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JES JOURNAL OF ENVIRONMENTAL SCIENCES

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Industrial and wildfire aerosol pollution over world heritage Lake Baikal

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ARTICLE INFO

Article history:
Received 13 August 2020
Revised 6 January 2021
Accepted 10 January 2021

Keywords:
Environmental impacts
Aerosol pollution
Ecosystem
Industrial emissions
Wildfires
Coal combustion
Shipping

ABSTRACT

Lake Baikal is the biggest reservoir of fresh water with unique flora and fauna; presently it is negatively affected by climate change, water warming, industrial emissions, shipping, touristic activities, and Siberian forest fires. The assessment of air pollution - related Baikal's ecosystem damage is an unsolved problem. Ship, based expedition exploring the Baikal atmospheric aerosol loading, was performed over the lake area in July 2018. We combine the aerosol near - water and vertical distributions over the Lake Baikal basin with meteorological observations and air mass transportation simulations. Lidar sounding of aerosol fields in the troposphere assesses the atmospheric background in the pristine areas and the pollution during fire-affected periods. Aerosol optical properties (scattering and spectral absorption) converted to the particle number size, black carbon (BC) mass, and Absorption Angstrom Exponent (AAE) provide the inside into aerosol characterization. Transport of industrial emissions from Krasnoyarsk and Irkutsk regions, and wildfire plumes from Republic of Yakutia relates the pollution sources to the increased concentrations of fine particle numbers, PM_{10} and BC mass over Southern and Northern/Central Baikal, respectively. The highest PM_{10} and BC are associated to the harbor and touristic areas of intensive shipping and residential biomass burning. Deposition estimates applied to aerosol data exhibit the pollution fluxes to water surface over the whole Baikal area. AAE marks the impact of coal combustion, residential biomass burning, and wildfires indicating the high pollution level of the Lake Baikal ecological system .

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https://doi.org/10.1016/j.jes.2021.01.011

Introduction

Aerosols play a key role in atmospheric chemistry affecting the processes in the Earth atmosphere-hydrosphere and the complex feedback between climate forcing and environmental responses Andreae and Crutzen (1997). Since significant climate changes, there is an intense increasing of aerosol-related impacts in the climate - sensitive regions such as the Arctic and Siberia (Abbatt et al., 2019; Kulmala et al., 2011). The pervasive and rapid surface water warming and eutrophication signals the urgent need to incorporate climate impacts into vulnerability assessments of air and water quality for lakes (O'Reilly et al., 2015). Accumulation of the pollution substance in water and sediments of the largest lakes of the world requires the identification of sources of the aerosol deposition to the lake surface (Ma et al., 2018; Oyo-Ita et al., 2017) which includes interactions between aerosol residence times, removal processes, and regional and global environmental change (Wiman et al., 1990).

Laser sensing methods are among the most effective for detecting sources of air pollution and monitoring the transboundary aerosol transport; they allow studies of the aerosol spatial distribution over the entire height of the troposphere, with high temporal resolution Samoilova and Balin (2008). The characterization of near-surface particulate matter (PM) mass and number size distribution linked with air mass transport makes it possible to identify the pollution sources and assess the consequences of emissions on the total environment (Tunved et al., 2013). In smoke plumes originated from wildfires the major growth in PM concentrations is caused by submicrometer particles (Saarnio et al., 2010). Transported fire plumes enhance the fine-mode aerosols whereas the coarse-mode aerosols remain unaffected (Ghosh et al., 2019).

Combustion is recognized as an important source of radiatively and chemically active aerosols containing black carbon (BC) which interact directly and indirectly with the Earth's radiation energy balance, cloudiness, and subsequently affect the regional and global climate (Bond et al., 2013). BC is the most important pollution contributor originating from fossil fuel (FF) combustion (transport, industry) and biomass burning (BB) (residential and wildfires) (Chen et al., 2009; Liu et al., 2018; Mousavi et al., 2018). It is widely variable from the background in pristine Arctic regions (Eleftheriadis et al., 2004; Popovicheva et al., 2019a) up to the large level in big cities of European part of Russia and China (Golitsyn et al., 2015; Popovicheva et al., 2020b), and Siberia (Kozlov et al., 2016). BC is mainly present in aerosols in urban environment, thus it concerns air quality and population health (Janssen et al., 2011).

Quantification of BC and brown carbon (BrC) in various source emissions is critical for assessment of combustion aerosol impact (Olson et al., 2015). Light absorption by atmospheric particulates emitted from FF combustion sources exhibits the relatively weak wavelength dependence with the Absorption Angstrom Exponent (AAE) close to 1 (Massabò et al., 2015), indicating that BC is the dominant absorbing aerosol component while BB aerosols are distinguished by the stronger wavelength dependency (Kirchstetter et al., 2004; Popovicheva et al., 2019b; Popovicheva et al., 2017b).

Lake Baikal is the deepest (1637 m) and oldest (25 million years) lake in the world; 70% of the species are endemics. Lake Baikal contains 20% of the world's unfrozen surface freshwater and the source of drinking water for villages around the lake. In 1996 Lake Baikal has been declared a UNESCO World Heritage Site. Whatever, the climate change negatively affects the atmosphere, precipitation, ice season and the temperature of lake surface water Shimaraev and Starygina (2010). The tendency of climate warming with the trend of annual air temperature of +1.3 °C in 100 years twice higher than the trend of global temperature of +0.6 °. Surface waters warmed 2.0 °C during 20 years; physical and biological changes associated with warming and eutrophication have been proved occurring in Lake Baikal (Izmest'eva et al., 2016).

Two decades ago the Baikal area was considered as a remote location in east Siberia, far away from industrial centers, and particles properties were studied as of remote continental aerosols (Bashurova et al., 1992). However, a number of studies of physical properties and selected chemical composition of aerosols in the Baikal region carried out during the last years had emphasized the significant role of the atmosphere pollution in the Lake Baikal ecosystem (Golobokova et al., 2018). Long-range transport of sulphur and nitrogen oxides from large coal power plants had caused extra sulphate and nitrate deposition and acidification of river waters in the Baikal region (Obolkin et al., 2016). It was found that while biogenic coast aerosols (> 2 µm) compose up to 60–80% of total volume (Matthias-Maser et al., 2000), the anthropogenic impact on submicron particles is of major concern.

In order to identify the pollution sources for the receptor region, the complementary air mass trajectories and chemical composition analyses are widely used: in urban areas (Manousakas et al., 2018; Popovicheva et al., 2020a), in remote Arctic and Siberian areas (Chang et al., 2011; Mikhailov et al., 2017). Morphological and elemental composition of individual particles in the Lake Baikal atmosphere have revealed the significant differences between the pristine northern and anthropogenically - influenced southern basin according to the impact of the long distance transport of polluted emissions from industrial centers of Irkutsk and Ulan-Ude (Van Malderen et al., 1996). Influence of the orographic features of the lake area on the formation of longitudinal and transverse sections of the altitude structure of aerosol fields was established in spatial distribution lidar studies of the atmospheric aerosols over Lake Baikal (Balin et al., 2007).

Wildfires is a major source of climate-relevant species emitted at northern latitudes (Lavoué et al., 2000). Their emissions are increased due to climate change, particularly during wildfires in the boreal forests of North America and Russia (de Groot et al., 2013; Jaffe et al., 2020; Konovalov et al., 2011) . Black carbon spatiotemporal variability for last two decades in the West Siberia and Russian Subarctic has revealed a strong impact of forest fires in the warm season (Kozlov et al., 2016), especially during extreme smoke haze events (Kozlov et al., 2014). Simulations of Siberian biomass burning in a chamber studies and natural prescribed burns showed the significant impact of the combustion phases (flaming & smoldering) on carbonaceous constituents (Cahill et al., 2008; Kalogridis et al., 2018). 70% of the Baikal region territory is covered by forest which is the greatest source of natural aerosols responsible for the maintenance of the stable atmospheric composition. Spatiotemporal structure of vertical aerosol fields formed as a result of forest fires near the lake shows the strong difference from a long away distance (Balin et al., 2016) .

Since the question of recent global changes in biocenosis and massive death of endemic species in the littoral zone of Baikal is unsolved to date (Timoshkin et al., 2016), the search of potential sources for increasing anthropogenic load is remaining to be continuously actual. Presently, a rising number of tourists and shipping is assumed negatively affected the Lake Baikal ecosystem, leading to multiplied amount of phosphorus and nitrogen compounds entering the lake (Kravtsova et al., 2014).

Dry deposition is an important process affecting the lifetime and spatial distributions of atmospheric aerosols (Wu et al., 2018). Water quality became at risk because of the direct deposition of combustion particulate matter from the atmosphere (Bazhenov et al., 2007) which is remaining not well quantified. Improved estimates for aerosol dry deposition on natural water are needed for the calculation of pollution fluxes to the Baikal lake.

The study of this article is focused on comprehensive observations of spatiotemporal aerosol distributions during the purposely designed expedition over Lake Baikal. A number of instrumentations for scattering and absorption near-surface and tropospheric measurements were installed onboard the research vessel cruising over the lake. They provided the distributions of aerosol tropospheric fields, particle number sizes and masses, and black carbon. Combined with meteorological data and simulations of air mass transportation to the sampling point, the highest PM_{10} mass, fine particle number, and BC concentrations are associated to industrial/wildfire emission polluted regions and touristic areas of intensive shipping and residential biomass burning. Absorption Angstrom Exponent (AAE) acts as a measure of spectral aerosol absorption, its high level identifies the coal combustion and biomass burning. Assessment of the aerosol deposition to water surfaces contributes to the determination of the impact which aerosols might have on sensitive Lake Baikal ecosystem.

1. Experimental

1.1. *Lake Baikal's region*

Lake Baikal is located in the south of eastern Siberia, Russia, close to the Mongolian border, see a map in Fig. 1. The lake is 636 km long and from 25 to 79.5 km wide, its reservoir is shaped as a half-moon in a mountain hollow. There are over 500 rivers discharging into Lake Baikal, of which the three major ones are the Upper Angara in the north, and Barguzin and Selenga rivers on the eastern lake shore. The vegetation in the lake surrounding is predominated by forests of coniferous (pine, cedar) and deciduous trees (birch, aspen).

Irkutsk region with well - developed industrial structure takes place west and northwest of the lake (Fig. 1). Detailed description of the Baikal region is presented in Supplementary Material. Irkutsk, Bratsk, and Angarsk are cities of highest particulate and gaseous emissions, as marked in Fig. 1. Vehicle exhaust emissions significantly impact, up to 50% of the total pollutions. Republic of Buryatia locates on the east and south-east of the lake, with Gusinoozersk and Ulan-Ude cities of biggest industrial developing (Fig. 1).

In July 2018, 214 fires with a total area of more than 760 thousand hectares and 89 fires with a total area of more than 1.3 million hectares were registered in Krasnoyarsk Region and the Republic of Saha (Yakutia) as reported by ISDM-Rosleskhoz FBU "Airlines Protection" (https://aviales.ru/).

1.2. *Expedition route over Lake Baikal*

The expedition was carried out onboard the research vessel “Akademik V. A. Koptuk” with displacement of 400 tons. The ship has main and auxiliary (diesel generator) engines of 230–240 and 80–100 rpm, respectively, operated with diesel fuel. The map of the ship route track over the water area of Lake Baikal and the parking places are shown in Fig. 1. Aerosol samples were collected during the whole expedition; sampling durations and routes corresponding to the sample collections are presented in Table S1. The description of the expedition route is presented in Supplementary Material.

1.3. *Meteorology and air mass transportation*

Analysis of the synoptic situation during expedition was carried out according to high-altitude synoptic maps (http://www.aari.ru/). Meteorological data (temperature, wind speed and apparent direction, precipitation) during the cruise were obtained from the shore meteorological stations taking place at the shorten distance from a ship route. Ship course was recorded by GPS. Real wind direction was estimated from the aforementioned data. Calm weather was purposely under attention because could lead to the aerosol pollution by the ship pipe exhaust.

The lake is surrounded by mountains reaching to maximum heights of 1500 m above the water level of the lake, which lies itself some 500 m above sea level (a.s.l.). In the southwestern part of the lake, the mountains are only about 500 m high above the lake surface. It makes the southern part of the lake open to air masses coming from the west. Moreover, the most common wind direction in this region is southwestern that carries anthropogenic emissions from Irkutsk industrial region. The movement of near-surface winds is strongly influenced by local conditions; the valleys of the bigger rivers provide possibilities for local air circulation can be also originated from temperature differences between air masses above the lake and over the surrounding land.

Fig. 1 – Map of Lake Baikal, Eastern Siberia, Russia. The expedition route track over Lake Baikal between places of ship parking on a coast is indicated by lines. Samples collected during a corresponding part of a route are shown by numbers, described in Table S1. Major industrial areas are indicated, with source emissions in tons per years according to official emission inventories. Insert is a research ship "Akademik Kovtuk".

To evaluate the air mass transportation the backward trajectories (BWT) were generated using NOAA HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model of the Air Resources Laboratory (ARL) (Stein et al., 2015) with coordinate resolution equal to $1^\circ \times 1^\circ$ of latitude and longitude. BWT were calculated for start and end times of sample collections, shown in Table S1. The potential source areas were investigated using 2-day backward trajectories, except those BWTs which transported from wildfire areas (p18,19, 20 and 22 in Fig. 1). For last ones we calculated 5 days ago in order clearly identify the wildfire impact. Air masses arrived at 250, 500, and 1000 m heights (a.s.l.) were simulated. There were no any reasonable differences between origin directions with respect the height, therefore further we report the results only for 500 m a.s.l. Fire information was obtained from Fire Resource Management System (Scanex) http://www.scanex.ru/cloud/karta-pozharov/. Daily maps were related to the computed trajectories, providing a clear picture of the geographical location of fires, with the several kilometer resolutions.

1.4. *Aerosol real time measurements and sampling*

Aerosol Raman polarization lidar LOSA-A2 was mounted on the back deck of the ship where the vertical sounding could be supplied. LOSA-A2 was custom made at the Institute of Atmospheric Optics, RAS (Tomsk) to conduct expeditionary studies of tropospheric aerosol fields, its working principles are described elsewhere (Nasonov et al., 2020). More detailed description of atmospheric sounding performed is presented in Supplementary material.

To determine the height of PBL and ML, a gradient method was used. It is based on higher aerosol concentrations in ML then in FA. Because air masses in ML and FA are hardly mixed, the interface between them is marked by a rapid decrease of the backscattering intensity. Assuming a nearly constant aerosol concentration in the boundary layer and a low aerosol concentration in the free atmosphere, the maximum of the vertical gradient determines the high of the ML.

Particle counter Handheld 3016-IAQ was used for measurements from the ship board opposite to the exhaust plume direction. It recorded the particles scattering and converted it to a particles number, meeting ISO 21501-4 calibration with NIST traceable PSL spheres. Particle size distribution was determined over six size (0.3, 0.5, 1.0, 2.5, 5.0, 10.0 μm) channels with the registration efficiency not lower than 50% and 100% for particles of radius > 0.2 μm and > 0.3 μm, respectively. Differential particle count data summarized over six channels were used for an estimation of particulate matter mass less than 10 µm (PM_{10}). The major constituents of aerosol in remote areas are organics, ammonium sulfates and nitrates, with sulfates to organics ratio 2:1 Seinfeld and Pandis (1998). Ammonium sulfate density is 1.76 g/cm^3, while organic aerosol density is 1.4 g/cm^3 (Vratolis et al., 2018). We also expect a large portion of the atmospheric aerosol in this remote area to be mineral dust with a density of 1.6 g/cm^3 as reported by (Kandler et al., 2007). Therefore, we assume that an appropriate aerosol density in the Baikal remote area is approximately 1.6 g/cm^3, an average over major constituent composition. During strong precipitation the measurements by a counter was not performed.

For the particulate sampling the proper precautionary measures were taken in order to prevent the possible contamination from potential sources such as the ship exhaust and lake water droplets. Therefore, the instruments which perform the filter sampling (aethalometer and sampling system) were installed at the upper deck of the ship rostrum as the cleanest location on the vessel. Aerosol equivalent black carbon (eBC) concentrations were determined using the aethalometer designed in Moscow State University (MSU) (Moscow) and Central Aerological Observatory (CAO) (Moscow Region) purposely for mobile campaigns and was used in (Popovicheva et al., 2017a; Popovicheva et al., 2017b). In this instrument, the light attenuation caused by the particles depositing on a quartz fiber filter is measured at three wavelengths (450, 550, and 650 nm). eBC concentrations were determined continuously by converting the time-resolved light attenuation to mass using 650 nm wavelength. The calibration parameter was derived during parallel long-term measurements against an AE33 aethalometer (Magee Scientific), that operates at seven wavelengths, three of them identical with the MSU unit. Data analysis from this AE33 calibration experiment was adjusted to provide the scheme of calculation for the absorption coefficient, as described elsewhere (Manousakas et al., 2020). The data retrieved from the aethalometer were postprocessed in order to remove outlier values. A value is considered as an outlier if it exceeds its previous value in the time-series by more than 3 times the standard deviation of the last 10 values. With this approach, extremely high concentrations that do not follow the general trend and can be attributed to local contamination (ship exhaust) were considered separately. If the direction of the apparent wind was blowing from the back towards the sampling site, we removed these extremely high concentrations from the database.

Moreover, the filter contamination might have occurred when the episodes of calm wind were happening, therefore these measurements are carefully considered in the discussion. Samples were collected at a flow-rate of 30 L/min using a low volume sampler loaded with 47 mm in diameter Whatman quartz microfiber filters (QM/A).

1.5. *Aerosol deposition*

The dry deposition velocity (V_d) depends on the size and density of particles as well as on meteorological conditions and the underlying surface character Slinn and Slinn (1980). In regional chemistry and aerosol transport models, the empirical parameterization of V_d in several dry deposition schemes is suggested which can vary over 2 orders of magnitude (Wu et al., 2018). Estimates for dry deposition account for the particle growth by water vapor condensation in the neighborhood of water surface which depends on the particle hygroscopicity. The extend of aerosol hygroscopicity is determined by the particle composition. Thus, freshly emitted combustion - produced BC is hydrophobic (Popovicheva et al., 2010) while ambient aerosols dominated by water-soluble substance like ammonium sulfates $(NH_4)_2SO_4$ are hygroscopic.

In order to calculate V_d for the aerosol mass deposited on water surface of Lake Baikal, the volume particle size distribution is acquired from the measured particle number size distribution data. Then, the geometric mean diameter (D_g) of the volume particle size distribution is computed for each measurement. Calculations of the deposition velocity for each D_g are following to Slinn and Slinn (1980), assuming a wind velocity from 1 to 5 m/s, in the range measured over the expedition route. Fluxes of deposited aerosols from the atmosphere on the underlying surface is determined as the product of their concentrations by the deposition velocity over an interval of observed time.

PM mass concentrations in the size distribution up to 10 μm are used for total fluxes of aerosol particles assuming they are dominated by $(NH_4)_2SO_4$. Solid particles of carbonates of alkali and alkaline-earth metals are widely distributed in the remote area of the Baikal region (Yermakov et al., 2007). In the humidified atmosphere the solid aerosols are involved into chemical heterogeneous reactions with gaseous NH_3 and H_2S0_4 through the stage of watering. As the result, the composition is changed to liquid $H_20/H_2S0_4/(NH_4)_2S0_4$. The proposed mechanism is in well correlation with chemical composition measurements (Golobokova et al., 2018). To emphases purposely the pollution from combustion sources, the fluxes for hydrophobic submicron black carbon aerosols are calculated. Additionally, to address the impact of particle nature and size on the deposition change, we calculate the hygroscopic submicron aerosol particles consisting of $(NH_4)_2SO_4$ as well.

1.6. *Analytic chemistry*

Off-line examination of light attenuation of particles deposited on quartz filter samples was performed using the multiple wavelength light transmission instrument (transmissometer) based on the methodology of Kirchstetter et al. (2004) and applied in (Popovicheva et al., 2019b; Popovicheva et al., 2017b) The intensity of light transmitted through quartz filters was measured at seven wavelengths from the near-ultraviolet to near-infrared spectral region. The dependence of the attenuation (ATN) on the wavelength λ is parameterized using a power law relationship:

$$\mathrm{ATN} = k\lambda^{-\mathrm{AAE}}, \tag{1}$$

where, the Absorption Angstrom Exponent (AAE) is a measure of strength of the spectral variation of aerosol light absorption. For a number of samples, the spectral absorption could not be measured because too low aerosol loading.

2. Results and discussion

2.1. *Tropospheric aerosol fields*

Meteorological situation and air mass transportation from remote source regions to the place of the sampling influenced the aerosol composition and its change during the ship route. Observed meteorological conditions such as temperature, wind direction, velocity, and precipitation are shown in Fig. S1. Air mass backward trajectories (BWT) arriving to the coast location of the sample collection during the a) southern and b) central and northern parts of the expedition route are plotted in Fig. 2.

At the beginning of the expedition from 15th to 17th July the Baikal region was under the influence of a powerful cyclone formed over Taimyr, the southern periphery of which was located in the area of Lake Hubsugul. This cyclone determined the rainy and cloudy weather; low cloud cover (1.0–2.5 km) was observed by lidar sounding. Since 18th July, the southern periphery of the cyclone has shifted to the west, the precipitation has stopped, and ground air flows have changed to the west and south-west direction.

On 18th and 19th July the sunny calm weather was observed. During these days the vertical structure of the aerosol field was typical for the background atmosphere, as illustrated in Fig. 3a for 19 July. At this time, the ship route was along the western shore of the lake from Listvyanka to the northwest, towards Olkhon Island. The planetary boundary layer (PBL) was observed below 2 km and the mixing layer (ML) of up to 1 km. Small values of the scattering ratio in Fig. 3a indicate the low aerosol loading, mainly in the ML. In the morning of 19th July a weak aerosol layer was observed because the route was closed to the touristic and navigable areas. Since the aerosol layers were unstable, during day warm up they began to stir. Thus, by the evening the ML was divided into 2 layers: the first one up to the height of 200–300 m and the second one up to the height of 400–600 m. Estimation of the total aerosol optical depth on 19th July according to the NAAPS model (http://www.nrlmry.navy.mil/aerosol/) shows the optical depth (τ) ˜ 0.1 which correspond to the average ones for the background atmosphere in summer.

In following days from 19th to 21th July, due to the reverse displacement of the southeast branch of the cyclone, the unstable cloud windy weather with precipitation and low daily temperatures was established. From 21st July air masses were transported from wildfire areas in the Krasnoyarsk and Yakutia regions. On the route Nizhneangarsk-Severobaikalsk in the night of 21 July the thin aerosol layer was started to be observed (Fig. 3b), the aerosol content in the atmosphere 2-3 times exceeded the background. At around 1:30 a.m., the aerosol layer at a height of 2–3 km began to separate on 200–300 m parts which subsequently expanded vertically, filling the atmosphere up to a height of 3.5 km. With a delay of about 1.5 h other aerosol layer at an altitude of 1.5 km was observed. At that time, the atmosphere optical depth was found high, τ ˜ 0.4–0.8, that is well related to wildfire smoke. Observation of the second aerosol layer in Fig. 3b may be explained by the smoke plume transportation above the ridge of 1.9–2.2 km height on the western shore of the lake.

On 22th July, the ship departed from Severobaikalsk towards the eastern shore of the lake and moved along it to the south. Smoke haze from the north had spread under the influence of local air flows gradually filling the basin. On 23th July when the ship passed along the Central Baikal near the Ushkany Islands, the aerosol layer nonuniformly filled the boundary layer to an altitude of ˜ 2 km, another weak aerosol layer was observed at an altitude of 3–3.5 km (Fig. 3c). Only in the evening of 24 July, the atmospheric loading began to decrease. On the night from 24th to 25 July Southern Baikal was affected by the cyclone which brought cloudy and rainy weather, background aerosol content in the atmosphere was again observed (Fig. 3d).

2.2. *Aerosol size, PM10 and black carbon mass*

Studies of the aerosol size distribution carried out in the Baikal region have showed the size range from a few nanometres to about 30 mM (Koutsenogii et al., 1993) while submicron particles were dominated (Van Malderen et al., 1996). Three types of the size distribution represented particles at different evolution stages and condensation nuclei formation (Bashurova et al., 1992). Significantly inhomogeneous distribution of the particle number density for both submicron and micron fraction was observed over the total area of Lake Baikal (Zagaynov et al., 1990).

Number particle concentrations as a function of size and periods of sampling obtained along the expedition route is shown in Fig. 4. Total mass of particles with a size less than 10 µm (PM_{10}) is estimated from the number concentrations integrated over the size. 1 h average PM10 mass and eBC concentrations along the expedition are shown in Fig. 5. Since calm weather can lead to additional pollution by the plume ship exhaust, the calm episode time durations are indicated in the same figure. There is no observed correlation for eBC concentrations with calm weather periods, however we should consider this possibility. Fig. 6 presents the time series of PM_{10} and eBC concentrations averaged over sampling periods along expedition route. In order to relate to geographical locations of sampling, we purposely show the spatial distribution of eBC mass concentrations averaged over sampling periods in Fig. 7.

2.2.1. *Southern Baikal*

On 15th July, air masses were originated over the less populated south-eastern region of the Republic of Buryatia. PM_{10} mass concentrations for aerosols collected in the habour Listvyanka were varied from 3 µg/m^3 in early morning up to 24 µg/m^3 at daytime, a typical trend for local residential area. On averaged PM_{10} mass was observed at the level of 9 µg/m^3. Fraction of fine aerosols less than 0.5 µm dominated at the average number concentration near 10^8 m^{-3}.

During the following day of 16 July the Baukal region was under influence of a low-gradient field of atmospheric pressure characterized by weak local winds in various directions. The Hamar Daban Range mountain framing the lake southern had part partially prevented the long- range air mass transport. On the Listvyanka-Tankhoi route the avareage number concentration of fine submicron particles and PM10 mass was decreased, the average eBC concentration was found of 1.7 µg/m^3. While the following Tankhoi-Kultuk route is characterized by the high BC variability according the wind direction. This case study is shown in Fig. S2.

Since 17th July morning, the meteorological situation over the South Baikal area was changed: calm weather was replaced by north-west wind. Until the morning of 18 July air masses were transported to Southern Baikal from the Krasnoyarsk region, they have passed the areas of forest fires and big industrial Bratsk and Sayansk cities in the Angarya River

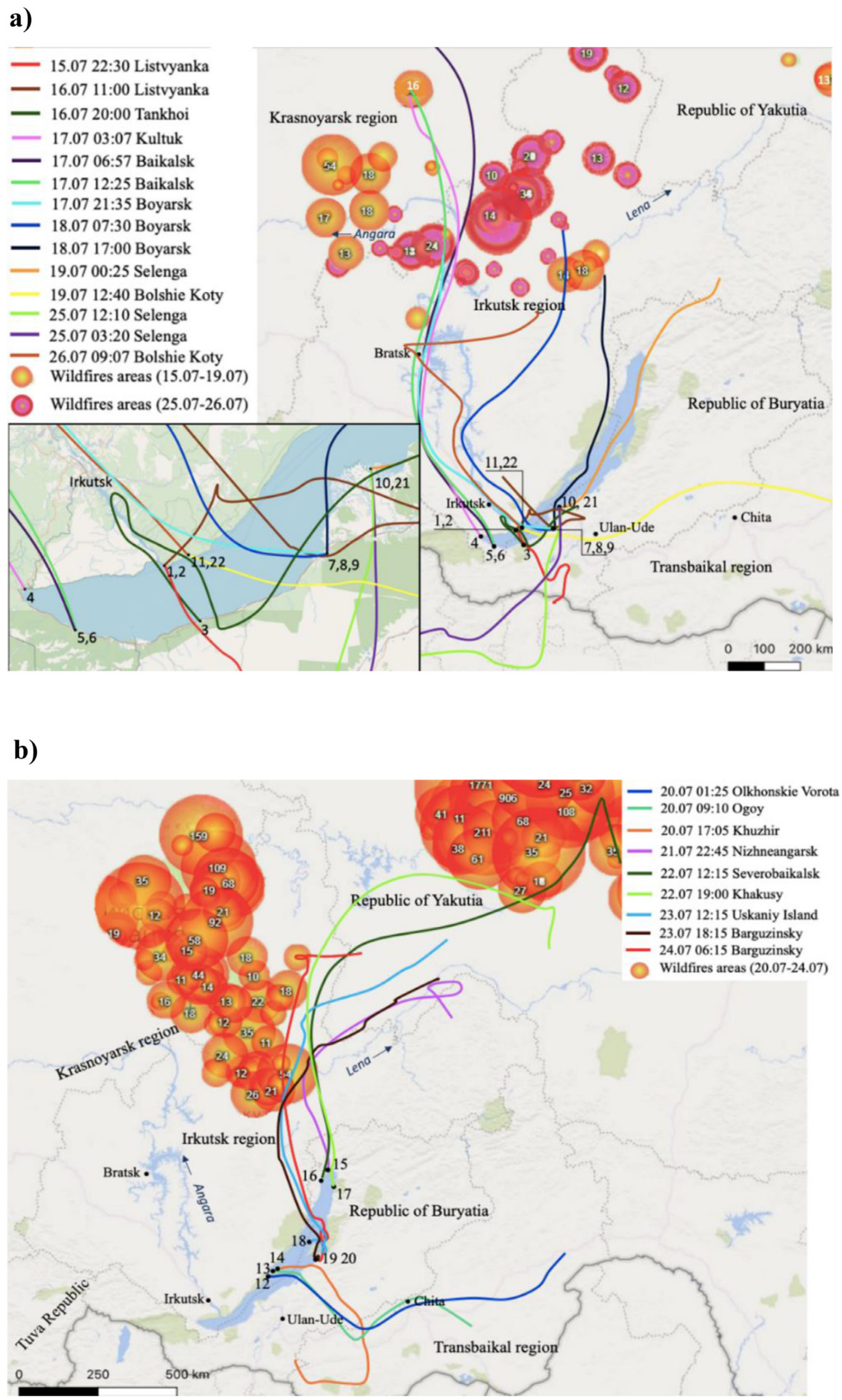

Fig. 2 – Air mass backward trajectories arriving to the location of start and end of sample collection at times indicated in the left top insert, during a) the southern and b) central and northern parts of the expedition route. The numbers indicate the sampling start on the coast, described in Table 1. Wildfire areas for corresponding periods are shown in left bottom insert from Scanex Fire Resource Management System. Numbers inside fire areas indicate the amount of ignitions.

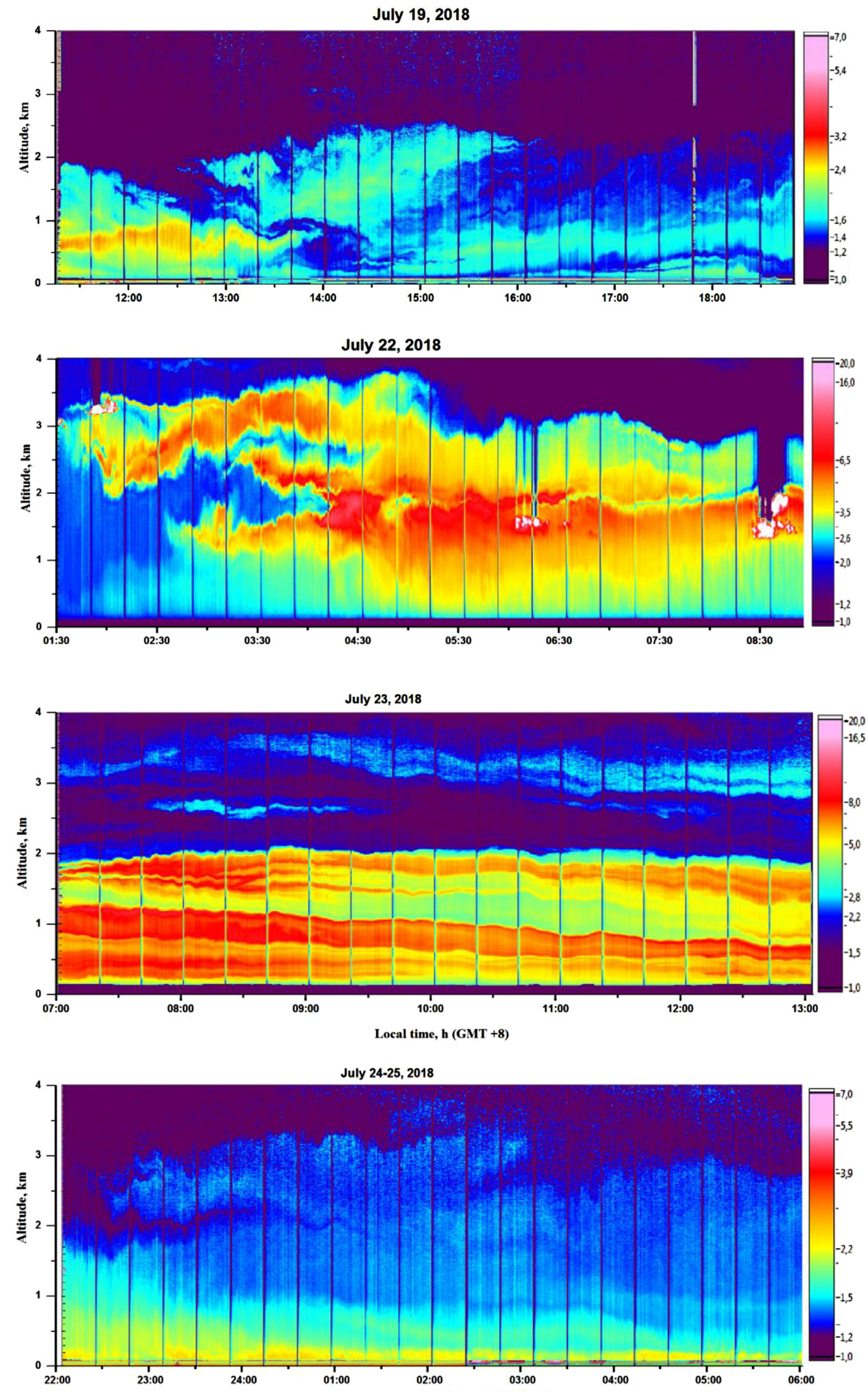

Fig. 3 – Spatiotemporal structure of aerosol fields on the route a) from Listvyanka towards Olkhon Island in the background troposphere on 19.08, b) from Nizhneangarsk to Severobaikalsk in wildfire plume on 21-22.08, c) along the Central Baikal near Ushkany Islands the aerosol layer nonuniformly filled the boundary layer on 23.07, and d) in Southern Baikal in cloudy and background atmosphere on 24.07 and 25.07. The color scale (from purple to red) corresponds to the scattering ratio R (532 nm).

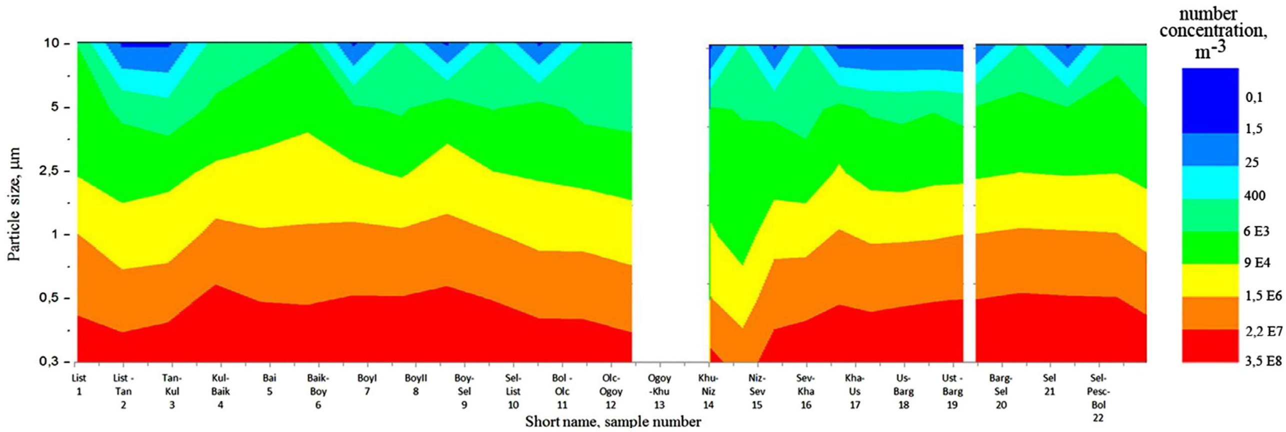

Fig. 4 – Spectral plot of particle number concentrations as a function of size and periods of sampling along the expedition route.

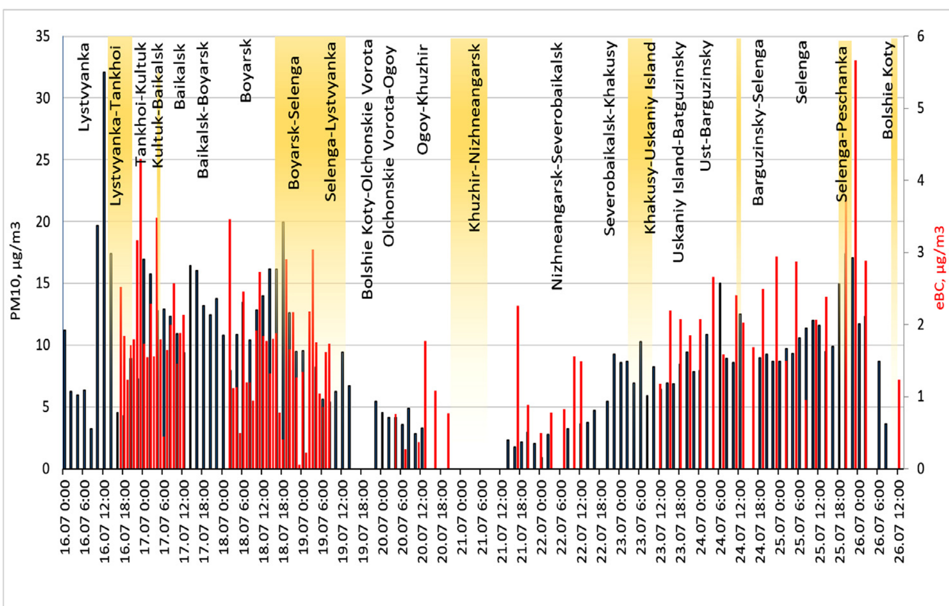

Fig. 5 – 1h average PM_{10} mass and eBC concentrations along the expedition. Calm episodes are indicated by shadows.

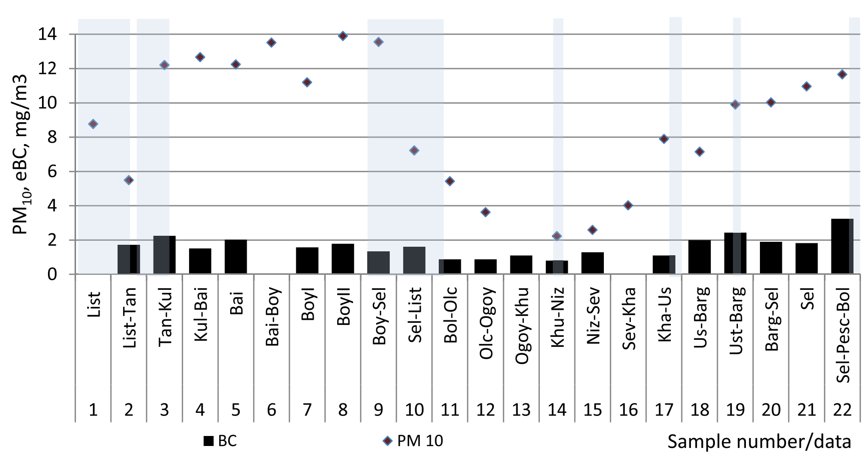

Fig. 6 – PM10 and eBC mass concentrations averaged per sampling periods along the expedition route. Sample numbers and names are presented according to Table 1. Calm episodes are indicated by shadows.

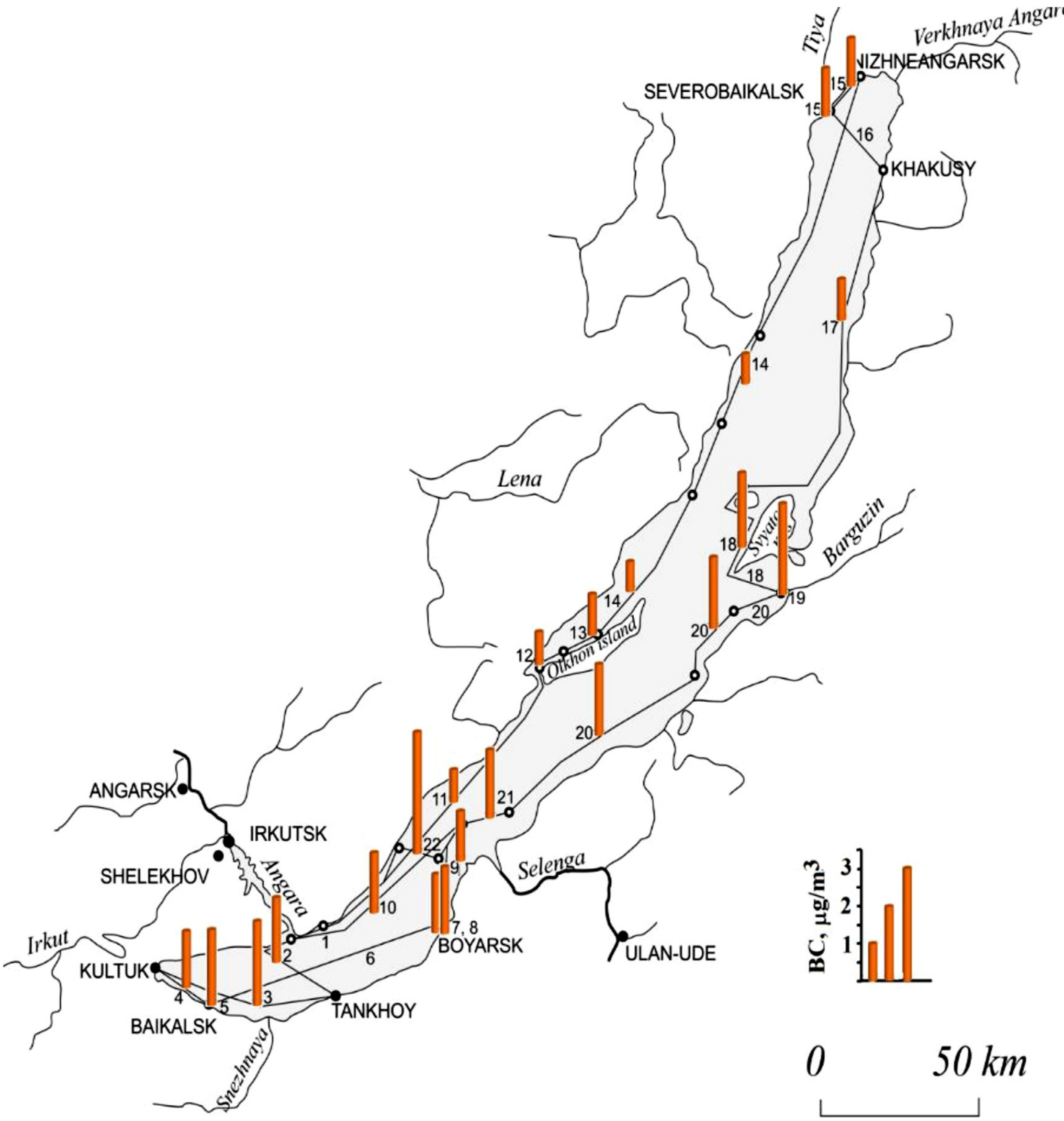

Fig. 7 – Spatial distribution of eBC averaged over sampling periods, µg/m^3. Samples numbers correspond to Fig. 1 and described in Table 1.

valley. On the following Kultuk-Baikalsk-Boyarsk route PM_{10} mass was increased at the beginning and then observed at the high level, on average 12 µg/m^3. Average concentrations of eBC were high, of 1.8 µg/m^3. And the number concentration for submicron particles were found similar as in Listvianka, near 10^8 m^{-3}. The longest dust event related to the number concentration of the course ˜ 10 µm particles up to $6{\cdot}10^3$ m^{-3} was observed on this route. We should note here that the South Baikal area is distinqueshed by unique ladle like orographic features. It is characterized by unique microclimate because of the closed relief. Thus, air mass flows through the Angara river valley bring the intensive pollution to water area.

Since 18th July morning the southern periphery of the cyclone has shifted to the west, the precipitation has stopped, and ground air flows have changed to the west and southwest direction. During the parking near Boyarsk and then on the Boyrsk - Selenga route air masses of few days aged were arrived, affected by wildfires in the Irkutsk region. They impacted PM_{10} and variable BC mass conventrations, on average 14 and 1.3 µg/m^3, respectively. The longest episode of the minimum hourly averaged BC around 0.1 µg/m^3 was observed on this route, exactly at night of 19 July. Significant change in air mass transportation from remote low-populated region of the Republic of Buryatia was observed on 19th July when the route over the southern part of Baikal was turned back to Listvynka, there PM_{10} mass were decreased down 7.2 µg/m^3.

2.2.2. *Central and Northern Baikal*

Since 19th July, the route took place from Listvianka toward to northern Baikal along the east coast of the lake that is the reserved and pristine region. Until Nizhneangarsk city which was approached in the evening of 21st July, the transportation of air mass was occurred from the east, from residential areas of the Republic of Buryatia and Transbaikal region, where they passed industrial cities namely Ulan-Ude, Selenginsk and Chita. However, rains and fogs in this time cleaned the atmosphere. On Bolshie Koty-Olchonskie Vorota-Ogoy route the number concentration of fine submicron particles and PM_{10} mass was decreased more than one order of magnitude, down to 3.5 µg/m^3, respectively. With respect to other areas, these characteristics can represent the background near-surface atmosphere. The route from Olchonskie Vorota to Ogoy and Khuzhir took place over Maloe more, it is the internal sea between the west coast of the lake and the coast of island Olhon. During this route BC was found at its minimal level around 0.27 µg/m^3, however it was increased up to 1.3 µg/m^3 during the parking near Khuzhir settlement, being affected by populated touristic place emissions. Then, until Nizhneangarsk city the lowest total number particles were observed. Thus, the lowest PM_{10} mass and BC, on average 2.2 and of 0.74 µg/m^3 was found.

However, in Nizneangarsk city the meteorological situation was significant changed; air masses started to arrive from north, from the Krasnoyarsk region where intensive wildfires were detected at this time. During next two days of 22, 23, and 24 July Severobaikalsk city, Khakusy, Uhkaniy islands and Barquzinsky bay were affected by remote and close fires in Northern Yakutia and Krasnoyarsk region, respectevely. On Nizhneangarsk -Severobaikalsk route PM10 mass started to increase to 2.6 µg/m^3. The increased BC episodes were happening then wildfire plumes could transport above the ridge on the lake shore. Figure S2 shows the case of high BC up to 2 µg/m^3 following the lowest one of 0.1 µg/m^3 .

The route from Severobaikalsk towards the eastern shore was characterized by the continuous increasing of PM_{10} mass. In days from 21st to 24th July, on the Nizhneangarsk - Barguzinsky route of the biggest impact of wildfires plumes from Republic of Yakutia and Irkutsk region, PM_{10} mass approached to 8 µg/m^3. This particulate loading 4 times exceeds one in the background atmosphere, well according to the smoke affected - aerosol field tropospheric structure observations. One can again observe the dominant abundance of the fine particle number concentration and increase of the averaged BC concentration up to 2 µg/m^3, approaching maximum of 2.4 µg/m^3 in the Barguzinsky gulf. On 24th July the Northern Baikal was dominated by weak local winds, at the same time it was possible to observe air flows coming from the north through the lake. The atmosphere was dominated by local circulation at that time.

On 25th July the Selenga river delta was affected by air masses from the south and southwest which were originated over desert and semi-desert areas of Mongolia. They could be a source of dust relating to PM10 mass increase up to 11 µg/m^3 and number of course particles up to 6•10^3 m^{-3}. On the Selenga - Bolshie Koty route the long parking in the Peschanka bay, the famous touristic and shipping activity place, took place. PM_{10} and BC concentrations approached the maximum, 12 and 3.2 µg/m^3, respectively. Finally, on 26th July when the cruise was finalized, northwest flows carrying air masses from industrial sources and wildfires of the Irkutsk region were repeated.

2.2.3. *Deposition fluxes*

The assessment of the environmental damage of Baikal's ecosystem, resulted from industrial emissions, touristic activities, and shipping, and related to long - range air mass transport and local impacts can be valuable from estimates of pollution deposition fluxes on water surface. Fig. 8 shows the fluxes for three types of particles: total PM_{10} composed from $(NH_4)_2SO_4$ for the case of deposition with relative humidity near 100% in the deposition layer, submicron $(NH_4)_2SO_4$ particles that grow to the equilibrium size exposed to a relative humidity of 99%, and hydrophobic BC submicron particles. Deposition of total PM_{10} mass have more than 2 orders of magnitude in comparison with submicron particles of the same (ammonium sulfates) nature due to a huge impact of coarse particles. Impact of hydroscopicity is significant: fluxes for submicron BC decrease twice in comparison with hygroscopic $(NH_4)_2SO_4$ particles. Increase of fluxes with the wind velocity, varied between 1 and 5 m/s, is found significant, especially in the case of BC, up to 3 times.

2.3. *Combustion sources and level of pollution*

Black carbon (BC) produced by high-temperature FF combustion sources fit within the Rayleigh scattering regime for near-visible wavelengths with theoretical λ^{-1} relationship relating to AAE equal 1. Brown Carbon (BrC) contributes significantly to the measured light absorption by increased AAE, showing it about 2.5 for wood smoke (Kirchstetter et al., 2004). During high regional biomass smoke its spectral features become pronounced, being an effective indication of BB as the major regional source for the haze at the molecular level (Popovicheva et al., 2017b). AAE value for coal combustion approaches 4.5 (Olson et al., 2015). In the residential emission inventory of Asian regions 15–18% of the total particle light absorption at ultra-violet wavelengths attribute to BrC from residential coal burning (Tian et al., 2019). Observations performed in urban environment indicate the impact of BB on urban aerosols, the AAE value above 1.3 is suggested to identify the periods most affected by BB (Diapouli et al., 2017). Thus, AAE acts as an optical marker for parametrization of FF and BB - affected periods (Popovicheva et al., 2020a) .

Episodes of the BC pollution can be identified by the increased ratio of BC to PM_{10} (BC/PM_{10}). In urban environment, this ratio is higher in traffic source emissions than that from other sources: in heavy-duty diesel source testing it was 0.77 while the lowest ratio was observed in residential wood combustion (0.095) (Chow et al., 2011). In urban environment the low BC/PM_{10} ratio suggested that the pollution episode was mainly influenced by solid fuel sources (e.g., coal and biomass burning) while the higher ratio in clean days indicated a predominant influence by traffic sources (Liu et al., 2018).

The spectral dependence of the Baikal aerosol light absorption is well approximated by a power law Eq. (2), providing the estimate for AAE shown in Fig. 9. Variation of AAE values during the whole sampling period exhibits the range from 0.9 to 3.5. Combining the obtained AAE with the origin of air mass arrived during sampling periods, and assuming data for PM10 mass and BC concentrations averaged over the same times, the potential sources influenced the obtained aerosol compo-

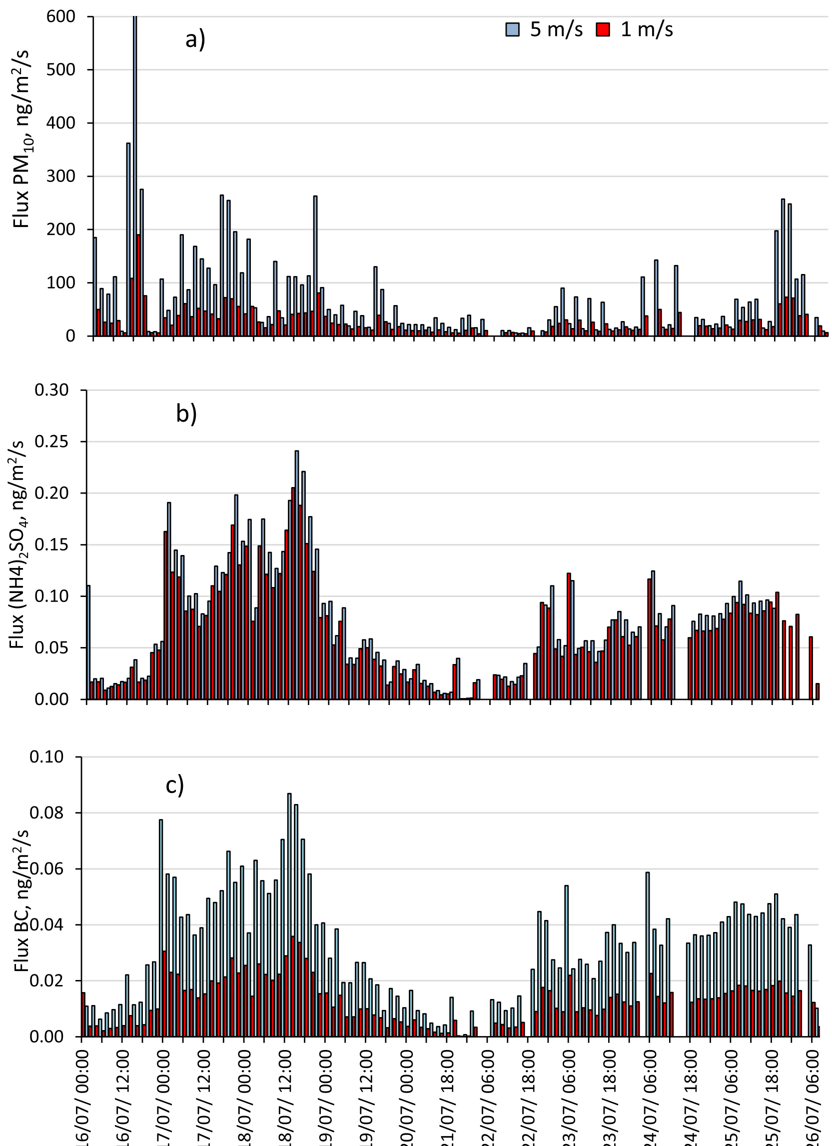

Fig. 8 – Fluxes of a) PM_{10} mass composed from $(NH4)_2SO_4$ for the case of deposition with relative humidity near 100%, b) submicron $(NH4)_2SO_4$ particles that grow to the equilibrium size exposed to a relative humidity of 99%, and c) hydrophobic BC submicron particle. Wind velocity are 1 and 5 m/s.

sition can be suggested. The list of suggested pollution sources is present in Table S1.

For aerosols sampled in the harbor Listvianka the AAE value around 1.8, typical for residential area influenced by local BB, was obtained. Because during this sampling period high PM mass and fine particle number concentrations were found while air masses originated from low - populated Buryatia area, it can be concluded that the harboar local residential sources including transport, shipping, and biomass burning had affected the aerosol composition at that time. High BC/PM10 ratio around 0.3 indicated the impact of intensive traffic near observation site. Low loading of the sample collected on the Lisvianka-Tankhoi route characterized by low PM mass could not allow to measure the spectral absorption, while high BC could be accumulated because calm weather happen. Thus, we can suggest that local BC source influenced the aerosol properties at that period.

Elevated AAE value up to 3.5 for Tankhoi-Kultuk route emphasizes coal burning as the pollution source identified by the high spectral absorption typical for lignite coal (Olson et al., 2015). This finding is confirmed by air mass transporation from industrial area of the Angara River valley and wind blowing from heat boilers in Baikalsk city. Then, during the whole route over Sourhern Baikal, high average PM mass and BC concentrations indicated industrial FF/coal combustion and BB, in correlation with air masses arrived from the Irkutsk region and affected by both wildfires and industrial emissions.The BC/PM10 ratio in the range 0.12–0.18 was typical for coal and BB sources. On the Baikalsk-Boyarsk route industrial FF was dominated because low AAE ˜ 0.8 was observed. During the

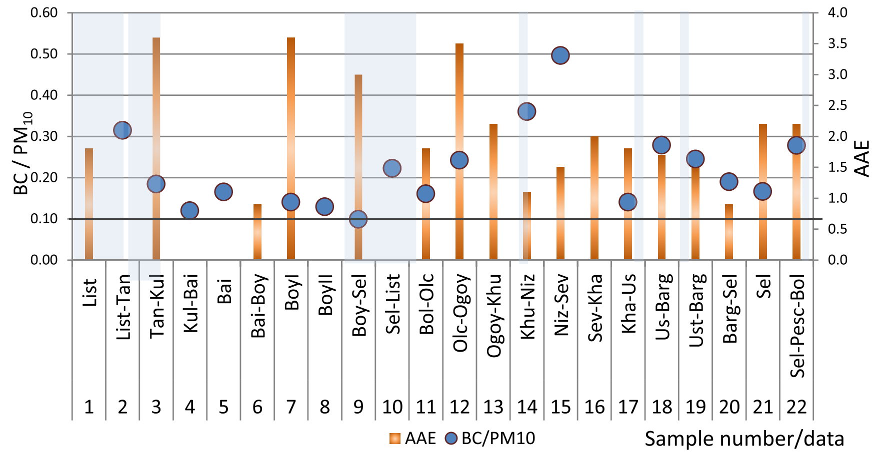

Fig. 9 – Absorption Angstrom Exponent (AAE) estimated as the strength of the spectral variation of aerosol light absorption from power law equation (2). Line of AAE equal 1 indicates high-temperature FF combustion sources fitting. Ratio of BC to PM_{10} mass concentration (BC/PM_{10}) averaged over sampling periods along expedition.

parking near Boyarsk and then on the Boyrsk - Selenga route high AAE values around 3 were observed, relating to coal burning and wildfires impacts.

On the route from the east lake coast to the west one, from Selenga to Listvianka, the significant change in the sources of pollution was happen, leading to significant, around twice, decreasing of PM_{10} mass and BC concentrations. Starting from that time and lasting until Severodvinsk city the period of the lower number for fine particles indicated much less impact of combustion processes. Air masses of east oridgin from Buriatia and Transbaikal regions brougt the residential BB plumes impacted the organic composition aerosols on the route until Olkhonskie Vorota that was indicated by the AAE value around 1.8.

The following route to Khuzhir took place in the region of Maloe more geografically and orograghically different from other water area of Lake Baikal. Being the internal sea between the mountainous west coast of the lake and the coast of island Olhon, this area is isolated and closed for air mass transportation. Here the pollution sources were discovered by the specific effect of local residential emisisons characterized by high AAE, low PM10 mass and BC concentrations as well as increased BC/PM_{10} ratio around 0.25. The AAE value of 3.3 and 2.2 were found on the Olhon - Ogoy and Ogoy - Khuzhir route, respectevely, indicating the local coal and wood combustion emissions from numerious touristic settetlements, campings and resorts located along Olhon island coast. Further, from Khuzhir to Nizhneangarsk, in the pristine area remoted from any anthropogenic sources the AAE value found around 1, it had indicated the impact of local BC emission because the BC/PM_{10} ratio was observed to be increased, up to 0.35.

From Nizhneangarsk the AAE value was increased and then until Barguzinsky bay was stable, around 1.8. Thus, the optical marker showed the continuous effect of the increased BrC in the aerosol composition from wildfire smoke transported from Yakutia region during this period. That was supported by increased fine particle number, PM_{10}, and BC as well as by aerosol field observations in the troposphere. The BC/PM_{10} ration was approached 0.5 on the route Nizhneangarsk - Severobaikalsk when the wildfire plumes were firstly approached the observation sites at low PM_{10} concentrations.

Only Barguzinsky - Selenga when air mass origin was significantly changed on the opposite orientated one from Mongolia region, the source of emissions for Baikal aerosols was changed to FF combustion dominated. AAE showed value equal 1 and BC/PM_{10} dropped to 2.0. In the Selenga region of intensive industrial residential sources, the AAE value was found consistently high, around 2.2 while the level of pollution, around 0.17, was similar to the Sourthern Baikal. The last the Selenga - Bolshie Koty route included the biggest touristic area Pestchanka, the AAE value was found also high, around 2.2, and level of pollution again approached the high level, of 0.3, in correlation with high PM and BC, indicating the impact of residential BB, shipping, and transport.

Conclusions

In summer 2018, in a season of Siberian wildfires, the ship - based expedition exploring the Baikal aerosol characteristics was performed over the lake area. Near - water surface sampling was supported by simultaneously operated lidar sounding of aerosol tropospheric fields. Geographically - distributed structure of aerosol fields during the expedition routes represent the observed variations from clean to the most polluted atmosphere by the best way. The applied approach has allowed to evaluate the interrelations between changes of the spatial structure of the aerosol loading, size distribution, spectral light absorption with meteorological and synoptic situations.

For the first time the distribution of PM_{10} and BC mass concentrations over Baikal water area was obtained; their spatial-temporal variability was found non-uniform over the lake water area indicating the impacts of emissions transported from industrial areas of the Angara river valley and Buryatia, from numerous popular touristic and residential places on the coast, and remote wildfires. The observational data for a measure of the aerosol spectral absorption, AAE, suggest that the most severe atmospheric pollution over the lake is associated with atmospheric transport from large coal - fired power plants both from the direction of Irkutsk and Buryatia region as well as from numerous local residential coal and biomass burning. Orographic features of a Baikal coastal zone such as gulfs, bays, and hills are found having the particular impact on pollution accumulation, more specifically in the Southern Baikal area, Maloe more, and Barguzin gulf. Significant increase of PM_{10}, BC and fine particle size distribution relating with AAE values typical for biomass burning is observed during the wildfire smoke, transported from the intensive wildfires in the Yakutia and Krasnoyarsk Kray and covered the lake water area from north along the east coast. Assessment of the deposition of hygroscopic ammonium sulphates and hydrophobic BC to water surfaces contributes to the determination of the impact which various aerosols might have on sensitive Lake Baikal ecosystem.

Declaration of Competing Interest

No interest conflict.

Asknowledgement

Expedition work was organized using the financial support of LIN SB RAS state task No. 0279-2021-0014. Data analyses was performed under the partial support of PEEX program.

O. Popovicheva thanks to RFBR project № 18-60084 for support of aethalometrical measurements and data analyzing.

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.jes.2021.01.011.

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