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ORIGINAL RESEARCH ARTICLE

Impact of wild forest fires in Eastern Europe on aerosol composition and particle optical properties☆

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Summary In this paper the authors discuss the changes of aerosol optical depth (AOD) in the region of eastern Europe and the Baltic Sea due to wild fire episodes which occurred in the area of Belarus and Ukraine in 2002. The authors discuss how the biomass burning aerosols were advected over the Baltic area and changed the composition of aerosol ensemble for a period of several summer weeks. The air pressure situation and slow wind speeds also facilitated the development of such conditions. As a consequence very high AOD levels were recorded, by an order of 3—4 higher versus normal conditions and they significantly increased the annual averages. On particular days of August 2002 the AOD values reached a level of over 0.7. On these days fine particles fully dominated the entire ensemble of aerosol particles. They were either sulfates or smoke particles. Such situation was unique over a period of many years and it had its serious consequences for the region and especially for the Baltic Sea.

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1. Introduction

Climate change results from both natural and human-induced modifications of the Earth's energy balance. These climate-related factors include variations in the amounts of greenhouse gases, aerosols, changes in land use, and the amount of energy Earth receives from the Sun (IPCC, 2007, 2013).

Atmospheric aerosols are responsible for radiative forcing on the surface, and they affect from local and regional weather patterns to global climate. Atmospheric aerosols are considered among the factors which play a significant role in the Earth's energy budget and thus affect global climate. However, as the IPCC reports show uncertainties are still significant regarding their radiative and climate effects (IPCC, 2007, 2013). Aerosol particles with their scattering and absorptive properties (so-called direct effect), are responsible for a number of environmental effects, i.e. they play a major role in the visibility problem, and they can also have a significant impact on UV radiation (Torres et al., 1998, 2007), and photochemistry in the boundary layer (Graber and Rudich, 2006; Kirchstetter et al., 2004).

Regional scale variations in radiative forcing have significant regional and global climatic implications, which cannot be addressed through the idea of global mean radiative forcing (Markowicz et al., 2011). Until now only a small number of studies has been dedicated to regional radiative forcing and response. It is still very difficult to describe a regional forcing and response in the observational record. Regional forcings may induce global climate responses, while global forcings can be connected with regional scale climate responses (Shindell et al., 2010; Tosca et al., 2013).

Aerosols' direct radiative effects are manifested in scattering and absorbing of radiation in both shortwave and longwave. The state of our knowledge of direct radiative forcing of aerosol particles is limited mainly due to difficulties of the proper quantification of global distributions and mixing states of aerosols. The state of mixing of aerosol particles affects particle optical properties, which thus are not well understood and are difficult to parameterize in climate models. Additionally, small-scale variability of such meteorological parameters as humidity and temperature, which also influences aerosol optical properties, is also difficult to represent in models (Byčenkiene et al., 2013; Petelski et al., 2014; Smirnov et al., 2011; Zielinski and Zielinski, 2002).

Wild fires have been modifying atmospheric composition for centuries. They have significant impact on the physical environment including modifications of land cover, land use, biodiversity or inducing climate changes and this impact is on both regional and global scales (Chubarova et al., 2012; Pio et al., 2008). Differences in aerosol composition on a regional scale as a result of advection of particulates from the regions of wild fires have been described before (Pio et al., 2008).

Additionally such disasters also have impact on human health and even on the socio-economic situations in the affected regions (Beringer et al., 2003). In order to describe and quantify the role of biomass burning as a source of atmospheric gases and aerosol particles to the atmosphere, information is required on the global magnitude of biomass burning (Chubarova et al., 2012; Zawadzka et al., 2013). Biomass burning aerosols include two important chemical components: black carbon and organic carbon. The first component primarily absorbs solar radiation, and the second scatters solar radiation (Cooke and Wilson, 1996; Haywood and Ramaswamy, 1998; Liousse et al., 1996). Takemura et al. (2002) made 3-D model simulations of radiative forcing of various aerosol species. In their model they divided all the main tropospheric aerosols into the following groups — carbonaceous (organic and black carbons), sulfate, soil dust, and sea salt aerosols. They compared their model simulations of total aerosol optical thickness, Ångström exponent, and single-scattering albedo for mixtures of four aerosol species with the observed data from both optical ground-based measurements and satellite remote sensing retrievals at a great number of stations. They reported the mean difference between the simulation and observations to be less than 30% for the optical depth and less than 0.05 for the single-scattering albedo in most regions (Takemura et al., 2002).

According to Bond et al. (2013) the estimate for the period 1750—2005, i.e. the industrial-era, direct radiative forcing of atmospheric black carbon is +0.71 W m^{-2} with 90% uncertainty bounds of (+0.08, +1.27) W m^{-2}. However, total direct forcing of black carbon from all known sources, with the pre-industrial background included, estimates to +0.88 (+0.17, +1.48) W m^{-2}. Bond et al. (2013) also report that: "the best estimate of industrial-era climate forcing of black carbon through all forcing mechanisms, including clouds and cryosphere forcing, is +1.1 W m^{-2} with 90% uncertainty bounds of +0.17 to +2.1 W m^{-2}".

The Baltic Sea is a unique basin of the World Ocean. It is a small and shallow sea located in the north-eastern part of Europe, and is surrounded by nine highly industrialized countries — Denmark, Estonia, Finland, Germany, Lithuania, Latvia, Poland, Russia and Sweden with some 85 million people living in the catchment area. Additionally, largely forest covered areas in Poland, Lithuania, Belarus and Ukraine are in the close vicinity of the Baltic.

According to Zdun et al. (2011) summer wind patterns for station in Gotland, located in the middle part of the Baltic Sea over a period between 1999 and 2003 show that around 50% come from easterly and southerly directions, thus crossing areas of Belarus and Ukraine. Zielinski (2004) reported that majority of winds in summer come to Sopot area, in the southern part of the Baltic Sea, from NE and SE directions, which is in accordance with Zdun's observations. Therefore, aerosol particles over the Baltic Sea, especially in summer, usually originate from a number of continental sources and it is difficult to observe one type of aerosols, e.g. marine over the Baltic. A number of aerosol studies have been conducted in the region, however, regular sunphotometric aerosol measurements have not been made in the Baltic area until 1999, when such studies had been originated within the NASA/AERONET program (http://aeronet.gsfc.nasa.gov/).

Year 2002 was unique in terms of air temperatures which were recorded in the region of central Europe and the Baltic area. Seven consecutive months were warm or very warm, and between February and September the multiannual average air temperature was higher than the normal temperature by over 1°C. May and August 2002 were extraordinarily warm. Such conditions were very conducive to wild fire outbreaks in the areas of Eastern Europe, which are widely covered by forests and meadows.

Years 2007 and 2008 were also very hot in the discussed region, however, warm periods were shorter than in 2002 and

lasted mostly 4 weeks. Then temperatures returned to the multiannual average for some time and warm periods returned. These years and 2010 were also fire abundant in the discussed region but the air mass trajectories were such that the smoke plume did not reach the area of the Baltic Sea. Thus these years have not been analyzed in this paper.

This paper presents information on the impact of wild fires in the Eastern Europe on aerosol optical properties in that region with a special emphasis on the Baltic Sea area. The authors discuss how the biomass burning aerosols were advected over the Baltic area and changed the composition of aerosol ensemble for a period of several weeks. Further the authors provide justification that those aerosols were fine particles and they increased the aerosol optical depth by an order of 3—4 versus normal conditions.

2. Description of the study area and research methodology

Location of the selected AERONET stations around the Baltic Sea is shown in Fig. 1. All three stations have used CIMEL sunphotometers with 7 wavelengths (http://aeronet.gsfc.nasa.gov/).

The authors decided to analyze the data from these three stations since all three of them operated in summer 2002 and are representative for this study. The AERONET stations selected for analyses in this paper include: Sopot station, on the Baltic Sea coast, operating between 1999 and 2002 and the Gotland station, located in the northern part of the island of Gotland, 50 m inshore. Owing to the location of the island in the central Baltic Sea this station is sometimes regarded as representative for the Baltic Sea conditions (Zdun et al., 2011). The data were collected at the station from 1999 to 2004. The third station analyzed is the Minsk station which has been operated since 2002. The station coordinates and the CIMEL wavelengths at each of the three stations are presented in Table 1.

For the analyses the authors have used only level 2.0 quality assured and with pre- and post-field calibration applied, cloud-screened data (http://aeronet.gsfc.nasa.gov/).

The type of aerosol particles can be deduced using two optical parameters: aerosol optical depth (AOD) and the Ångström parameter (Bokoye et al., 1997; Zielinski and Zielinski, 2002). Both of these parameters strongly depend on the sources of particle generation and the type of air masses (Smirnov et al., 2011). On a regional scale or in cases of such regional seas like the Baltic Sea, aerosols are often difficult to define since both marine and continental types of particles mix in the air, depending on wind direction. Such mixture comprises particles of different optical properties. The Ångström parameter is used to estimate particle size. Large values of this parameter indicate fine particles while small values indicate coarse aerosols (Zawadzka et al., 2013).

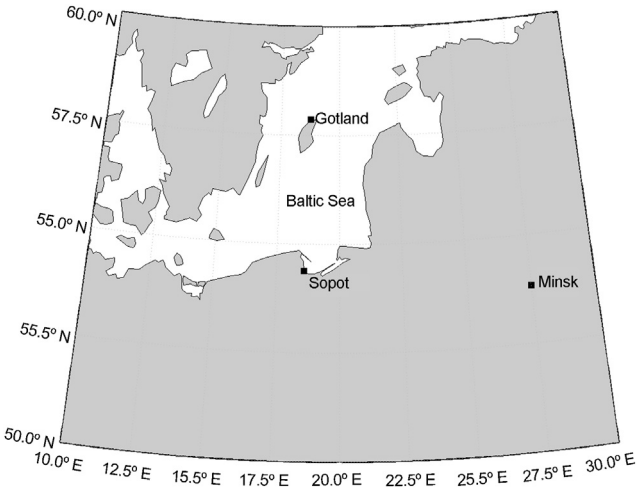

Figure 1 Selected AERONET stations located in the vicinity of the Baltic Sea.

The wavelength dependence of aerosol optical depth can be expressed as follows (Carlund et al., 2005; Eck et al., 1999; Smirnov et al., 1994):

$$\tau(\lambda) = \beta(\lambda)^{-\alpha}, \tag{1}$$

where α is the Ångström parameter, $\tau_\alpha(\lambda)$ is AOD at a wavelength λ, and β is Ångström coefficient of turbidity.

The spectral dependence of the AOD as well as the Ångström exponent are sensitive to the calibration coefficients. In this study we used the Ångström exponent α defined at two wavelengths as follows:

$$\alpha = -\frac{\ln(\tau_1/\tau_2)}{\ln(\lambda_1/\lambda_2)}. \tag{2}$$

The air mass back trajectories have been calculated at 500, 1500 and 3000 m using the NOAA-HYSPLIT model (Draxler and Rolph, 2010). In order to obtain additional information about transport of air pollution a long-range pollution transport model was applied. For this purpose the authors used the Navy Aerosol Analysis and Prediction System (NAAPS) (Christensen, 1997; Witek et al., 2007). The NAAPS simulations are presented in Fig. 11. Additionally, the AOD results have been supported by the analyses of satellite aerosol optical depth data from the Moderate Resolution Imaging Spectroradiometer (MODIS), mounted onboard the Aqua and Terra satellites.

3. Results and discussion

In summer 2002 vast areas in Belarus and Ukraine were subject to serious and long-lasting wild fires, which were a source for significant production of biomass burning

Table 1 Three selected AERONET stations; coordinates and CIMEL wavelengths at each site.

AERONET station	Coordinates	CIMEL wavelengths [nm]
Sopot, Poland	54°27′03″N, 18°33′54″E	340, 380, 440, 500, 675, 870, 1020
Gotland, Sweden	57°55′N, 18°57′E	340, 380, 440, 500, 675, 870, 1020
Minsk, Belarus	53.92°N, 27.60°E	340, 380, 440, 500, 675, 870, 1020

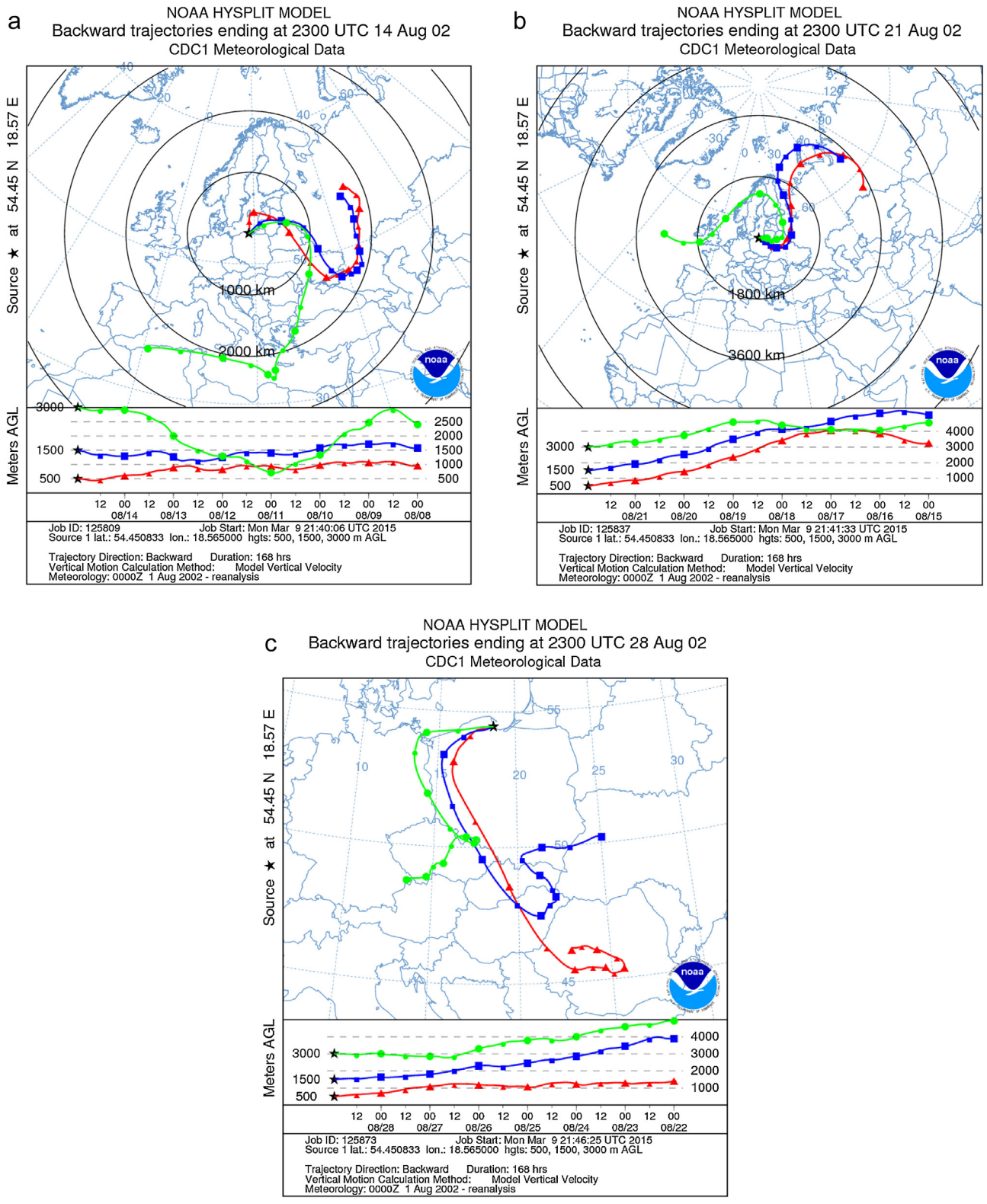

Figure 2 The 168 h NOAA HYSPLIT air-mass back trajectories for August 2002 at 500, 1500 and 3000 m above sea level (a.s.l.) ending in Sopot (on 14 August (a), 21 August (b) and 28 August (c)). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

particulates injected to the atmosphere, which reached atmospheric layers beyond the boundary layer (Evangeliou et al., 2015). The fire breakouts started in July and their peak was observed in late July and all over August 2002. In August 2002 wind patterns were such that the locally produced aerosols in the region of Belarus and Ukraine were transported over the area of the Baltic Sea (Figs. 2 and 3).

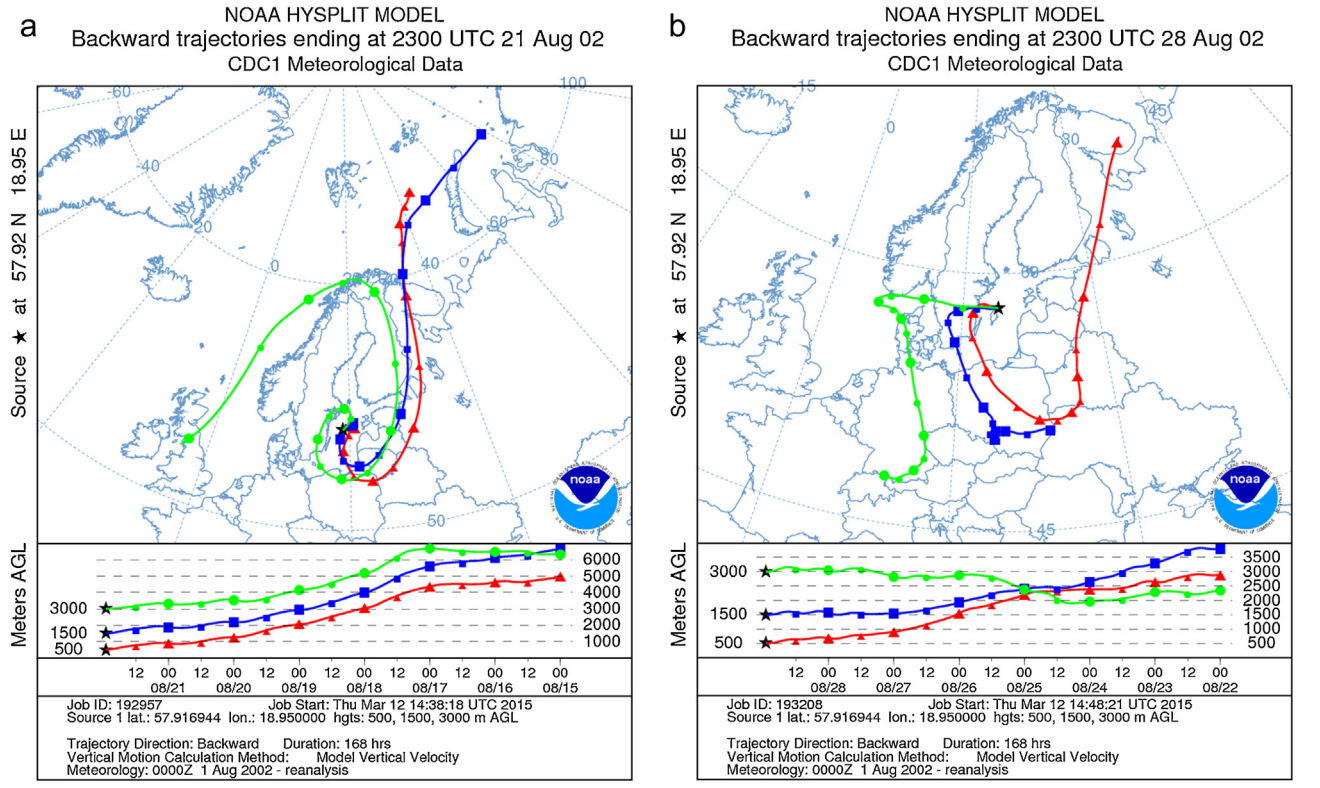

Figure 3 The 168 h NOAA HYSPLIT air-mass back trajectories for August 2002 at 500, 1500 and 3000 m above sea level (a.s.l) ending in Gotland (on 21 August (a) and 28 August (b)). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Figs. 2 and 3 show that between 8 and 28 August 2002 air masses moved over the area of wild fire regions towards the Baltic Sea. For the station in Sopot this is especially true for the first 2 decades of August and less for the last 10 days. The trajectories for Sopot crossed the wild fire areas at all three chosen altitudes, i.e. 500, 1500 and 3000 m a.s.l. In case of the Gotland station basically entire month is characterized by the air masses which crossed the region of wild fires at all three altitudes. In majority of cases all air masses passed over the station in Minsk.

Figs. 4 and 5 show level 2.0 AOD values at all three stations for entire 2002 and August 2002, respectively.

These 2 figures provide a general overview of the AOD changes over a period of an entire year. They reveal that AOD values in summer, especially in August are significantly increased in comparison to other months in 2002. The average annual AODs at all stations are unusually high and they differ from those in 1999 and 2003 (Table 2). These two years have been chosen for comparison since there are level 2.0 data available from the 3 stations to compare with year 2002 and especially August 2002.

The differences presented in Table 2 clearly show that the AOD values were dramatically increased in 2002, especially in August 2002 in comparison with 1999 and 2003, when no wild fires of the same intensity as in 2002 were observed in the region. The differences at stations in Sopot and Gotland reach a level of 2.85 and 4.88 in August 1999 and 2002, respectively. The difference for the Minsk station between August 2002 and 2003 is of an order of 3.42. This tendency is true for all other years at all three stations, whenever data are available. Table 3 shows the level 2.0 Ångström values calculated in the same manner as in case of the AODs presented in Table 2.

Clearly the values of the Ångström exponent are high, which is normal for summer conditions since prevailing winds reach the Baltic stations from the continental areas, thus transporting large amounts of dust and generally small particles. Therefore, the annual exponent differences are far less pronounced than in case of the AODs, still the August values are the ones which differ from other years the most. The situation during particular days in August was such that on certain days the AOD values were almost unrealistically

Table 2 Average annual and August level 2.0 AOD values at 500 nm (Sopot and Gotland) and at 440 nm (Minsk) for selected years (1999, 2002 and 2003).

Station	1999	August 1999	2002	August 2002	2003	August 2003
Sopot, Poland	0.113	0.145	0.239	0.413	N/A	N/A
Gotland, Sweden	0.089	0.065	0.170	0.317	0.142	0.204
Minsk, Belarus	N/A	N/A	0.431	0.557	0.224	0.163

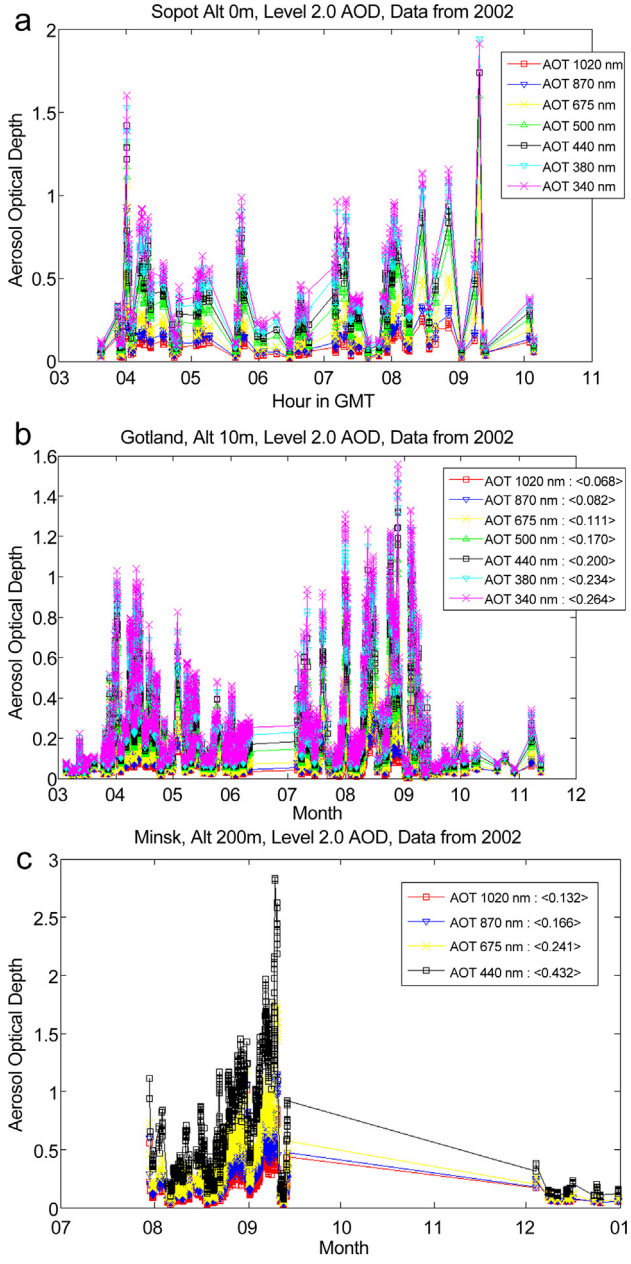

Figure 4 Aerosol optical depth measured in 2002 at three AERONET stations: Sopot (a), Gotland (b) and Minsk (c). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

high for regular aerosol conditions in the region. Table 4 shows the daily averaged level 2.0 AOD values at stations in Sopot and Gotland for all available measurement days at both stations in August 2002.

Table 4 The daily averaged level 2.0 AOD values (500 nm) at stations in Sopot and Gotland for all available measurement days at both stations in August 2002.

Day in August 2002	Sopot	Gotland
1	0.400	0.471
4	0.547	0.079
7	0.319	0.087
8	0.234	0.093
9	0.186	0.118
15	0.731	0.661
18	0.206	0.074
21	0.374	0.093
27	0.755	0.545

The data presented in Table 4 clearly indicate high AOD values at the station in Sopot during all measurement days. However, between 7 and 9 August 2002 these values are only slightly higher than on "clean" years of 1999 and 2003 (Table 2). The AOD values on 15 and 27 August 2002 are so high that they exceed the regular values (Table 2) by an order of almost 4 or 5. There is a similar situation at the station in Gotland, however the 7–9 August 2002 values are practically indicating very clean atmosphere in the station area. Again 15 and 27 August 2002 show outrageously high AODs. Therefore, in the reminder of the paper the authors concentrate on these two days. Fig. 6a–d shows level 2.0 AOD values (at seven wavelengths) on 15 and 27 August 2002 at the stations in Sopot and Gotland.

On both days and at both stations the AOD values are very high at all seven wavelengths. With exception to 27 August 2002 at the station in Gotland the AODs are rather uniform during the measurement period. The average daily level 2.0 Ångström parameters calculated from 440 and 870 nm wavelengths for 15 and 27 August 2002 for stations in Sopot and Gotland were 1.475, 1.595 and 1.492, 1.662, respectively. All these values indicate presence of fine particles in the atmosphere, which were absolutely dominating on these two days (Fig. 7a–d).

On both days fine particles were dominating the atmosphere measured at both stations and on 27 August 2002 they were basically the only fraction present in the air over the both stations. This indicates that the particles could have been advected to the measurement area from other regions and thus the NOAA HYSPLIT model was applied to calculate the air mass back trajectories. The 96 h air mass back trajectories calculated for 15 and 27 August 2002 for the stations in Sopot and Gotland are presented in Fig. 8a–d.

On 15 August 2002 air masses were advected to both stations from the area of Belarus and Ukraine at all three

Table 3 Average annual and August level 2.0 Ångström values (440/870 nm) at stations in Sopot, Gotland and Minsk for selected years (1999, 2002 and 2003).

Station	1999	August 1999	2002	August 2002	2003	August 2003
Sopot, Poland	1.232	1.693	1.188	1.561	N/A	N/A
Gotland, Sweden	1.120	1.510	1.258	1.650	1.003	1.290
Minsk, Belarus	N/A	N/A	1.369	1.505	1.312	1.586

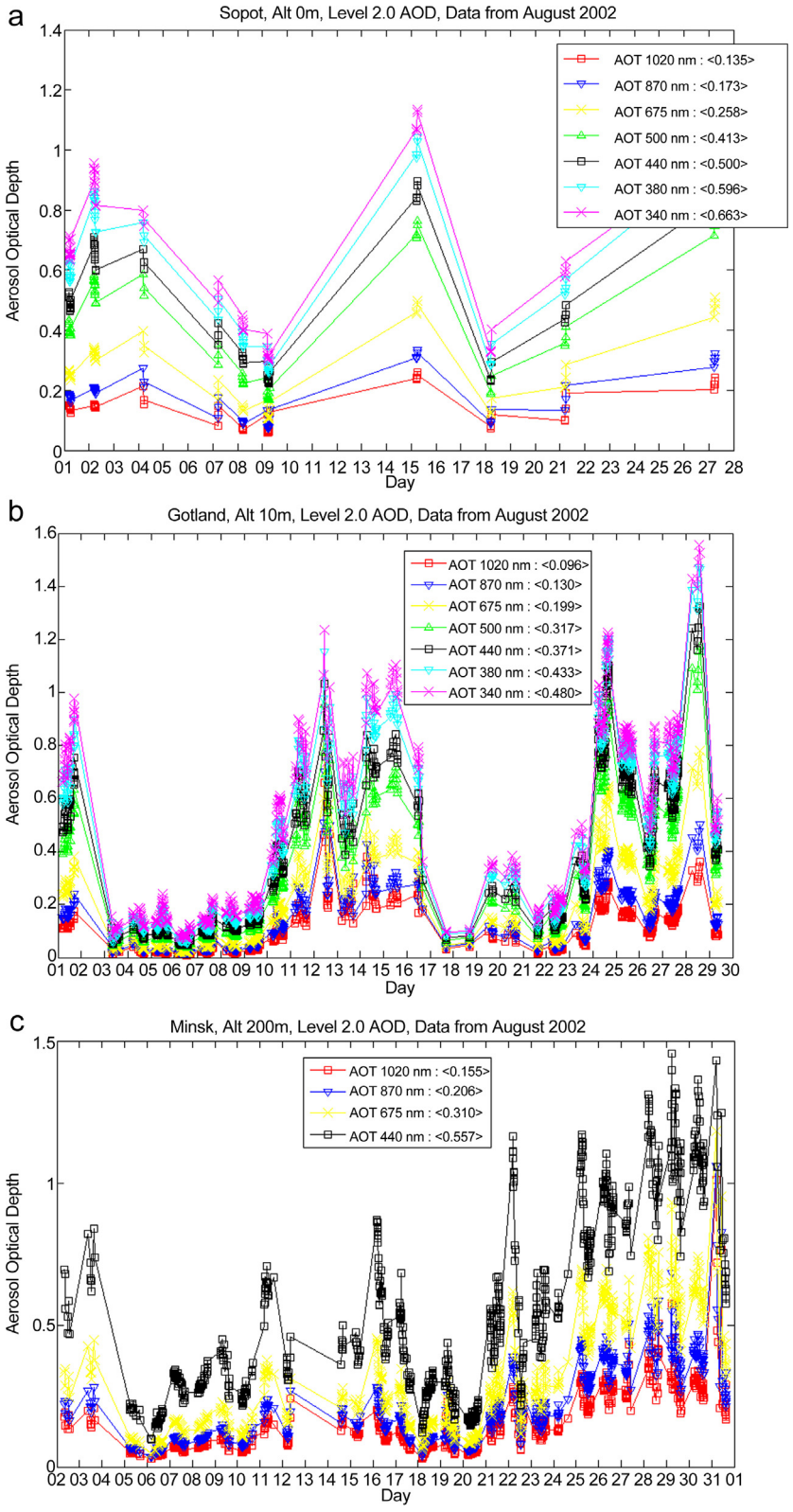

Figure 5 Aerosol optical depth measured in August 2002 at three AERONET stations: Sopot (a), Gotland (b) and Minsk (c). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

chosen levels, 500, 1500 and 3000 m a.s.l. Considering that majority of particles represented the fine mode the indication is that these particles must have travelled, and probably stayed in the measurement areas for some time, at higher altitudes, above the boundary layer (the 3000 m a.s.l. trajectory is the one which crossed over the fire areas). On 27 August 2002 the situation is slightly different since the Sopot trajectories again crossed the fire areas, especially the

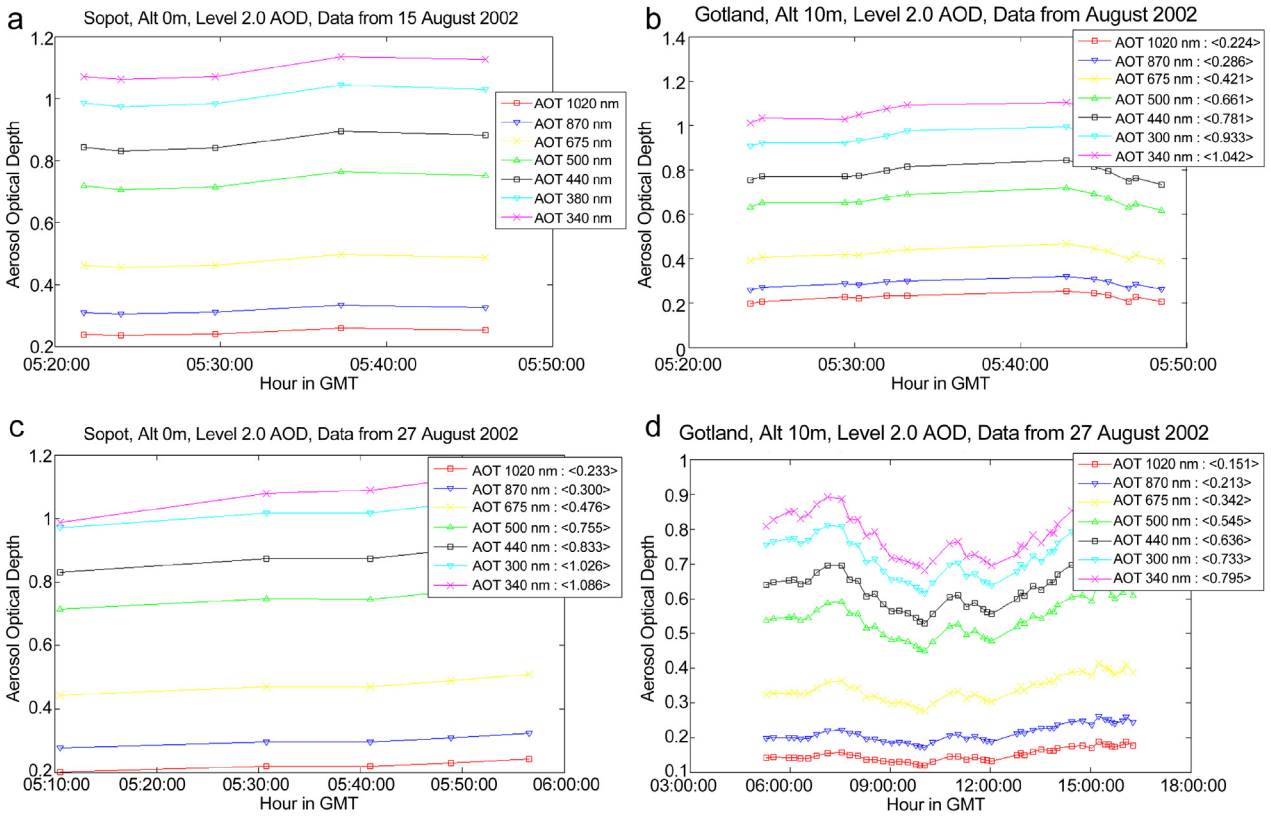

Figure 6 Level 2.0 AOD values (at seven wavelengths) on 15 and 27 August 2002 at the stations in Sopot and Gotland. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

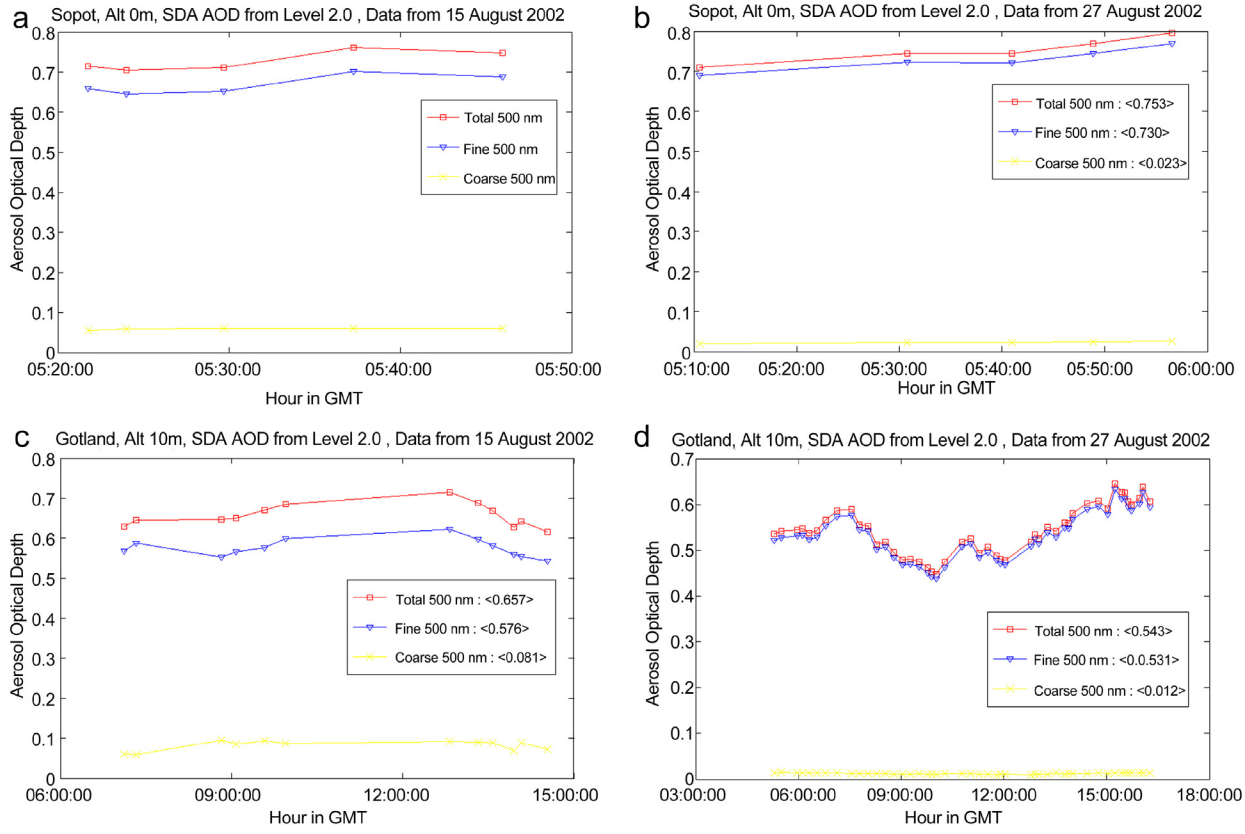

Figure 7 The level 2.0 SDA fine/coarse AOD values on 15 and 27 August 2002 for stations in Sopot and Gotland. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

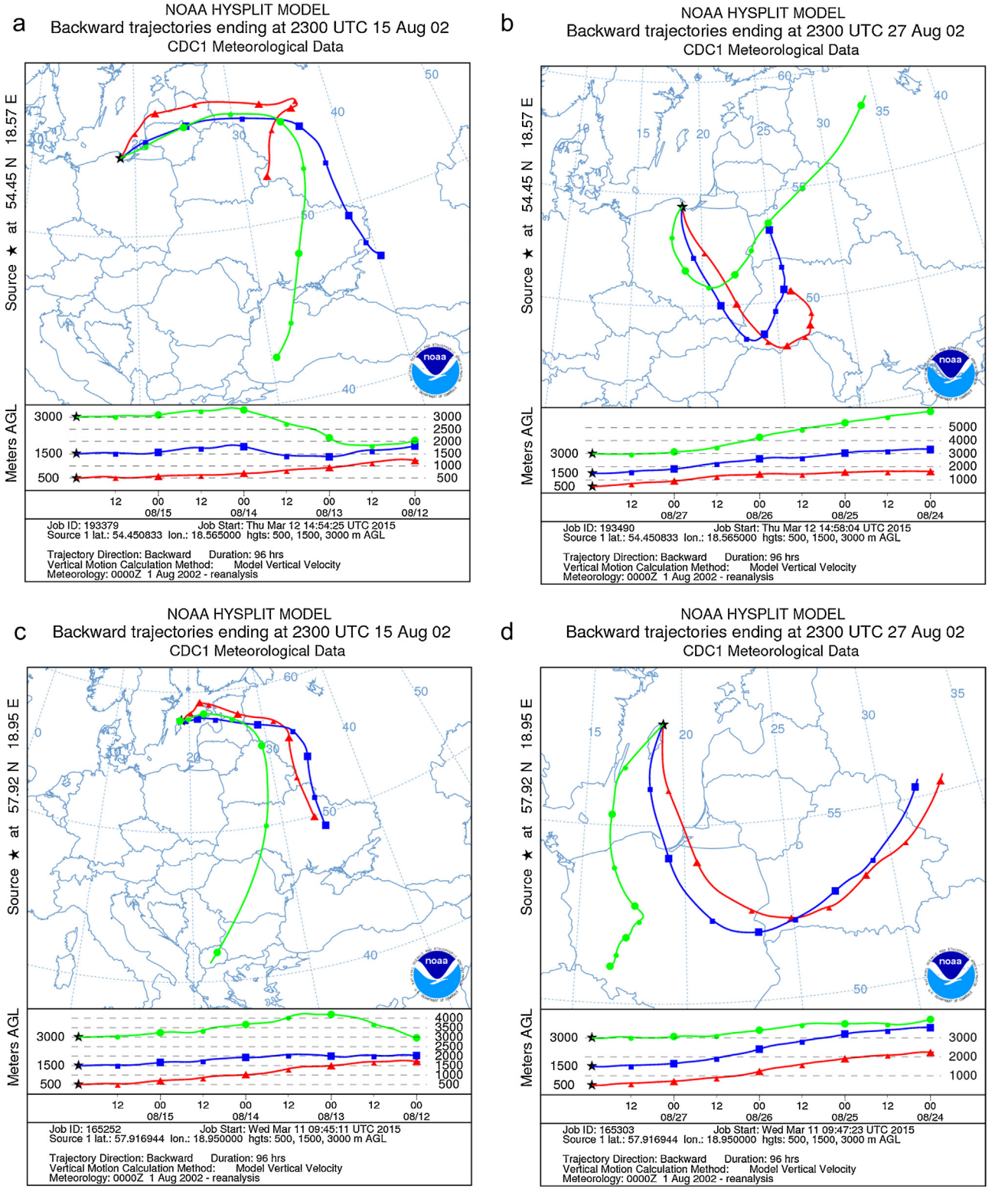

Figure 8 The 96 h air mass back trajectories at three altitudes calculated for 15 and 27 August 2002 for the stations in Sopot and Gotland. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

3000 m one, but in case of the Gotland station they also travelled over the Baltic Sea, earlier crossing Sopot. In this case it seems like the air masses might have transported fine particles over the Gotland station at altitudes lower than 3000 m but higher than 1500 m a.s.l.

The entire region on both days showed much higher AOD values than expected. Fig. 9 shows the MODIS obtained AODs at 550 nm for the Baltic and Poland, Lithuania, Belarus and Ukraine regions for 15 and 27 August 2002.

The increased AODs for the Belarus, Ukraine and the Baltic region (0.6—0.7) are evident on both days and in comparison to the rest of Europe. These pictures together with the air mass back trajectories show that the fine particles were advected to the Baltic region with south westerly winds which moved the air masses from the areas of wild fires.

Between June and mid-September 2002 the Baltic region was within the low pressure system from the area of the Mediterranean with very low speed winds from the south and south-east. On 15 August 2002 the Baltic region and especially both stations were in between the high pressure system over Scandinavia and Finland and low pressure system over Ukraine. On 27 August 2002 the situation was different since

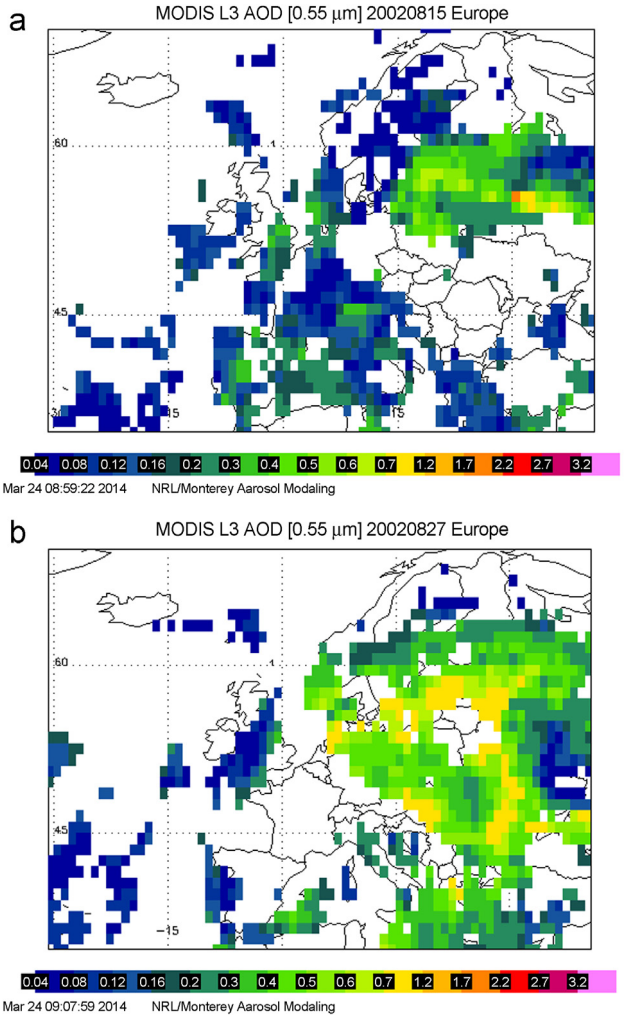

Figure 9 MODIS derived AODs at 550 nm for the Baltic and Belarus and Ukraine regions for 15 and 27 August 2002. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

both western part of the Baltic and Scandinavia were under the low pressure system and the eastern part of Europe, including Ukraine, under the high pressures system. As a result in both cases Sopot and Gotland stations were affected by the slow descent of air masses (Fig. 10a and b).

Additional information about potential transport of air pollution from the simulations of optical depths can be obtained from a long-range pollution transport model. For this purpose the authors used the Navy Aerosol Analysis and Prediction System (NAAPS) (Christensen, 1997; Witek et al., 2007). The NAAPS simulations of optical depth for 15 and 27 August 2002 for the discussed region are presented in Fig. 11.

Fig. 11 reveals that the pollution plumes on 15 and 27 August 2002 might have differed in both composition and dynamics. On 15 August 2002 the optical depth values were high in the entire region and they increased between midnight and noon. The optical depth values indicate presence of sulfate particulates in the air, while their surface concentrations are relatively low. They are produced by chemical reactions in the atmosphere from gaseous precursors. The two main sulfuric acid precursors are sulfur dioxide (SO_2) from anthropogenic sources, biomass burning and volcanoes, and dimethyl sulfide (DMS) from biogenic sources, especially marine plankton. Clearly their increased presence in the air may be connected with wild fires in the region. On 27 August 2007 situation is different since majority of optical depth values are related to a smoke plume, presumably black

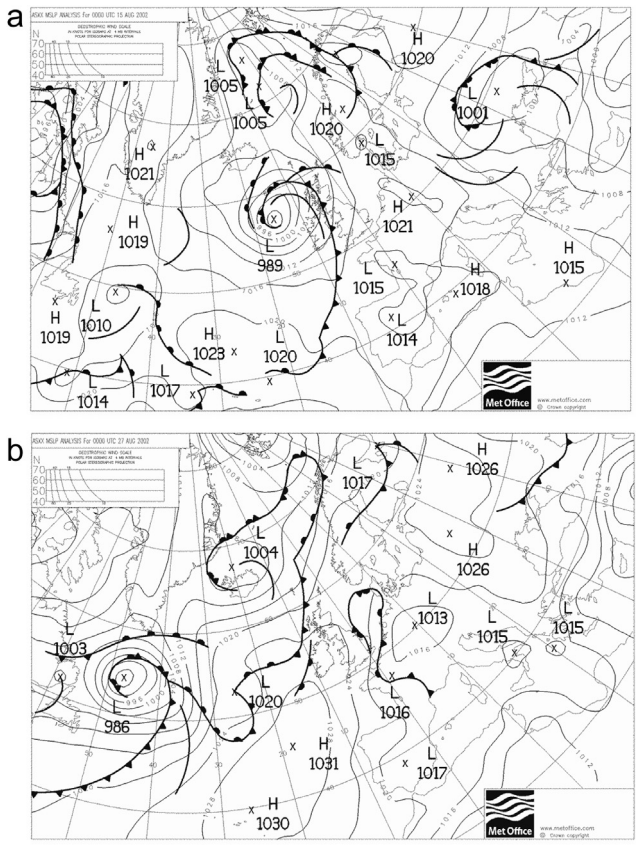

Figure 10 Air pressure systems on 15 and 27 August 2002 in the study area. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

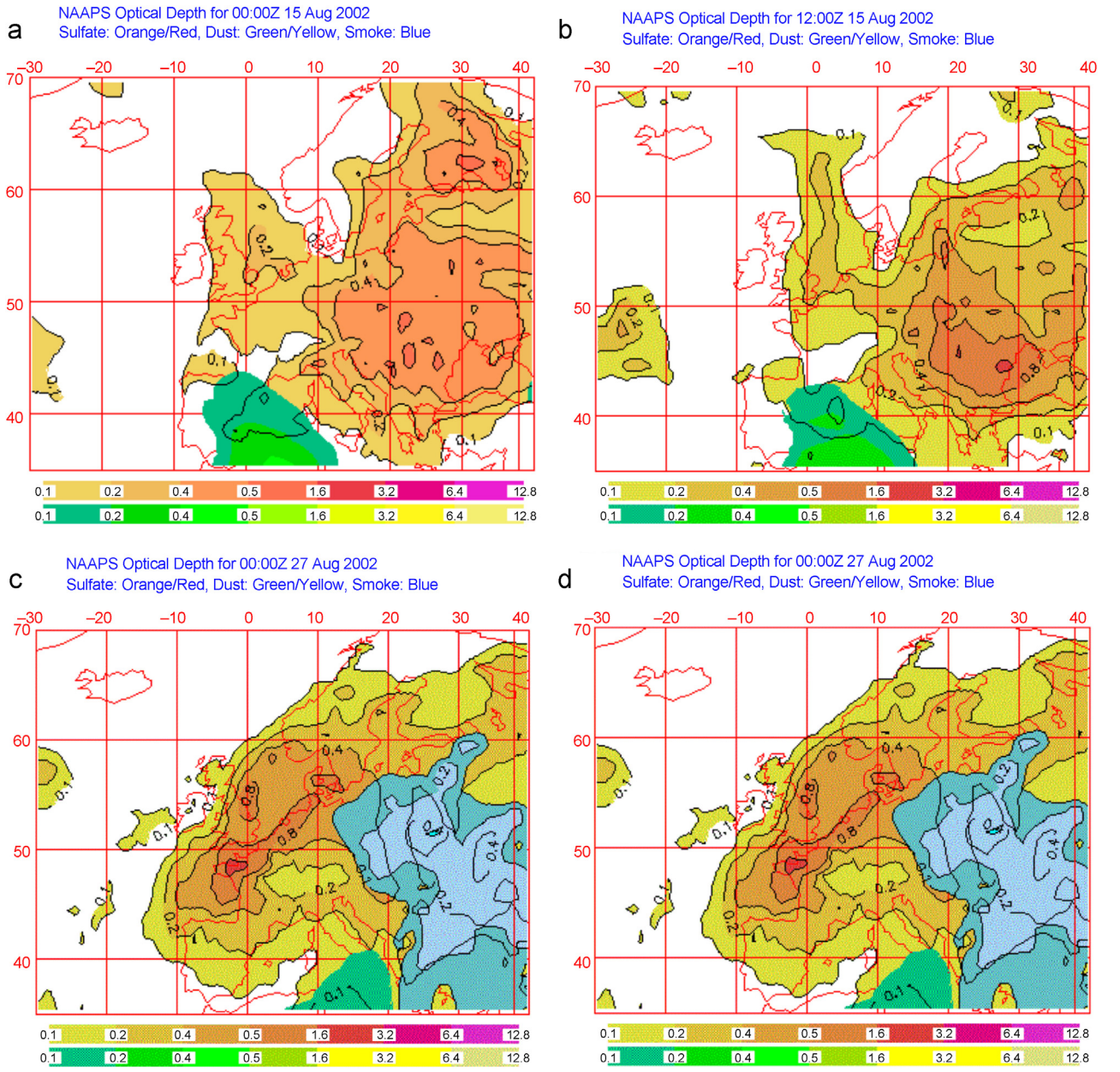

Figure 11 The NAAPS simulations of optical depths for 15 and 27 August 2002 at 00:00 and 12:00 for the discussed region. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

carbon, which did not really change much with time. The aerosol concentrations are slightly increased and also did not change significantly during the day.

4. Conclusions

The authors discussed the changes of aerosol optical depth in the region of eastern Europe and the Baltic Sea due to wild fire episodes which occurred in the area of Belarus and Ukraine in 2002. It is evident that during August 2002 the entire region was under the influence of polluted air masses from over the wild fire areas. The air pressure situation and slow wind speeds also facilitated the development of such conditions. This resulted in very high AOD levels, which significantly increased the annual averages. On particular days of August 2002 the AOD values reached a level of over 0.7. In all cases fine particles had been responsible for such situation and they fully dominated the entire ensemble of aerosol particles. They were either sulfates or smoke particles. Such situation was unique over a period of many years and it had its serious consequences for the region and especially for the Baltic Sea.

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