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Variability of aerosol properties over Eastern Europe observed from ground and satellites in the period from 2003 to 2011

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Abstract

The paper presents the study of aerosol variability in the period from 2003 to 2011 over Eastern Europe region with latitude ranging from 40° N to 60° N and longitude from 20° E to 50° E. The analysis was based on the POLDER/PARASOL and POLDER-2/ADEOS satellites and AERONET ground-based sunphotometer observations. The aerosol optical thickness (AOT) of the studied area is characterized by the values (referenced to 870 nm wavelength) ranging from 0.05 to 0.2 except the period of July–August 2010 with strong forest and peat wildfires when the AOT typical values range from 0.3 to 0.5. The analysis of seasonal dynamics of aerosol loading has revealed two AOT high value peaks. The first peak observed in April–May is the result of solitary transportation of Sahara dust in the atmosphere over Eastern Europe, infrequent agricultural fires, transportation of sea salt aerosols by southern winds to Ukraine and Moldova from the Black and Azov Seas. The second peak in August–September is associated with forest and peat wildfires, considerable transportation of Sahara dust and presence of soil dust aerosols due to harvesting activity. The maximum values of AOT are observed in May 2006 (0.1–0.15), April 2009 (0.07–0.15) and August 2010 (0.2–0.5). Furthermore, the study has identified a distinct pattern of anthropogenic aerosols over the industrial areas, especially in the central Ukraine, eastern Belarus, as well as Moscow, Nizhny Novgorod and Stavropol regions in Russia.

The comparison of the fine mode AOT (particle radius $< 0.3\,\mu$m) derived by standard algorithm POLDER/PARASOL from reflected polarized radiances with those recomputed from AERONET inversions was performed over a number of AERONET sites: over Kyiv and Sevastopol sites for the period of 2008–2009 and over Moscow, Minsk, Belsk, and Moldova sites for the period of 2005–2009. The correlation coefficients are 0.78 for Moscow, 0.76 – Minsk, 0.86 – Belsk, 0.93 – Kyiv, 0.81 – Moldova and 0.63 for Sevastopol sites. The deviations are explained by the spatial inhomogeneity of the surface polarization that has stronger effect on aerosol retrieval for clear atmospheric conditions with low aerosol loading when surface impact on satellite observations is

more pronounced. In addition, the preliminary analysis of the detailed aerosol properties derived by new generation PARASOL algorithm was accomplished. The AOT and single scattering albedo retrieved by the algorithm over Kyiv were compared with the closest AERONET retrievals within two hour of satellite overpass time and the stable atmospheric conditions.

1 Introduction

Aerosols influence not only the radiative balance of terrestrial atmosphere and therefore the climate on the regional and global scales, but also have considerable impact on air quality. Aerosol particles are diverse in size, shape and chemical composition (Seinfeld and Pandis, 2006). They are produced both by natural sources, such as forest fires, sea spray, desert sandstorms and volcanic eruptions, and by human activities, such as industry, fossil fuel and biomass burning, deforestation and others (Chin et al., 2009). Depending on physical and chemical properties aerosols can cause an increase or decrease in the planet's temperature.

Up-to-date knowledge of aerosol formation and conversion physics is based on research of optical phenomena and cloud formation processes in the atmosphere. The amount of aerosol in the atmosphere is usually quantified by mass concentration or by optical measure referred to as aerosol optical thickness (AOT). Aerosol mass in the atmosphere comprises about 10^{-9} of all air mass (the fraction of aerosol particles is smaller than 10^{-6} in dusty air), which is by three-four orders of magnitude smaller than the mass fraction of water vapor (Seinfeld and Pandis, 2006). In spite of this fact, the role of aerosols in atmospheric processes is very important, especially in the formation of clouds and interaction with water vapor, since in the atmosphere without condensation nuclei a cloud can form only at high altitude due to condensation on the ions. The mass of water vapor per unit volume of air is by several orders of magnitude greater than the mass of aerosol particles, and this has a considerable effect on the variability of aerosol optical characteristics (Chin et al., 2009). Almost at all wavelengths the coefficients of aerosol extinction, scattering and absorption are approximately of the same order as for all the atmospheric gases, but aerosol optical characteristics are much more variable both in time and space. Aerosols affect Earth's energy budget by scattering and absorbing radiation and by modifying the amounts as well as microphysical and radiative properties of cloud cover (IPCC, 2007).

The aerosol parameters and their impact on the climate are studied by using the ground-based measurements, for example, AERONET, SKYNET, AEROSIBNET, GLOBE networks, and satellite observations of optical radiation scattered by the Earth atmosphere and surface. These methods are used to determine spatial and temporal variation of aerosol physical and chemical properties in order to solve the aerosol-climate interaction problems.

Both ground-based and satellite data are obtained by passive and active remote sensing techniques that have been developed to study aerosol optical properties. The passive remote sensing techniques are based on the measurements of solar radiance scattered by surface and atmosphere aerosols whereas active techniques use lidar sounding devices. The satellite techniques enable us to derive the aerosol distribution over the globe by scanning the atmosphere and surface along and across satellite ground tracks at specific overpass time. There are several data sets of global aerosol properties over land and oceans available from various satellite sensors, for example MODIS, MERIS, MISR, AVHRR, POLDER, TOMS and OMI (King et al., 1999; Mishchenko et al., 2007; Kaufman et al., 2002).

The ground-based technique allows the continuous accumulation of data over long periods of time (years and decades) but only at specific observational locations. From data collected by both ground-based and satellite techniques, aerosol properties are estimated as the parameters of a model solving the inverse problem (King et al., 1999; Dubovik et al., 2002, 2011; Kokhanovsky and de Leeuw, 2009). The extreme complexity and variability of Earth's atmosphere-surface system are the main source of retrieval errors. The best results on aerosol properties and dynamics are obtained by the combined analysis of space-borne and ground-based remote sensing data. In particular, the retrievals of aerosol parameters from satellite remote measurements are more problematic than those from ground-based observations, so extensive validation of satellite retrievals is required. The combination of satellite and ground-based observations makes possible the retrieval of the vertical distribution of aerosols as well. In the paper we analyze and compare the results of the aerosol distribution and variability over Eastern Europe region obtained by ground-based sunphotometers and satellite data collected in 2003–2011.

2 Aerosol dynamics over Eastern Europe

The field of interest in this work is Eastern Europe region with latitudes from 40° N to 60° N and longitudes from 20° E to 50° E where a lot of natural and anthropogenic aerosol sources are located. This region is characterized by numerous agricultural, forest and peat fires, as well as soil erosion in steppe area. Besides, there are many existing and potential aerosol pollution sources: transport, intensive agriculture, heavy industry, metallurgy, as well as exploitation of open mines.

The forest and peat wildfires in Moscow Region in September 2002 caused increase of AOT and transportation of aerosols by air masses to Moldova through Belarus and Ukraine (Aculinin et al., 2004; Eck et al., 1999). Moreover, ten cases of very high AOT value during 2001–2005 (mainly in August) were observed with UV-Raman lidar over Thessaloniki, Greece (Amiridis et al., 2009). These events were identified as agricultural burning across Russia in the latitudinal belt between 45° N and 55° N, as well as in Eastern Europe (Baltic countries, Belarus and Ukraine). It was found that the optical characteristics of smoke aerosol were variable and this behaviour was mainly attributed to the fact that the burned regions were located at variable distances from the lidar of Thessaloniki which affected the optical characteristics of the advected smoke aerosols due to different ageing processes. The particle size is likely to increase with the age of the advected air mass (Amiridis et al., 2009).

It was noted that the number of agricultural burn events decreased in 2002 and 2003 compared to 2001 in Eastern Europe and western Russia (Korontzi et al., 2006). The agricultural fire activity showed two peaks, the first occurred in April and May and the second – in August, which is associated with burning croplands and peats. The seasonal and interannual trends in agricultural fire activity are consistent with the regional agricultural practices. Stohl et al. (2007) observed emissions from agricultural fires in the Baltic countries, Belarus, Ukraine and Russia at the end of April and beginning of May 2006 that caused the most severe air pollution episode over the recorded period at the Zeppelin research station in Spitsbergen. Amiridis et al. (2010) showed by CALIPSO data that aerosol top heights of the vertically homogenous smoke layers ranged from 1.6 to 5.9 km over the locations of agricultural fires in south-west Russia and Eastern Europe in 2006–2008 (during July and August). The contribution of wildfires and agricultural fires in this region to the European fine fraction aerosol optical thickness are estimated to be 20–35 % in April and 28 % in August through the period from 2002 to 2007 (Barnaba et al., 2011).

Anomalously high surface temperatures (35–41 °C) and low relative humidity (9–25 %) from mid-June to mid-August 2010 were ideal conditions for the wildfires to thrive. The analysis showed that the region around Moscow in the centre of western Russia was most severely impacted by wildfire emissions (Witte et al., 2011; Konovalov et al., 2011). Chubarova et al. (2012) presented the results showing extremely high daily average AOT at wavelength 500 nm (AOT500) on 6–8 August that reached the absolute maximum of 6.4 in Moscow and 5.9 in Zvenigorod on 7 August.

An incidental soil dust event was observed in Europe in 23–25 March 2007. It originated from south Ukraine (Kherson, Mykolaiv, Zaporizhia regions) known for its erodible lands (Birmili et al., 2008; Bessagnet et al., 2008). The strong surface wind (15–30 m s^{-1}) blowing in dry weather conditions picked up the soil dust and water soluble potassium (K^+) concentrated in humus from the top layers of “chernozem” (very fertile black soil) to the altitude of 1.8 km by a boundary layer jet (Bessagnet et al., 2008). This dust plume with dominant particle size 1–5 µm was transported through Ukraine, Moldova, Slovakia, Czech Republic, Austria, Poland and Germany to Belgium, North France and the UK.

The Sahara is the major source of mineral dust that often spreads across the Mediterranean to Europe. According to Israelevich et al. (2012) whose research is based on data from MODIS for the period from 2001 to 2010 the Sahara dust is transported to Eastern Europe region mostly in autumn (August–September) and sometimes in spring (April).

3 Description of ground-based and satellite data

3.1 Ground-based data

In this section we analyze the data obtained by ground-based network AERONET (AErosol RObotic NETwork). The network provides continuous time series data with a high temporal resolution obtained for a site of measurements during many years (Holben et al., 1998). The main instrument used by AERONET to measure aerosol properties measurements in spectral range 350–1650 nm is the automatic sun tracking and sky scanning sunphotometer CIMEL CE-318 (CIMEL Electronique, France; http://www.cimel.fr/?instrument=photometre-multi-bandes-soleilciel). The aerosol data obtained from AERONET include AOT, the Ångström exponent, precipitable water vapour, aerosol volume size distribution, real and imaginary part of refractive index, single scattering albedo (SSA), absorption and extinction optical thickness of aerosols, asymmetry factor and some others (Holben et al., 1998). The technical specification of the sunphotometer, measurement sequences and technique for retrieving aerosol optical characteristics, data processing, and the accuracy of measurements are described in (Dubovik and King, 2000; Eck et al., 1999; Dubovik et al., 2000) and in Technical and Quality Assurance Documents available on AERONET official website (http://aeronet.gsfc.nasa.gov).

The analysis of diurnal cycle of AOT500 and Ångström exponent computed in the wavelength range from 440 nm to 870 nm using 2000–2005 measurements over Chisinau, Moldova is presented by (Aculinin, 2007). The results of one year sun-photometer operation, monthly statistics and aerosol particle characteristics over Kyiv AERONET site in 2008–2009 are presented by Danylevsky et al. (2011). The combination of multi-wavelength lidar data with aerosol column integrated products retrieved from AERONET is a powerful approach to determine vertical distribution of fine and coarse mode AOT (Chaikovsky et al., 2004). According to Chaikovsky et al. (2007) and Pietruczuk and Podgórski (2009) whose research is based on data from October 2004 to December 2006, the most powerful aerosol source region was located in the south-east, south and south-west directions relatively to the monitoring stations Minsk (Belarus) and Belsk (Poland). About 60 % of aerosols in Minsk and about 50 % of aerosols in Belsk are shown to have transboundary origin. Different microphysical, optical and radiative properties of aerosol were analyzed using 10-yr measurements (2001–2010) at Moscow site by Chubarova et al. (2011a). The high correlation of the AOT values, the Ångström exponent and the effective radius between the observations at Moscow and Zvenigorod sites confirm that natural processes are the dominating factor influencing aerosol properties even over Moscow megacity area (Chubarova et al., 2011b).

In our paper we have analyzed the cloud-screened and quality-assured data (Level 2.0) obtained in the region under study. In order to characterize decadal period of aerosol pollution the dependence of AOT and Ångström exponent on time is given in Fig. 1. The same period of time 2001–2011 has been selected for AERONET sites: Moscow (55.7000° N, 37.5100° E, Russia), Minsk (53.9200° N, 27.6010° E, Belarus), Belsk (51.8367° N, 20.7917° E, Poland), Kyiv (50.3636° N, 30.4966° E, Ukraine), Moldova (47.0001° N, 28.8156° E, Moldova), Sevastopol (44.6158° N, 33.5173° E, Ukraine). It should be noted that the chosen sites started operating at different times: Moldova – in September 1999, Moscow – in August 2001, Belsk – in April 2002, Minsk – in July 2002, Sevastopol – in May 2006, and Kyiv – in April 2008. Moreover, the measurements were distributed non-uniformly through the year, for example, in winter the number of observational days was significantly smaller than in other seasons.

The two peaks of aerosol amount seen in Fig. 1 show the increase in AOT which was observed almost every year in each site. The first peak (in April–May) is partially explained by agricultural fires that were started by farmers who burn their fields before a new growing season. This practice is illegal in the European Union but is still widely used in Eastern Europe for crop rotation and controlling insects. It is quite common that agricultural fires get out of control and devastate the nearby forests. Correspondingly, the values of Ångström exponent are higher in April–May which could be explained by the increase in fine fraction aerosols such as smoke particles. Moreover, Sahara dust in the atmosphere layer over Eastern Europe can increase AOT value in spring. The contribution of pollen is neglected since the observed minimum radius of blossom dust, for example, during the flowering period of *Myosotis* (Forget-me-not) in March–April is 6–10 µm. According to Jaeglé et al. (2011) the lifetime of sea salt aerosols with particle size up to 4 µm is 12–25 h. The sea salt aerosols are transported to Ukraine and Moldova from the Black and Azov Seas by the southern wind in spring. Therefore, monthly mean volume size distribution (Fig. 2) shows the presence of 0.5–4 µm particles over Sevastopol and Moldova sites in April–May.

The second peak in August–September is associated with the fires in croplands, as well as forest and peat wildfires in addition to the considerable transportation of Sahara dust. Furthermore, harvesting causes increase in soil dust amount in the atmosphere and the particles with size 1–5 µm can be transferred on hundreds kilometers in strong surface wind weather conditions. The Ångström exponent values depend on the events that prevail and therefore on aerosol characteristics.

The monthly mean volume size distributions for April, May, August and September averaged through 2000–2011 are shown in Figs. 2 and 3 where they display certain common features. The analysis has been accomplished for each month of investigation period and separate site.

The fine fraction particles peak was found for Moscow region in April 2002 and 2005, May 2005 and 2006, August 2002 and 2010 (particularly due to forest and peat wildfires), September 2002, 2003 and 2008. The coarse fraction particles peak was observed in April 2009, May 2006, August 2007 and 2010, September 2002, 2003 and 2008. The volume fraction of coarse particles is larger than that of fine particles in May 2007 and May 2011.

The fine fraction particles peak was observed predominantly in Minsk in April 2006 and 2010, in May 2005, 2006 (particularly due to forest, peat, grass and shrubbery wildfires) and 2007, in August 2002, 2006 and 2010, in September 2002 and 2008. The coarse fraction particles peak was registered in April 2008 and 2009, May 2003, 2006 and 2007 (due to soil dust event in southern Ukraine described above), August 2002 and 2010, September 2002, 2003 and 2010.

The maximums of fine fraction particles were detected in Belsk in April and May 2006, August 2002 and 2010, September 2002 and 2007, while coarse fraction particles prevailed in April 2002 and 2009, May 2002 and 2006, August 2002, 2008 and 2010, September 2002 and 2008.

The fine fraction particles peak was observed in Kyiv in April 2009, 2010 and 2011, in August 2008 and 2010, in September 2008, while coarse fraction particles were registered mostly in April 2009 and 2011, in May 2008 and 2009, in August 2008 and 2010, in September 2008 and 2009.

The fine fraction particles prevailed mainly in Moldova in April 2003, in May 2006, in August 2001, 2002, 2005 and 2008, in September 2002 and 2006. The coarse fraction particles were observed predominantly in April 2000 and 2003, May 2003 and 2007 (due to soil dust event in southern Ukraine in 2007), August 2001 and 2007, September 2002 and 2006.

The maximums of fine fraction particles were detected in Sevastopol in April 2007 and 2009, in August 2008, in September 2007, while coarse fraction particles were observed in April 2008 and 2009, in May 2007 (due to soil dust event in southern Ukraine) and 2010, in August 2007, 2008 and 2010, and in September 2010.

Near-surface wind speed is known to cause the increase in amounts of several aerosols types, for example, sea salt, dust, primary organic particles, and burn particles (IPCC, 2007). In order to qualify the possible reasons of AOT change the data from IRI/LDEO Climate Data Library (http://iridl.ldeo.columbia.edu) were used to characterize 925 hPa wind fields over Eastern Europe for the period 1961–1990 on the base of NOAA NCEP CPC CAMS_OPI climatology (Kalnay et al., 1996). The southwest and west winds dominate in January, south and southwest – in February and March, south – in April. The southeast wind dominates over Ukraine and Belarus while over northwest Russia the southwest wind prevails in May, north and northwest – in June and July. The west wind blows in August–September in the northern part the region under study particularly in Belarus and northwest Russia while the east wind from the Caucasus meets with the west wind and produces a north and northeast wind over south Ukraine, the Black and Azov Seas. The west and southwest wind predominates in October–December.

3.2 Satellite data

The experience of satellite remote sensing has showed that only multi-spectral, multi-directional and polarized measurements provide the robust aerosol properties retrieval over land because radiance reflected from Earth's surface has small polarization while fine aerosol particles polarize light substantially, and thereby the polarized satellite measurements are adequate only for aerosol studies over land. The POLDER (POLarization and Directionality of the Earth Reflectance) instrument (Deschamps et al., 1994) meets these features because it carries out multi-spectral (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 ground trace) measurements of intensity and linear polarization degree of back-scattered solar radiation. The POLDER-1 instrument was launched on board ADEOS satellite in August 1996 with operation time till June 1997. The POLDER-2 instrument flew aboard ADEOS-2 from April to October 2003 and POLDER-3 was launched aboard French microsatellite PARASOL (part of the

A-Train satellite constellation) in December 2004 and has been collecting data since March 2005.

The spectral distribution allows us to estimate aerosol particle sizes and thus their scattering phase function, as well as the AOT. The polarization provides information on aerosol refractive index and shape (spherical or non-spherical), which improves the precision of the scattering phase function. The current standard aerosol inversion strategy detailed in Deuzé et al. (2001) 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, mean refractive index $m = 1.47 - 0.01i$ and Ångström exponent between 1.8 and 3. The aerosol parameters are adjusted to give the best agreement between the measured and simulated multidirectional polarized radiances at 670 nm and 865 nm wavelengths. The surface contribution to the polarized reflectance is based on priori values (as a function of observation geometry and surface type) derived from statistical analysis of POLDER data (Nadal and Bréon, 1999).

The characteristics of Earth's surface are taken into account in the form of empirical coefficients which are chosen for different classes of land surface according to the main International Geosphere-Biosphere Programme (IGBP) biotypes and the normalized difference vegetation index (NDVI) (Nadal and Bréon, 1999). Moreover, the polarized light reflected by the surface is wavelength independent.

The spectroradiometer POLDER data were processed and were described at http://www.icare.univ-lille1.fr/parasol/. Earth's surface reflection coefficients are obtained from maximum 16 directions for POLDER/PARASOL and from 14 directions for POLDER-1, -2 with resolution 6 × 6 km, while aerosol parameters are estimated with resolution 3 × 3 pixels, i.e. approximately 18 × 18 km. The spectral polarized radiances (Level 1 data) performed at 670 nm and 865 nm wavelengths are used to derive AOT over cloud-free regions (Level 2 data).

The monthly mean (Level 3 data) AOT values at 865 nm for April, May, August and September from POLDER-2 and POLDER/PARASOL through 2003–2011 period except 2004 are used to analyze aerosol loading in the atmosphere over Eastern Europe. The results are presented in Fig. 4. The territory is characterized by low AOT in comparison with industrial regions of East Asia and African dust events.

The high value of AOT in April–May and August–September is generally explained by agricultural fires, pollution of uncontrollable forest and peat wildfires, as well as transportation of Sahara dust over the Mediterranean to Eastern Europe in April and August–September. According to the National report on technological and natural safety (2011) by the Ministry of Ecology and Natural Resources of Ukraine the number of forest, peat and agricultural fires and burnt areas has decreased over the last 5 yr. In 2005, the total of 4223 fires took place with 26 km^2 burnt areas. The number of fires had been decreasing since 2005 and reduced to1780 events with 6 km^2 burnt area in 2011. The most extensive wildfire events were observed in May 2006 when aerosol amount in the atmosphere increased due to transportation of aerosols from the wildfires in Belarus and in western Russia in April 2009. The AOT values registered at 865 nm ranged from 0.1 to 0.15 in the first event and 0.07 to 0.15 in the second event. The aerosol transboundary transportation from western Russia and north Ukraine strong forest and peat fires in the mid-July–August of 2010 produced increased AOT values to 0.2–0.5 (Fig. 4). Sea salt aerosols are usually transported to Ukraine and Moldova from the Black and Azov Seas by south winds in April–May but the detection of 0.5–4 µm particles from POLDER/PARASOL is problematic.

According to our research the values of AOT are higher over the central Ukraine (Kyiv, Cherkasy, Kirovograd and Vinnytsia region) and Kryvii Rig, Mykolaiv, Zhitomyr, Rivne, Lviv and Chernigiv cities; over the whole territory of Moldova; over Minsk, Baranovichi, Gomel and Vitebsk cities in Belarus; over Moscow region, Tver, Voronezh, Volgograd, Stavropol, Ryazan, Nizhny Novgorod, Astrakhan and Krasnodar cities. However, the amount of aerosols has decreased since 2009 which is caused particularly by reduction of burnt areas. Moreover, a small decrease of anthropogenic aerosols in industrial regions can be observed from May 2009 to August 2011 in the result of global economic crisis when many factories and plants reduced their output.

4 Comparison of AERONET and POLDER data

4.1 Current standard aerosol inversion

The significant correlation (0.84) between satellite and ground-based measurements has been obtained over land surface (Bréon et al., 2011). However, some discrepancy in the aerosol radius cutoff leads to underestimation of the satellite retrieval results compared to AERONET data. In the regions where dust-loaded atmospheres are excluded, i.e. in the regions affected by biomass burning or pollution aerosols, the comparison with AERONET measurements shows better results with small bias (Bréon et al., 2011; Tanré et al., 2011). A specific comparison study (Fan et al., 2008; Su et al., 2010) of AERONET data over East Asia with POLDER/PARASOL observations demonstrates the POLDER instrument capability to determine the anthropogenic contribution ($r < 0.3\,\mu\text{m}$, where r – particle radius) to regional aerosol load. Additionally, Gu et al. (2011) have showed that the sensitive radius for polarized aerosol retrieval is 0.35 µm for all seasons in 2005–2009 for Beijing (China) and Kanpur (India).

The PARASOL satellite overpass time for the investigated territory is 10:00–11:30 UT depending on the orbit. The domains with 3 × 3 pixels of Level 2 (54 × 54 km) were selected so that the central pixel covered the site equipped by sunphotometer. Moreover, we chose the days when minimum 6 pixels from the domain were retrieved on condition that measurements were collected at least at 9 different angles. Since POLDER derived AOTs are sensitive to the fine aerosol mass concentration (Fan et al., 2008; Su et al., 2010) in our research we estimate the aerosols with $r < 0.3\,\mu\text{m}$. Consequently, the AOT values of fine mode aerosols have been computed by AERONET retrieval software for all the data from the stations. We selected the AOTs that were measured within ±30 min of the satellite overpass times and on condition that AOT variation in this lapse of time did not exceed 25 % of the daily mean. Furthermore, we excluded AOT zero values from our analysis as well. After selecting the required data we compared POLDER AOT derived from the central pixel and recomputed AERONET AOT which satisfied the above requirements. The results of the comparison for Kyiv and

Sevastopol sites over the period from 2008 to 2009 and for Moscow, Minsk, Belsk and Moldova sites over the period from 2005 to 2009 are presented in Fig. 5.

In general, there is a good agreement between the AERONET fine mode AOT and the POLDER AOT over all sites except Sevastopol. The correlation coefficients are 0.78 for Moscow, 0.76 for Minsk, 0.86 for Belsk, 0.93 for Kyiv, 0.81 for Moldova and 0.63 for Sevastopol sites. As seen from Fig. 5 the slopes are approximately 1 for Minsk (0.97), Kyiv (0.99) and Moldova (1.01), and equal 0.83 for Moscow, 1.13 for Belsk and 0.74 for Sevastopol. It should be noted that the number of data increased for the sites at smaller latitude and greater quantity of sunshine days, i.e. Moldova and Sevastopol. The high deviation in Sevastopol site may be explained by the spatial variation of surface polarization at the coastal land. The surface impact on polarization is relatively greater when the amount of aerosol in the atmosphere is small. The underlying surface in the investigated region varies highly in spring and autumn due to snow and vegetative cover. The deviation could also be explained by the same cutoff of fine mode particles ($r < 0.3\,\mu\text{m}$) while the presence of different aerosol types over each site can lead to changing this limit. For instance, AERONET station in Sevastopol is a coastal site where sea salt particles are present throughout the year and the fine aerosol mass concentration is typical for this territory (Figs. 2 and 3).

4.2 Potential of new generation algorithms for retrieving enhanced aerosol properties from space

Finally, we have analyzed a limited data set of aerosol properties derived over AERONET sites by the new generation of PARASOL aerosol retrieval algorithm. In contrast to most satellite retrieval approaches the new state-of-art algorithm (Dubovik et al., 2011) does not use look-up table. Instead, it implements all the radiative tranfer calculations on line and searches for the solution in continuous space of aerosol solutions. This algorithm is an attempt to enhance aerosol retrieval by emphasizing statistical optimization using the positive data redundancy (Dubovik, 2004; Dubovik et al., 2011) available from advanced multi-angular polarimetric satellite spectral observations such as those by POLDER/PARASOL. Based on this strategy, the algorithm aims at retrieving an extended set of aerosol parameters including particle size distribution, complex refractive index, as well as some parameters characterizing aerosol particle shape and vertical distribution. The high potential of this approach is confirmed by "blind test" intercomparison of satellite retrievals over dark surface by Kokhanovsky et al. (2010). The results of these tests showed that new PARASOL retrieval algorithm provides the most accurate and complete aerosol retrieval from synthetic data.

In addition, the approach allows for retrieving not only optical properties of aerosol but also underlying surface reflectance over land. In order to achieve a robust retrieval of both, the algorithm uses the so-called a "multi-pixel" retrieval regime where the inversion of satellite data is performed simultaneously for a large group of pixels. The approach allows for applying additional a priori constraints on temporal variability of surface reflectance and spatial variability of aerosol properties. The first applications of the algorithm to real PARASOL data by Dubovik et al. (2011) show a promising agreement of the retrieved values of POLDER AOT with AERONET data with correlation coefficient of ~ 0.9 for Banizoumbou (Niger) and ~ 0.87 for Mongu (Zambia).

In comparison with PARASOL standard algorithm the new algorithm provides more aerosol parameters and describes the surface reflectance with higher robustness. However, since the algorithm performs all radiation computation on line, the time required to implement the algorithm is significantly longer compared to the conventional look-up table algorithms. This limitation is expected to be addressed in the near future by parallel programming and implementing the retrievals at the network of computers that operate in parallel.

The new algorithm was applied for POLDER/PARASOL data obtained over Kyiv AERONET site for the period from 2008 to 2011. According to the numerical tests (Dubovik et al., 2011) multi-pixel retrieval was selected for data processing. The data were chosen in conditions when time difference between AERONET measurement and POLDER/PARASOL overpass did not exceed 120 min: 18 eligible days in 2008, 14 days in 2009, 7 days in 2010 and in 2011. It should be noted that the atmosphere usually changes in 1–2 h, therefore the days with stable AOT value and cloudless weather conditions according to AERONET measurements were compared. The time difference between satellite and ground-based data time (Δt) acquisition is smaller in spring and autumn and larger in summer while in winter the eligible measurements were not collected due to clouds and snow (Fig. 6a).

The comparison of AOT440, SSA440 (Fig. 6b) and AOT870, SSA870 (Fig. 6c) retrieved from POLDER/PARASOL observations over Kyiv with the corresponding values provided by AERONET in 2008–2011 are presented in Fig. 6. As can be seen from the illustrations, the variability of AOT and SSA retrieved from PARASOL is in good agreement with the dynamics of aerosol loading observed by AERONET. Such outliers can probably be explained by sky inhomogeneity in PARASOL observed pixels. The AOT440 reaches the maximum value 0.96 (see Fig. 6b), which is caused by smoke aerosols from the forest and peat fires in August 2010. It should be emphasized we have reported only preliminary results from those that can be provided by the new PARASOL algorithm. We expected to achieve retrievals of even higher quality once the algorithm is fully tuned and validated in operational PARASOL data processing.

5 Conclusions

The Eastern Europe region which has been of primary interest in this work is located on (LAT: from 40° N to 60° N, LONG: 20° E to 50° E) and is often considered to be a source of both natural and anthropogenic aerosols. We have analyzed aerosol distribution for the period of 2003–2011 using satellite POLDER/PARASOL and POLDER-2/ADEOS-2 measurements of aerosol characteristics and AERONET ground-based sunphotometer data. Cloud-screened and quality-assured AERONET data (Level 2.0) have been used to analyze the data obtained over Moscow, Minsk, Belsk, Kyiv, Moldova, and Sevastopol sites. The typical aerosol loading over these territories in the period of 2003–2011 is characterized by the AOT values (at 870 nm) ranging from 0.05 to 0.2.

The observed seasonal variability of aerosol amount is characterized by two high peaks in April–May and August–September. The first peak is explained by incidental transportation of Sahara dust in the atmospheric layer over Eastern Europe, infrequent agricultural fires, transportation of sea salt aerosols (with particle sizes 0.5–4 µm) to Ukraine and Moldova from the Black and Azov Seas by southern winds. Correspondingly, the values of Ångström exponent are higher in April–May which is explained by the increased amount of fine fraction particles.

The August–September peak is associated with forest and peat wildfires, and autumn season of transportation of Sahara dust. The strong surface wind can cause the transportation of the soil dust aerosol (1–5 µm) on hundreds of kilometers during harvesting activity. According to the analysis the region is characterized by numerous agricultural (August–September), grass, shrubbery wildfires (May 2006, Belarus), forest and peat fires (August 2010, Russia, north Ukraine). These seasonal and inter-annual trends in agricultural fire activity are consistent with regional agricultural practices.

The aerosols observed over industrial areas of Eastern Europe region are presumably of anthropogenic origin. According to the analysis of the AOT distribution, higher values are observed over the central Ukraine and Belarus, the whole territory of Moldova, over Moscow and Nizhny Novgorod regions. A small anthropogenic aerosols decrease over industrial regions can be observed from May 2009 to August 2011, which is probably caused by the global economical crisis when many factories and plants reduced the production.

The comparison of the AOTs obtained from POLDER/PARASOL observations standard algorithm with fine mode AOTs (particle radius $<0.3\,\mu$m) recomputed from AERONET inversions has shown that the correlation coefficients for satellite and ground-based data are 0.78 for Moscow, 0.76 for Minsk, 0.86 for Belsk, 0.93 for Kyiv, 0.81 for Moldova and 0.63 for Sevastopol. The slope is close to 1 for Minsk, Kyiv and Moldova, and 0.83 for Moscow, 1.13 for Belsk and 0.74 for Sevastopol. The deviation is explained by spatial variations of surface polarization used in the retrieval, since the surface impact on satellite observations is higher in the atmospheric conditions with small aerosol loading.

The results obtained by the algorithm developed recently by Dubovik et al. (2011) have been compared with the data from AERONET for Kyiv site. The satellite and ground-based data were chosen within two hours of satellite overpass time in stable atmospheric conditions for 46 clear days in 2008–2011 period. Since the new retrievals from POLDER/PARASOL have preliminary character, the presented analysis of the newly derived aerosol characteristics is somewhat limited and presents the results over Kyiv in the conditions of low loading of anthropogenic aerosols. The comparison of the derived aerosol properties with available observations by AERONET ground-based sunphotometer indicates rather promising consistency of PARASOL derived optical thickness and single scattering albedo with those obtained by AERONET.

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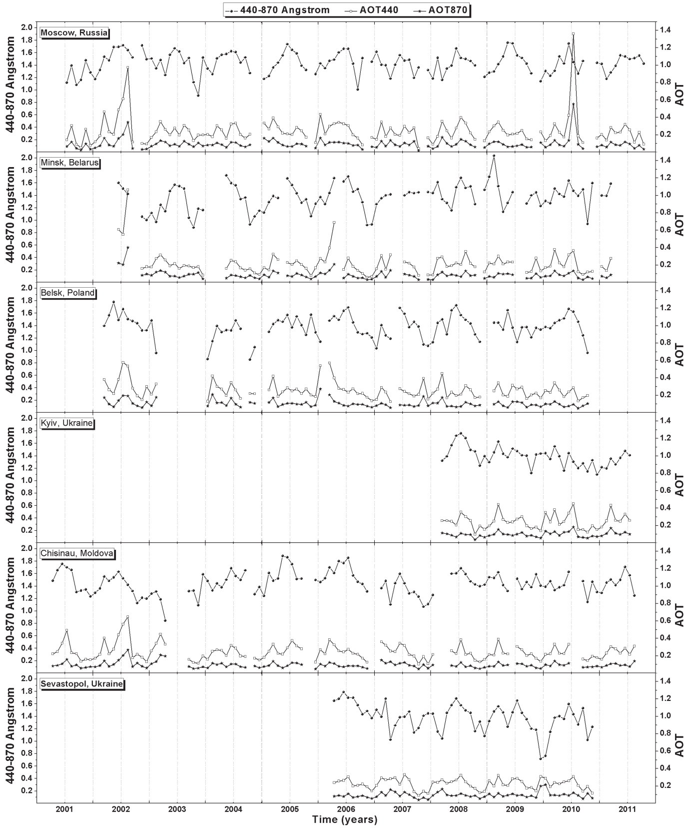

Fig. 1. Monthly mean AOT at 440, 870 nm and Ångström exponent computed from 440 to 870 nm for 2001–2011 from Moscow, Minsk, Belsk, Kyiv, Moldova and Sevastopol sunphotometers.

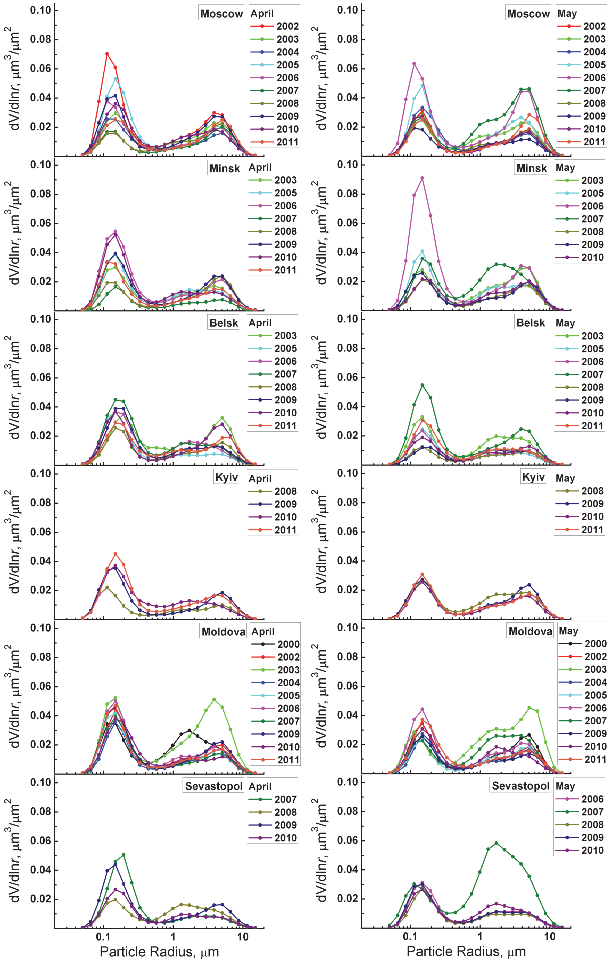

Fig. 2. Monthly mean volume aerosol particle size distribution in April and May averaged through 2000–2011 period.

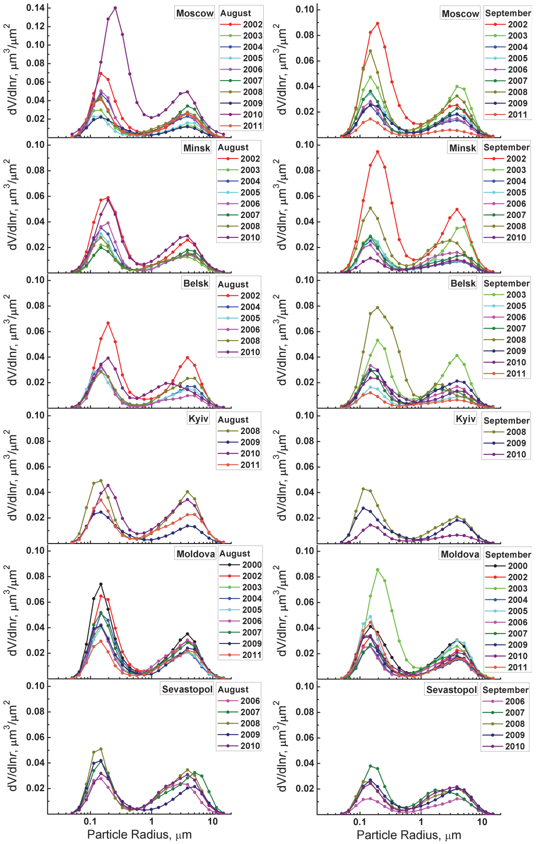

Fig. 3. Monthly mean volume aerosol particle size distribution in August and September averaged through 2000–2011 period. Note different axis scale of volume size distribution for August for Moscow site.

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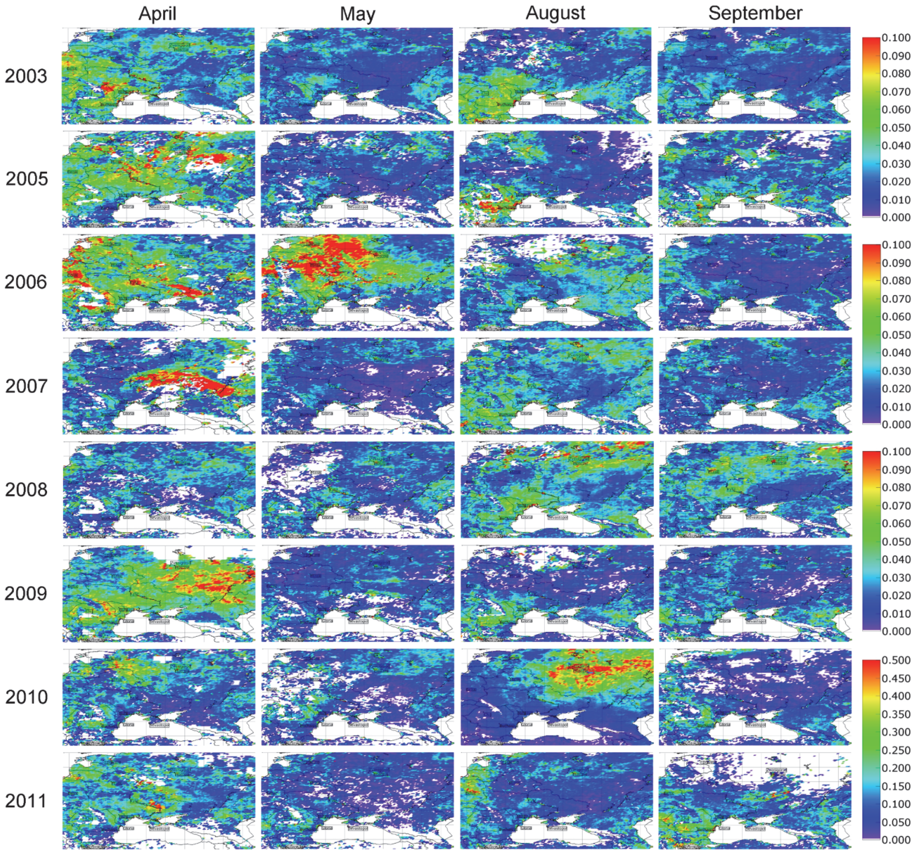

Fig. 4. Maps of monthly average AOT at 865 nm received from POLDER-2/ADEOS-2 and POLDER/PARASOL for April, May, August, September through 2003–2011. Note bottom AOT scale with maximum value 0.5 for August 2010 map.

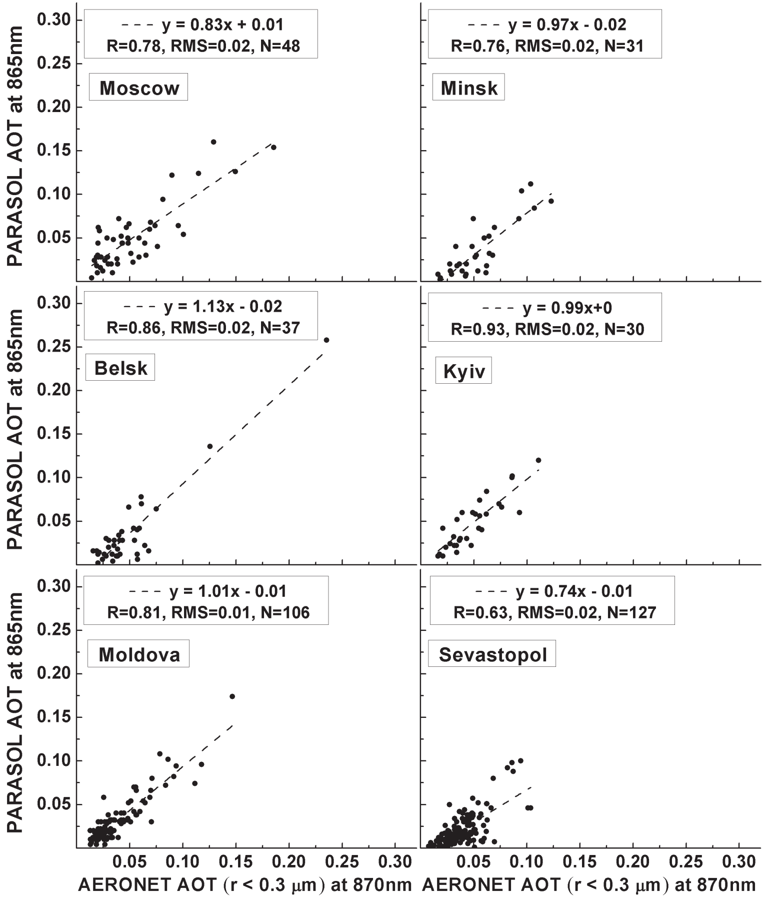

Fig. 5. Comparison of POLDER/PARASOL AOT865 and AERONET fine mode ($r < 0.3\,\mu m$) AOT870 for Moscow, Minsk, Belsk, Kyiv, Moldova and Sevastopol sites.

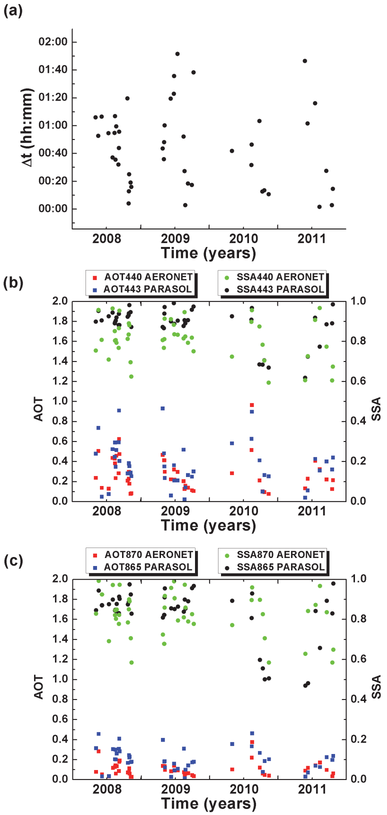

Fig. 6. Time difference between AERONET measurements and POLDER/PARASOL overpass time **(a)**. The comparison of AOT440, SSA440 **(b)** and AOT870, SSA870 **(c)** retrieved from POLDER/PARASOL in the period from 2008 to 2011 over Kyiv with the corresponding values provided by AERONET.