

## BIOMASS BURNING IMPACT ON BLACK CARBON AEROSOL MASS CONCENTRATION AT A COASTAL SITE: CASE STUDIES

V. Ulevičius, S. Byčenkienė, N. Špirkauskaitė, and S. Kecorius

*Center for Physical Sciences and Technology, Savanorių 231, LT-02300 Vilnius, Lithuania*

E-mail: ulevicv@ktl.mii.lt

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During 25 March – 5 April 2010 intense wildfires in the Kaliningrad region (Russia) occurred. The resultant smoke plume blanketing the Lithuanian western part was seen in satellite images. Concurrently, an extremely high black carbon (BC) aerosol mass concentration was observed at the background Preila site (55°55' N, 21°00' E, 5 m a.s.l., Lithuania). The surface measurements and calculation of Ångström exponent of the absorption coefficient  $\alpha$  carried out separately for shorter and longer wavelengths (i. e.,  $\lambda = 370\text{--}520$  nm and  $\lambda = 590\text{--}950$  nm) showed that high levels of BC aerosol were related to the transport of air masses rich in biomass burning products from the Kaliningrad region caused by active grass burning. During this event the BC aerosol mass concentration of 1-hour average reached  $13000\text{ ng m}^{-3}$ , while normally annual mean concentration values are about  $750\text{ ng m}^{-3}$ . The transport of the burning products from fire areas is associated with southeastern flow and strong advection of warm and dry air from South Europe in the lower troposphere. During the event the highest mean values of Ångström exponent of the absorption coefficient  $\alpha_{370\text{--}520}$  and  $\alpha_{590\text{--}950}$  were observed ( $2.0 \pm 0.4$  and  $1.6 \pm 0.3$ , respectively). The mean values of Ångström exponent of the absorption coefficient during the study period obviously indicate that a major part of carbon mass in aerosol particles transferred by the regional air masses comes from the wildfire location.

**Keywords:** black carbon aerosol, aethalometer, biomass burning, Ångström exponent of the absorption coefficient

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### 1. Introduction

The biomass burning has long been recognized as a significant source of reactive species such as aerosol, carbon monoxide (CO), hydrocarbons, and nitrogen oxides ( $\text{NO}_x$ ), which play an important role in the chemistry and radiative budget of the troposphere [1, 2]. During biomass burning periods, the visibility in affected areas can be heavily reduced, and the health effects on the local population can be substantial. Moreover, biomass burning particles are efficient cloud condensation nuclei, they can influence the formation of clouds and affect the Earth's climate by directly scattering and absorbing the atmospheric radiation [3–6]. Along with absorbing aerosols, black carbon (BC) or elemental carbon fraction is known to be most effectively absorbing solar radiation [7, 8].

In Eastern Europe the open biomass burning of agricultural residues and other waste remains a significant issue [9–11]. Smoke aerosols released from biomass fires are a complex of chemical mixture of organic and inorganic materials, simultaneously involving the gaseous, liquid, and solid phases [2, 12–14]. The

long-range transport of emissions from wildfires from Ukraine and European part of Russia increases the particular matter concentrations in Eastern and Northern European countries, frequently during spring and summer [15–17]. After emission into the atmosphere, BC is processed by the condensation of secondary material such as sulfates, nitrates, or organic matter increasing the size and hygroscopicity of the particles [18]. During atmospheric ageing, the refractory component of BC is chemically quite stable and thus is relatively invariant, making BC an ideal tracer of primary pollution [19]. Although concentrations of BC are determined permanently at many locations worldwide such as North America [20, 21], South America [22, 23], Africa [24], and the Mediterranean [25], the reports have shown that biomass burning particles found in different regions vary in concentrations. The variety of particle characteristics may be due to several factors such as intensity of fires, moisture, weather conditions, and other variables.

In this work, the regional air mass transport event that occurred in Lithuania on 25 March – 5 April 2010 was investigated. The potential source areas of particles

and the presence of open biomass burning were investigated using terrestrial information operationally derived from satellite data including those from NASA's MODIS instrument. Air mass backward trajectories and smoke dispersion modelling data were used to identify the origin and to characterize the regional transport from open biomass burning as a particle source of the atmospheric pollution into a synoptic weather context. The BC aerosol mass concentrations in conjunction with analysis of Ångström exponent of the absorption coefficient were investigated.

## 2. Methods

### 2.1. Site description

The black carbon aerosol mass concentration measurements were performed at the Preila environmental pollution research station ( $55^{\circ}55'N$ ,  $21^{\circ}00'E$ , 5 m above the sea level) in the coastal/marine environment. This station is located on the Curonian Spit, which separates the Curonian Lagoon and the Baltic Sea, and thus can be characterized as a regionally representative background environment. The Curonian Lagoon, the largest coastal bay in the Baltic Sea, is a highly eutrophied water body. It is an enclosed shallow (mean depth 3.7 m) bay connected to the Baltic Sea by the narrow (400–600 m wide) Klaipėda strait. One of the nearest industrial cities, Klaipėda, is at a distance of about 40 km to the north and the other major city, Kaliningrad (Russia), is 90 km to the south from Preila.

### 2.2. Fire smoke transport and air mass isentropic backward trajectories

To identify possible events of regional air mass transport, the location of a thermal anomaly was detected with the MODIS sensor using data from the middle infrared and thermal infrared bands [26]. The MODIS monthly fire maps, available since 2001, have shown that these events of biomass burning in March–April occur annually. However, the duration, geographical extent, and emission of the smoke from these fires differ year by year. The results of the Navy Aerosol Analysis and Prediction System (NAAPS) model were used to determine the distribution of smoke aerosols from fires (model description and results are available from the web pages of the Naval Research Laboratory, Monterey, CA, USA; <http://www.nrlmry.navy.mil/aerosol/>). The NAAPS model has recently been modified to

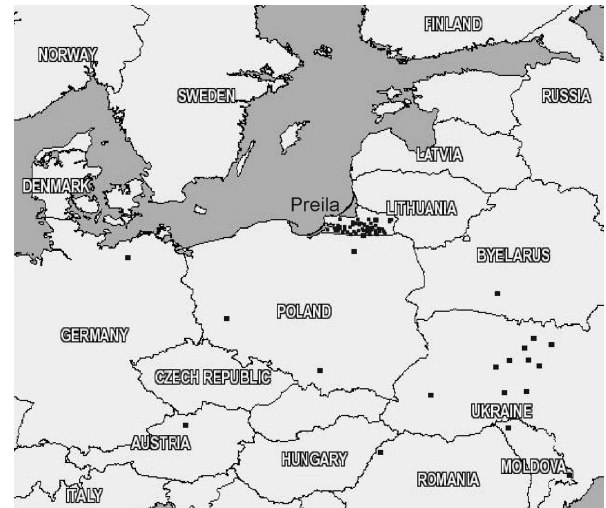


Fig. 1. Locations of the active fires identified from MODIS observations (Europe subset: Terra 1 km true colour image for 3 April 2010).

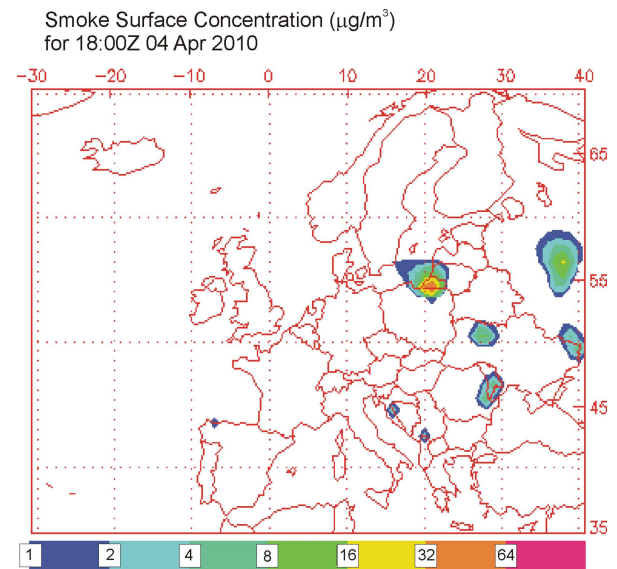


Fig. 2. Smoke mass mixing ratio at the surface by NAAPS.

incorporate real-time observations of biomass burning based on the joint Navy/NASA/NOAA Fire Locating and Modeling of Burning Emissions system (FLAMBE, <http://www.nrlmry.navy.mil/flambe/>) [27, 28]. The aerosol characteristics with respect to categorized air mass isentropic backward trajectories for initial estimation of the wildfire potential source location and quantitative contribution have been analysed. Air mass isentropic backward trajectories were analysed using the HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT4) model [29] with the Final Analyses (FNL; 2010) and the Global Data Assimilation System (GDAS) meteorological databases at the NOAA Air Resources Laboratory's web server

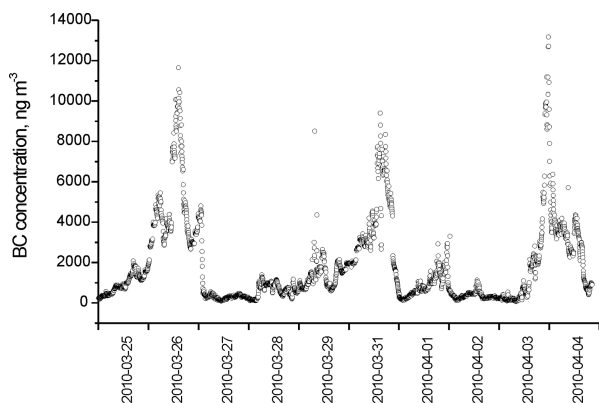


Fig. 3. Time series of BC mass concentration.

[30]. The weather and baric topography maps from the European Meteorological Bulletin (Germany weather service, <http://www.wetterzentrale.de>) were analysed.

### 2.3. Black carbon aerosol mass concentration measurements

A Magee Scientific Co. Aethalometer™, Model AE40 Spectrum, manufactured by Optotek, Slovenia was deployed at the site and provided real-time, continuous measurements of the BC mass concentration. The optical transmission of carbonaceous aerosol particles was measured sequentially at seven wavelengths  $\lambda$  (370, 450, 520, 590, 660, 880, and 950 nm). The concentration of black carbon corresponds to the 880 nm wavelength.

The aethalometer data recorded with a 2-minute time basis were compensated for loading effects using an empirical algorithm [31].

A power law fit is commonly applied to describe the wavelength dependence of absorption coefficient  $b_{\text{abs}}$ :

$$b_{\text{abs}} \propto \lambda^{-\alpha}, \quad (1)$$

where  $\lambda$  is the wavelength and  $\alpha$  is the usually called Ångström exponent of the absorption coefficient and is computed by fitting an exponential curve [32]. The wavelength dependence of the light absorption can be better approximated by separate exponential fits of the lower (370–520 nm) and higher (590–950 nm) wavelengths (coefficient of determination  $r^2 > 0.99$ ) obtained by an exponential curve fit over all seven wavelengths.

Spatial and temporal variations of PM<sub>2.5</sub> and PM<sub>10</sub> within Klaipėda city were investigated using the data sets collected from air quality monitoring site of the Lithuanian Environment Protection Agency. The mass concentrations of PM<sub>2.5</sub> and PM<sub>10</sub> were obtained with the automatic analyzers using the French Environment S.A model 1M monitors based on the  $\beta$ -attenuation method. The measurements of the carbon monoxide and nitrogen oxides were performed continuously with the Environment S.A model CO11M (infrared gas absorption method) for CO and model AC31M (chemoluminescent method) for NO<sub>x</sub> [17].

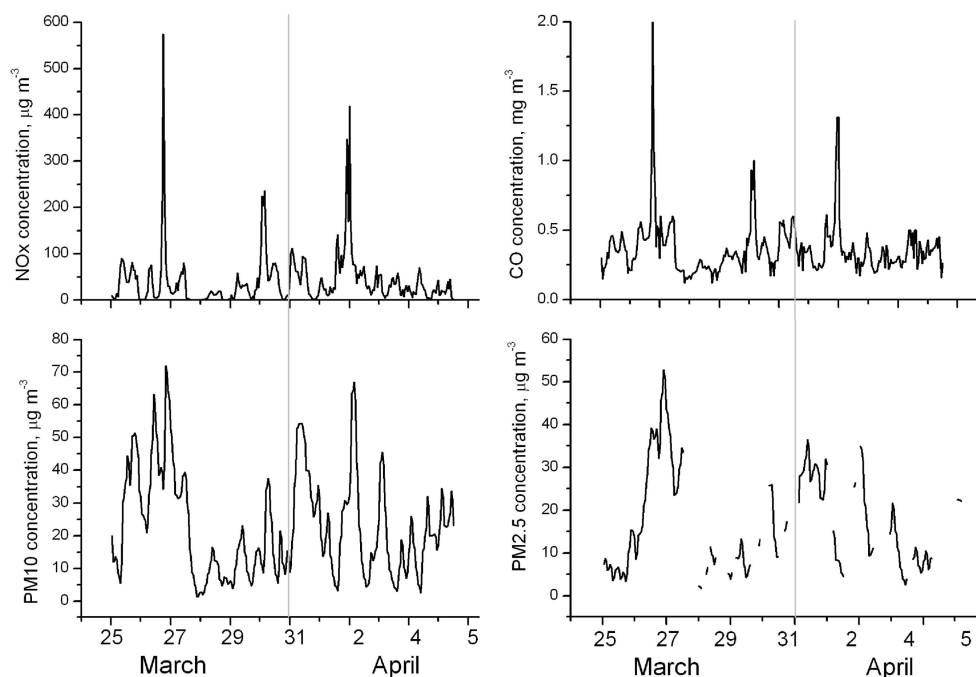


Fig. 4. Time series of NO<sub>x</sub>, CO, PM<sub>10</sub>, and PM<sub>2.5</sub> concentration as measured at the Klaipėda AQ station during March–April 2010.

### 3. Results

#### 3.1. Analysis of high black carbon concentration episodes

In this study we focus on the strongest period of wildfire in the Kaliningrad region that occurred on 25 March – 5 April 2010. They commonly start in March, and the number of fires is largest in April. The intensity of these fires varies from year to year, which is largely caused by meteorological conditions in spring, such as the occurrence of precipitation, the temperature, etc. For example, in Fig. 1 the Fire Mapper – a web mapping interface that displays hotspots/fires detected by the MODIS Rapid Response System – products have provided data about the area and coverage of fires.

Satellite image and smoke dispersion by NAAPS (Fig. 2) suggested substantial emissions and regional transport from the large biomass burning in the Kaliningrad region at that time. During this period territories close to Preila were covered with high BC aerosol mass concentrations that were clear outliers in their respective series, both for hourly concentrations and 24-h means (Fig. 3). Biomass burning products were redistributed over Lithuania by the large-scale atmospheric circulation. As shown in Fig. 1, the MODIS clearly illustrates the fire location. Model forecasts by NAAPS for this period suggested substantial emissions and regional transport of smoke (for smoke, we assume particles of  $0.01 \mu\text{m}$  radius) (Fig. 2) from the large fires burning in the Kaliningrad region at that time.

During the study the NAAPS model predicted the optical depth of 0.4 at a wavelength of  $0.55 \mu\text{m}$  for three components: sulfate, dust, and smoke over the Lithuanian western part due to smoke from the wildfires. It should be mentioned that the measured BC aerosol mass concentration might originate from some other burning region, which was located closer to the site but missed by MODIS, possibly due to clouds.

During this event the BC aerosol mass concentration of 1-hour average reached  $13\,000 \text{ ng m}^{-3}$ , while normally annual mean concentration values are about  $750 \text{ ng m}^{-3}$  [33]. During the study period on 25 March – 5 April 2010 the urban air quality monitoring station

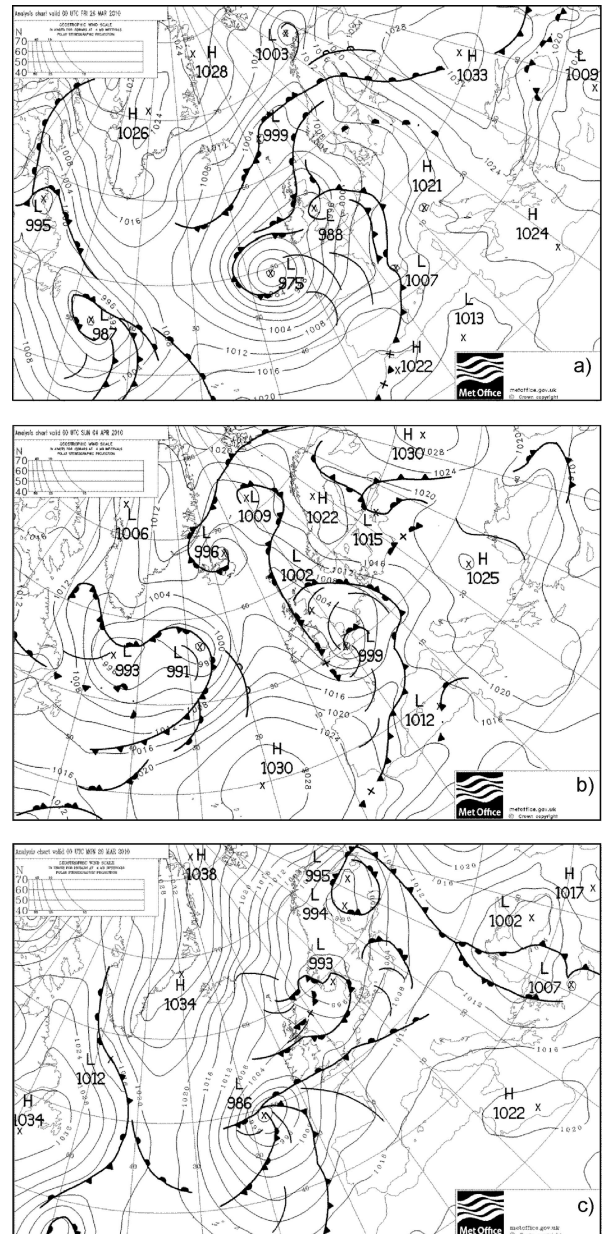


Fig. 5. Weather maps from the European Meteorological Bulletin (Germany Weather Service, <http://www.wetterzentrale.de>) for 00 UTC (a) 02 Lithuania local wintertime, 26 March; (b) 03 Lithuania local summertime, 29 March; (c) 03 Lithuania local summertime, 4 April 2010.

recorded moderate or higher concentrations of PM<sub>2.5</sub>, PM<sub>10</sub>, NO<sub>x</sub>, and CO (Fig. 4). As can be seen from

Table 1. The mean daily meteorological parameters at the Klaipėda (Šilutės motorway) air quality station during the study period (25 March – 5 April 2010).

Days	25	26	27	28	29	30	31	1	2	3	4	5
$T, ^\circ\text{C}$	9.2	10.4	3.4	3.8	5.3	8.9	13.5	4.7	4.6	2.8	9.5	5.3
Relative humidity (RH), %	80	72	96	59	91	85	72	86	71	75	74	74
Pressure, hPa	1014	1003	990	1003	1005	1002	999	1013	1015	1016	1015	1020
Wind speed, $\text{m}\cdot\text{s}^{-1}$	3.3	1.6	3.2	1.4	0.8	2.5	2.4	0.4	1.3	0.3	2.6	1.0

Fig. 4, appreciable increase in the PM10 concentration has been monitored during the course of the pollution event that suggests that a high concentration event has been a widespread regional transport event. A strong CO and NO<sub>x</sub> enhancement (from 0.5 to 2 mg m<sup>-3</sup> and from 100 to 500 μg m<sup>-3</sup>, respectively) was observed. Bearing in mind that BC found during pollution events is mainly the product of incomplete combustion of biomass, the relative contribution compared to that from other sources is not certain. The pollution events at the urban site were not influenced only by urban sources but also by biomass burning during early spring [34].

The dispersion of biomass smoke and resulting high concentration events, their severity, duration, and affected areas depend on meteorological conditions. For the current event, the weather maps showed that its ex-

ceptionally long duration was caused by a long-lasting anticyclonic system over western Russia, which caused the fire fumes to be transported from the burning region. Strong temporal variations in the course of the BC aerosol mass concentration showed a possible correlation with the change of atmospheric processes resulting in primary carbonaceous aerosol outflow from biomass burning areas in the Kaliningrad region. During 25–26 March, deep cyclone (975 hPa) over the eastern Atlantic Ocean and high pressure (1033 hPa) systems over Russia formed the atmospheric circulation over Lithuania associated with south–eastern flow and strong advection of warm air from South Europe in the lower troposphere (Figs. 5(b), 6). On the next days (27–30 March) low pressure with cold or occluded fronts system and the north–eastern flow transporting polar air masses dominated over the Baltic countries (Fig. 5).

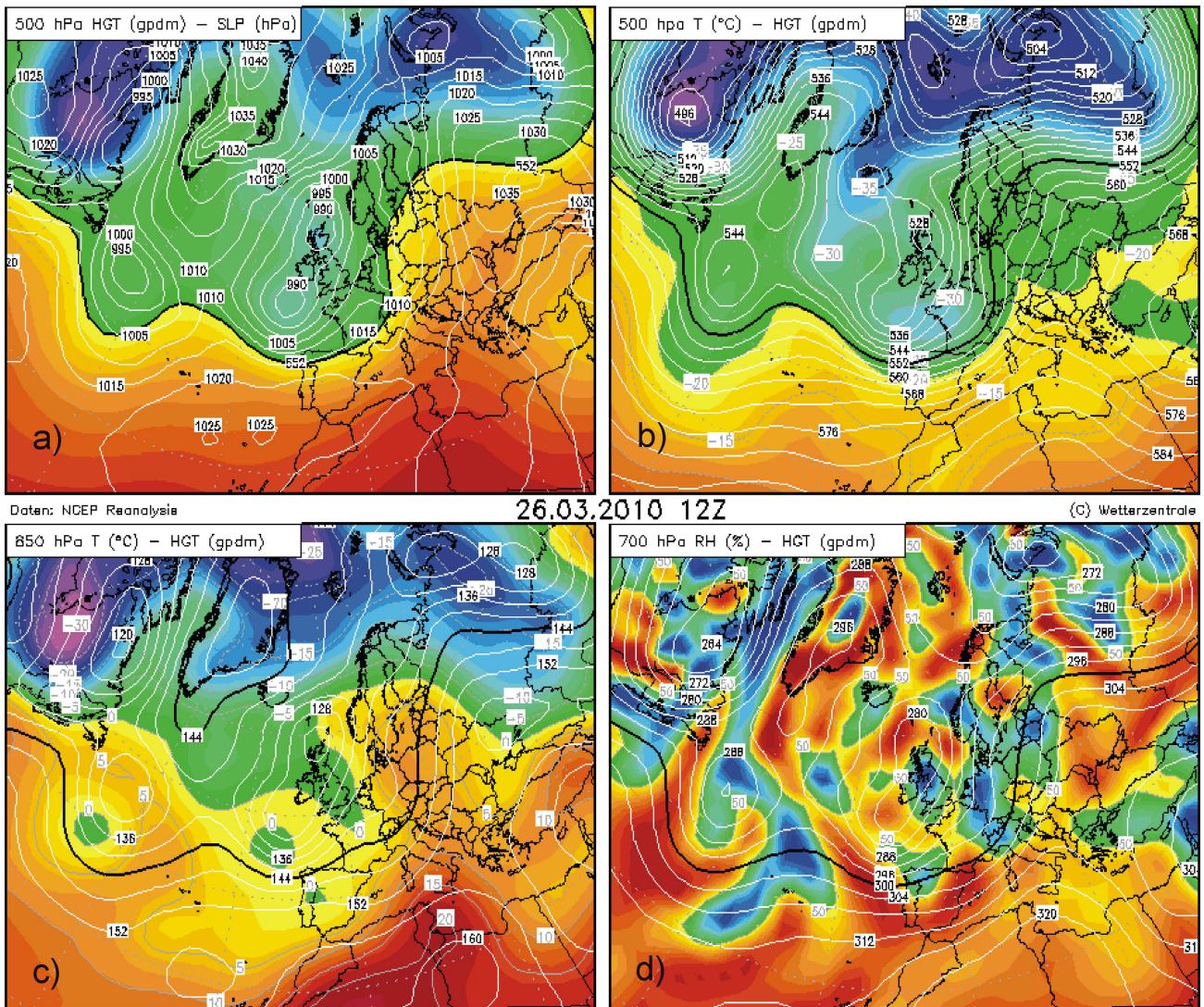


Fig. 6. Baric topographic maps: (a) Absolute topography (AT) 500 hPa above sea level; (b) AT 500 hPa T °C; (c) AT 850 hPa T °C; (d) AT 700 hPa RH % for 26 March 2010 12 UTC.

Meteorological data for the study period confirm a significant drop in the temperature (5–7 °C) and an increase in the relative humidity (Table 1).

During these days the BC mass concentration yet again reached the background level ( $750 \text{ ng m}^{-3}$ ) (Fig. 3). In all probability, this was related to the passage of cold front with precipitation cleaning the ground level air from biomass burning products at the Preila site and a radical change of air mass advection direction was observed. A sharp increase of the BC mass concentration on 31 March and 4 April (Fig. 3) was clearly associated with a change of the atmospheric circulation type. This was the beginning of wavy several day-long warm and dry air outburst from Southeastern Europe (mainly from Greece) (Figs. 5(c), 7(c)). As seen in Fig. 6(c, d) the tongue of warm and dry air mass extending from

Greece to Norway across the Baltic Sea was observed in the lower troposphere because the middle troposphere was covered by the processes of transport of air masses sprawling from northern latitudes. Arrival of the fresh air mass at the study site from south has shown a sudden changing of daily average temperature both on 31 March and 4 April (Table 1). The short-term advection of the air mass has shown the variation of the meteorological parameters since on the next days the transformation of air masses occurred.

Analysis of air mass isentropic backward trajectories established the transport of air masses from south over biomass burning areas, especially in lower troposphere to the north towards the Baltic countries (Fig. 8). The air mass backward trajectory analysis showed that these particles from the Kaliningrad region with air masses

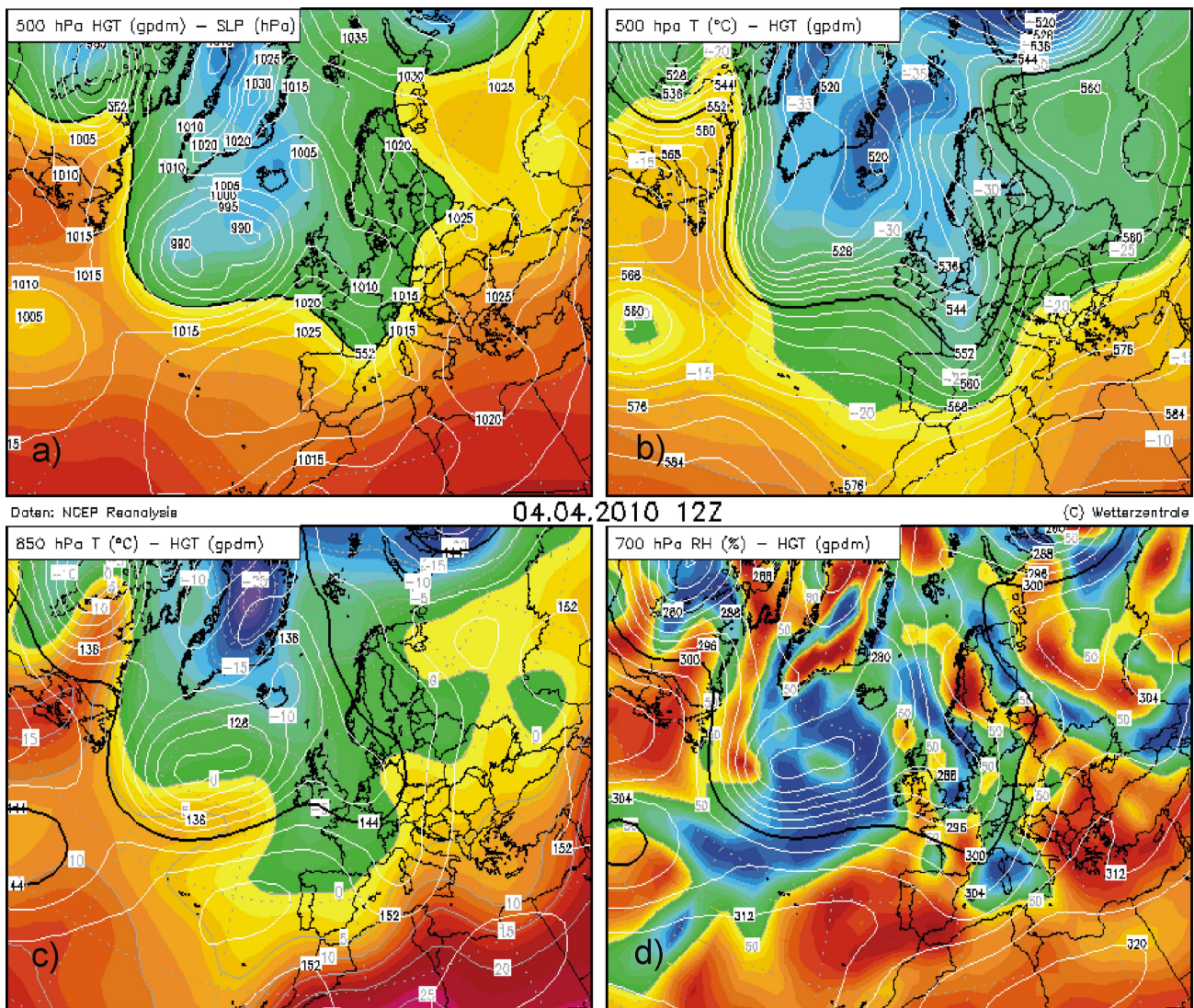


Fig. 7. Baric topographic maps: (a) AT 500 hPa above sea level; (b) AT 500 hPa  $T$  °C; (c) AT 850 hPa  $T$  °C; (d) AT 700 hPa RH % for 4 April 2010 12 UTC.

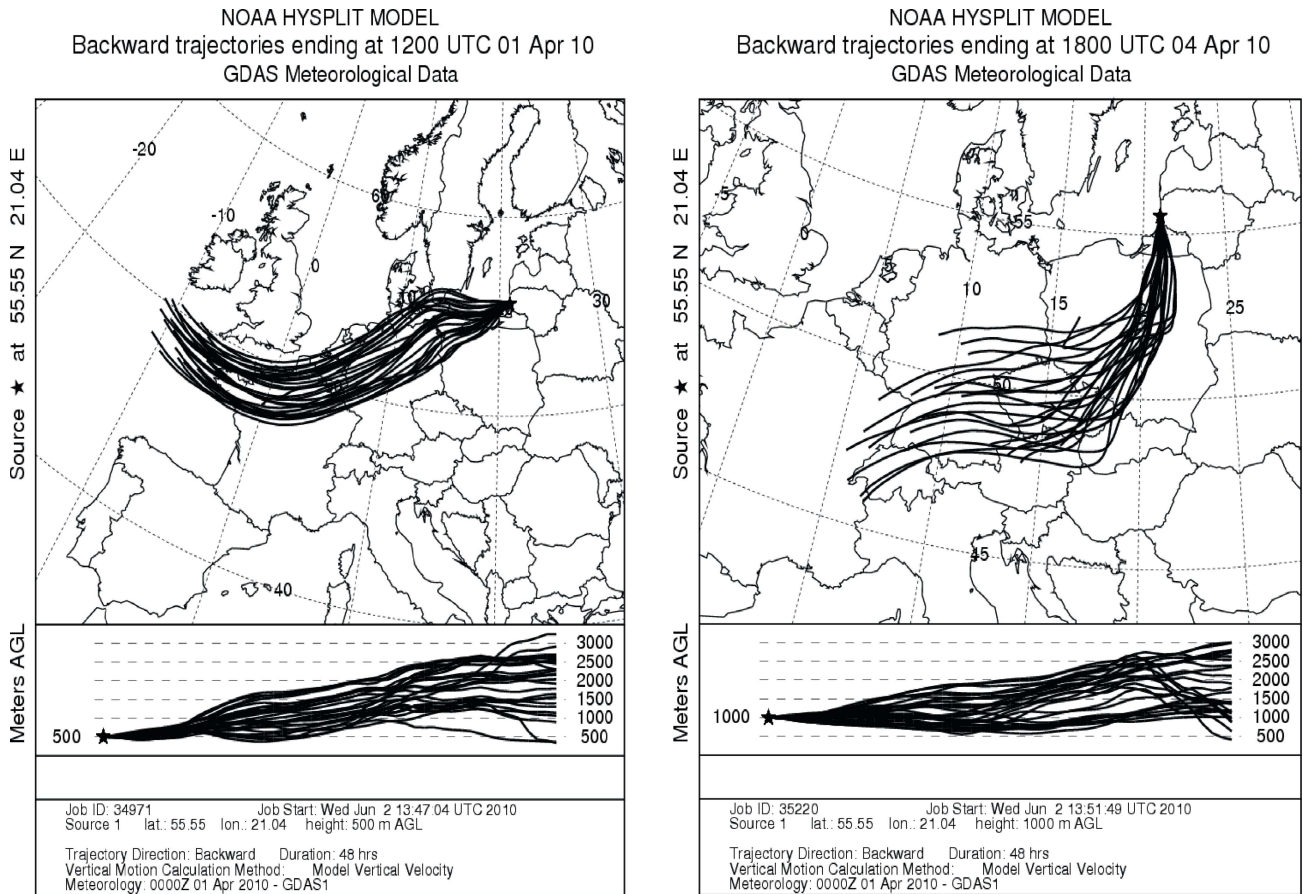


Fig. 8. Air mass regional transport from biomass burning territories in Kaliningrad region on 1 (left panel) and 4 April (right panel) 2010.

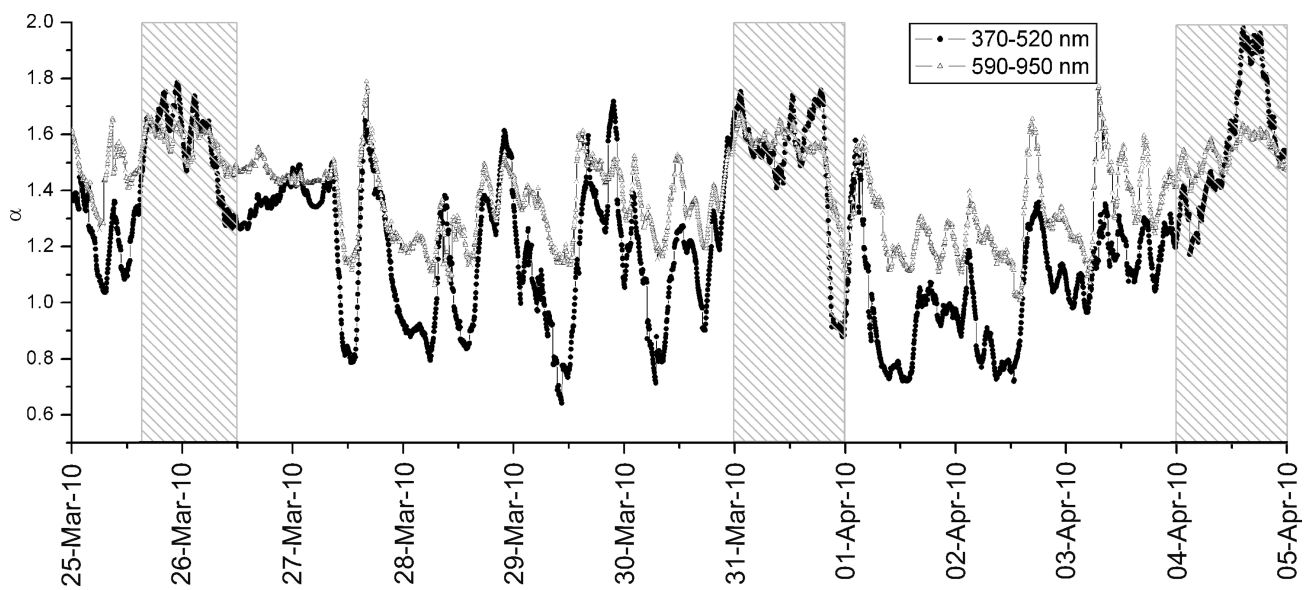


Fig. 9. The light Ångström exponent of the absorption coefficient:  $\alpha_{370-520}$  and  $\alpha_{590-950}$ .

were brought to Lithuania and caused three abrupt increases in the BC concentration (Fig. 3).

Moreover, for the study period the site was under the influence of frontal air systems (Fig. 5(b)). The dominant local wind directions were to be either from the north (N) or northeast (NE) (Table 1). It is obvious that during this period polluted air from the biomass burning regions was transported to the site.

### 3.2. The absorption exponent $\alpha$ as a function of wavelength range

The Ångström exponent of the absorption coefficient was calculated to explain the BC aerosol origin. The Ångström exponents were calculated by fitting  $b_{\text{abs}}$  for the whole available wavelength intervals: the low wavelength interval 370–520 nm and the high wavelength interval – 590–950 nm (Fig. 9).

The contrast between the events with biomass smoke and those with less smoke is most evident in the short wavelength interval. On 5 April the  $\alpha$  values were significantly higher for the shorter wavelengths ( $\alpha_{370-520} \approx 2.0 \pm 0.4$ ) compared to the longer wavelengths ( $\alpha_{590-950} \approx 1.6 \pm 0.3$ ) (Fig. 9). On non-event days, the light absorption coefficients  $\alpha_{370-520}$  and  $\alpha_{590-950}$  were within a narrow range of 1.2, with weak diurnal cycles. These high values of the absorption exponent are indicative of biomass combustion [32], in our case – wildfires in the Kaliningrad region, as indicated by the air mass isentropic backward trajectory analysis and weather maps. Kirchstetter et al. [35] reported  $\alpha$  value of 2.2 for outdoor firewood burning, 1.8 for a savanna fire, and 0.8–1.1 for traffic-dominated sites. Schnaiter et al. [36, 37] reported  $\alpha$  value of 1.1 for uncoated diesel soot (measurements with  $\lambda = 450, 550,$  and  $700$  nm). Previous investigations showed that the type of wood being burned also influenced the  $\alpha$  value as shown by Day et al. [38] where they measured fresh wood smoke from seven types of forest wood with an aethalometer ( $\lambda = 370-950$  nm) and reported  $\alpha$  values between 0.9 and 2.2.

## 4. Summary and conclusions

A combination of ground-based and satellite observations was used in this study to investigate different sources of the high black carbon aerosol mass concentration over Lithuanian western part on 25 March – 5 April 2010. During this period the measured black carbon concentration 1-hour average of  $13\,000 \text{ ng m}^{-3}$  exceeded previously established mean values of  $750 \text{ ng m}^{-3}$ .

During the open biomass fire events in the neighbourhood of Lithuania under specific weather conditions the abrupt increases of the BC concentration in the ground level air at the Preila site were observed. Transport of the burning products from fire areas is associated with southeastern flow and strong advection of warm and dry air from South Europe in the lower troposphere. During the event in 2010 the highest mean values of Ångström exponent of the absorption coefficient  $\alpha_{370-520}$  and  $\alpha_{590-950}$  were observed ( $2.0 \pm 0.4$  and  $1.6 \pm 0.3$ , respectively). The highest values were comparable to research studies on wood smoke, i. e.,  $\sim 2$ . This confirms the influence of the relative contribution of wood burning on the Ångström exponent of the absorption coefficient  $\alpha$ . Observation data from other countries demonstrate that particulate matter levels have been increasing in northern Europe and Scandinavia during spring periods, indicating that these were long-range transport pollution events.

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## BIOMASĖS DEGINIMO ĮTAKA JUODOSIOS ANGLIES AEROZOLIO MASĖS KONCENTRACIJAI JŪROS PAKRANTĖJE: ATVEJO ANALIZĖ

V. Ulevičius, S. Byčenkienė, N. Špirkauskaitė, S. Kecorius

*Fizinių ir technologijos mokslų centras, Vilnius, Lietuva*

### Santrauka

Tirtas juodosios anglies aerolio masės koncentracijos padidėjimas Lietuvos pajūryje tolimosios pernašos metu iš gaisrų apimtų teritorijų Kaliningrado srityje 2010 metų kovo 25 – balandžio 5 dienomis. Didelis išmetamų teršalų iš gaisrų apimtų teritorijų kiekis sukūrė dūmų šleifą, nusidriekusį link Lietuvos jūros pakrantės. Tuo metu Preilos aplinkos užterštumo tyrimų stotyje buvo stebėta didelė juodosios anglies aerolio masės koncentracija. Tyrimų metu Preiloje juodosios anglies vidutinė valandos masės koncentracija siekė 13000 ng m<sup>-3</sup>, kai vidutinė metinė vertė siekia 750 ng m<sup>-3</sup>. Teršalų koncentracijos matavimai ir sugerties koeficiento Angstromo

eksponentės  $\alpha$  analizė, atlikta atskirai trumpų ir ilgų bangų diapazonuose (t. y.  $\lambda = 370\text{--}520$  ir  $590\text{--}950$  nm), patvirtino, kad didelė juodosios anglies aerolio masės koncentracija buvo susijusi su biomasės degimo produktų pernaša iš Kaliningrado srities aktyvių gaisrų židinių. Nustatytos didžiausios vidutinės sugerties koeficiento Angstromo eksponentės vertės  $\alpha_{370\text{--}520}$  ir  $\alpha_{590\text{--}950}$  siekė atitinkamai  $2,0 \pm 0,4$  ir  $1,6 \pm 0,3$ . Siejant juodosios anglies aerolio koncentracijos kaitą su atmosferos cirkuliacija nustatyta, kad didžiausia koncentracija buvo susijusi su pietryčių srautu ir stipria šilto ir sauso oro advekcija į Lietuvos pajūrį iš Pietų Europos.