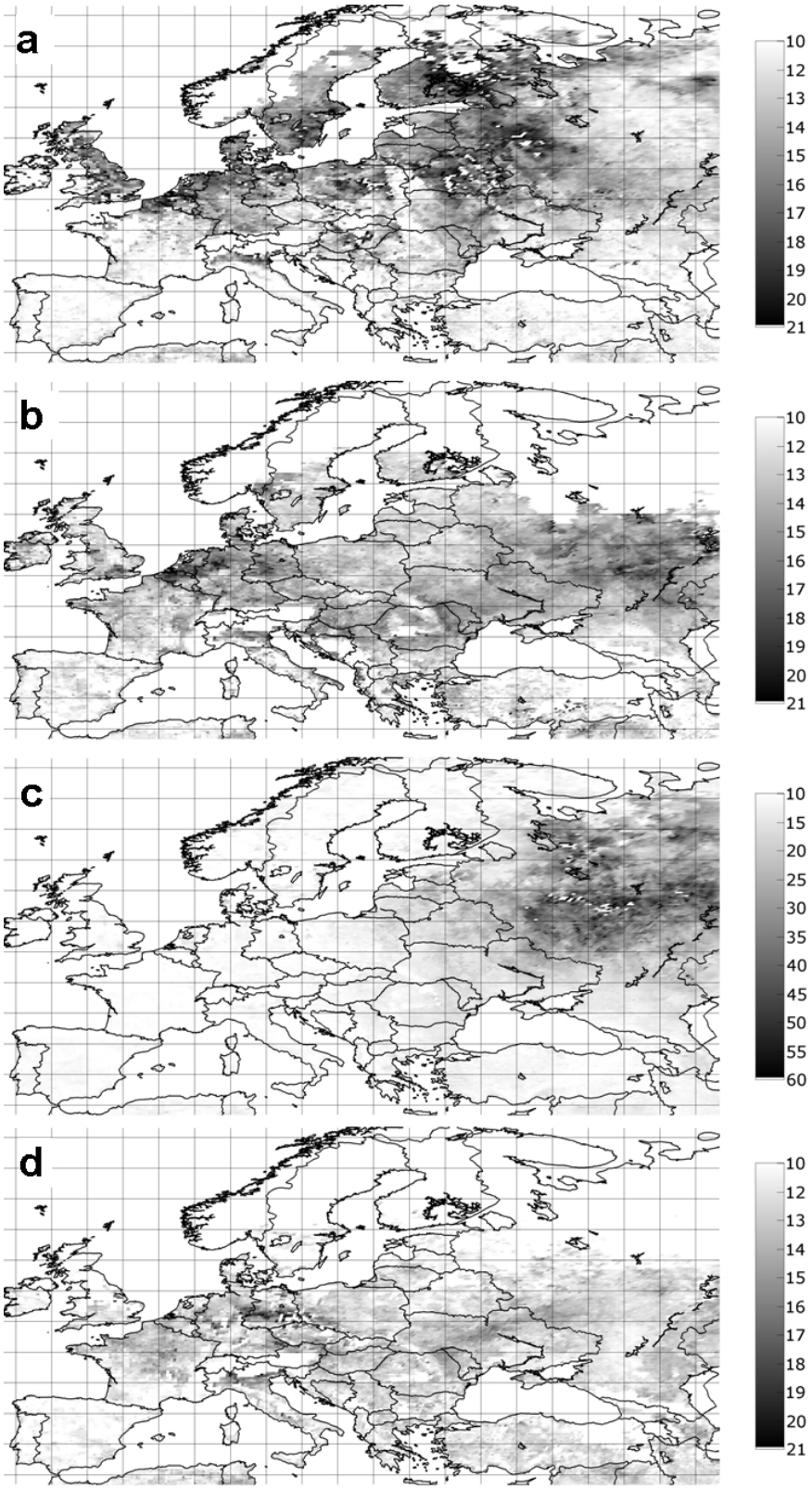

Fig. 3: Spatial distributions of monthly mean $PM_{2.5}$ for: a) May 2006; b) April 2009; c) August 2010; d) April 2013. White: missing data or ocean. Note different scale for August 2010.