Conference paper

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Organic carbon in atmospheric precipitation in the urbanized territory of the South of Western Siberia, Russia

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Abstract: This paper presents the results of studying the contents of total (TOC) and dissolved (DOC) organic carbon in atmospheric precipitation and their deposition fluxes on the territory of the city of Barnaul. Samples of atmospheric precipitation (rain and snow) were collected from May 2016 to December 2020 in the city center, additionally at the end of winter 2018–2019 samples of snow cover were taken in the territory of the city and its environs. The studies showed a significant content of organic carbon (OC) in atmospheric precipitation: the weighted average concentrations for the study period were 7.2 ± 0.6 and 4.2 ± 0.4 mg/L for TOC and DOC, respectively. The annual flux of OC deposition with atmospheric precipitation on the territory of Barnaul over the past three years has varied within 2.4–3.9 t/km^2 for TOC and 1.4–2.1 t/km^2 for DOC. To visualize the spatial distribution of organic matter over the territory of Barnaul, simple kriging was used, implemented in the Geostatistical Analyst module (ArcGIS® Desktop). The flow of organic carbon input into the snow cover during the winter period was used as data for the geostatistical model. According to the model, the deposition of OC from the atmosphere occurs unevenly throughout the urban area and depends on the location and intensity of pollution sources.

Keywords: Aerosols; atmospheric deposition; atmospheric precipitation; chemistry and climate; geostatistical model; organic carbon; snow cover.

Introduction

The contribution of organic carbon (OC) to the chemical composition of atmospheric aerosol, especially the finely dispersed fraction, is very significant and, according to various data, can be 10–50 % [1], 40–65 [2], 80 % [3, 4] and even 92 % [5]. The initial sources of OC are both biogenic (vegetation emissions, ocean emissions, volcanic eruptions) and anthropogenic (burning urban waste, biomass and various fuels) origin [6, 7]. A significant part of OC is also formed as a secondary aerosol product in the atmosphere under photochemical

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oxidation of volatile organic compounds [8, 9]. The contribution of the initial biogenic sources to OC emissions is quite significant and on a global scale it is up to 80 % [10]. However, despite the predominance of biogenic emissions, the presence of a significant correlation of organic carbon with elemental carbon [11], as well as with other pollutants, indicates the importance of anthropogenic emissions in the input of OC in atmospheric precipitation [10]. Precipitation is known to be an important source of OC in surface waters [12] and can have a significant impact on the hydrological cycle [13]. Deposits of inorganic components and nutrients from the atmosphere have a serious impact on the environment and are well studied [14]. At the same time, insufficient attention is paid to the deposition of OC due to the chemical complexity of its mixtures, but at the same time it plays a fundamental role in the functioning of ecosystems and understanding the rate of its deposition is important for estimating the global carbon budget [15].

The aim of our work is to study the concentration of total (TOC) and dissolved (DOC) organic carbon and the fluxes of their deposition in with atmospheric precipitation on the territory of the south of Western Siberia (within the city of Barnaul).

Materials and methods

Study site and sample collection

Samples of atmospheric precipitation (rain and snow) were collected after each event (but not more often than once a day) from May 2016 to December 2020 in the center of Barnaul in an open area (the roof of the building of the Institute of Water and Environmental Problems of the SB RAS), located at a height of 25 m from the surface of the earth. Rain samples were collected in a plastic bottle fitted with a plastic funnel. Snow samples were collected in a plastic box placed on the bottom of a barrel equipped with a windscreen to prevent snow from blowing out. Snow cover was sampled at 27 points in Barnaul and its environs in March 2019 at the time of maximum snow accumulation (Fig. 1). A composite snow sample was formed from 5 to 10 single samples, which were taken with a plastic pipe with the inner diameter of 4.5 cm.

The city of Barnaul (53°20′N, 83°46′E) is located in the south of Western Siberia, Russia. The openness from the Arctic Ocean and the semi-desert regions of Central Asia creates the possibility of the influx of various air masses, contributing to significant variability of weather conditions and the accumulation of impurities in the

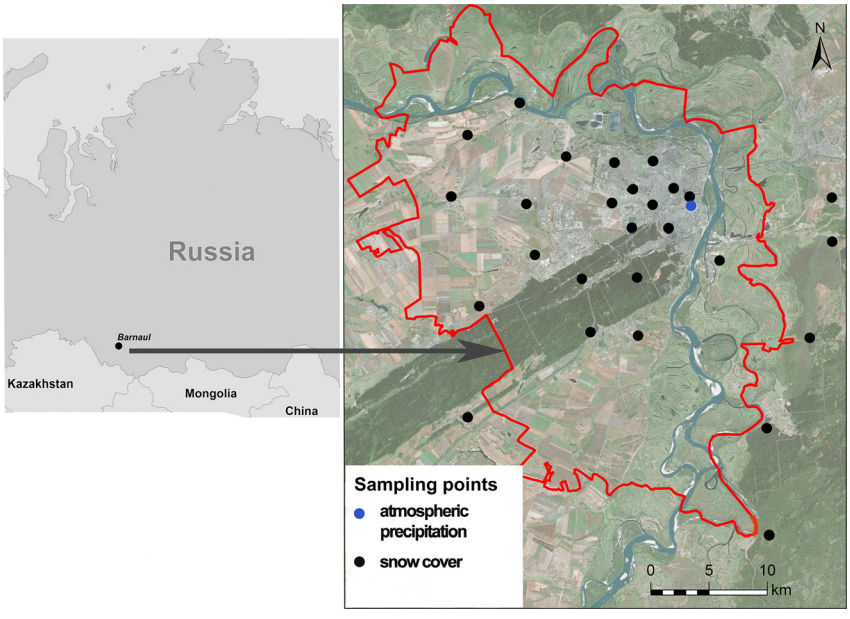

Fig. 1: The sampling points of atmospheric precipitation (2016–2020) and snow cover in and around of Barnaul in 2019.

surface part of the atmosphere. Barnaul has a continental climate with long cold winters and short hot summers. Average annual precipitation is 433 mm. The prevailing wind directions are southwest, west and south, blowing at an average annual speed of 1.8 m/s. A stable snow cover forms on average in the first decade of November and completely collapses at the beginning of April.

Chemical analysis

The organic carbon (OC) content was estimated based on the chemical oxygen demand (COD) values. Previous studies show this possibility due to the existence of a linear relationship between OC and COD [16, 17]. The COD was determined according using a DR 2800 spectrophotometer (HACH Lange Company, Germany). Unfiltered samples were analyzed to estimate total organic carbon (TOC), and samples filtered in an argon flow (filter pore diameter 0.45 μm) to estimate dissolved organic carbon (DOC). The content of OC in precipitation and melt water was calculated as follows: OC = 0.375·COD [18, 19].

Data processing

The volume-weighted mean (VWM) concentrations (mg/L) of OC are calculated by Eq. (1):

$$\mathrm{VWM} = \frac{\sum_{1}^{n} (\mathrm{Cn} \cdot \mathrm{Vn})}{\sum_{1}^{n} \mathrm{Vn}}, \tag{1}$$

where VWM – volume-weighted mean concentration, mg/L; Cn – concentration (mg/L) and Vn (L) – the atmospheric precipitation amount of an individual precipitation event.

To interpolate the spatial distribution of organic matter, simple kriging is used from the family of geostatistical kriging models implemented in the Geostatistical Analyst module of the ArcGIS® Desktop geographic information product. Kriging methods are based on statistical models that include autocorrelation (statistical relationships among the measured points). Not only do these techniques have the capability of producing a prediction surface, but they can also provide some measure of the certainty or accuracy of the predictions [20, 21]. The dataset has been converted using the normal score transformation to standard normal distribution. Because density of sampling points location is different on the territory sections, then data are decluttering by cells.

The fluxes of OC input during the winter period from the atmosphere to the underlying surface were used as data for the geostatistical model. These fluxes were calculated using the following formula (2):

$$\mathrm{P} = \frac{\mathrm{C} \cdot \mathrm{V}}{\mathrm{S} \cdot \mathrm{N}}, \tag{2}$$

where P – the mass of the OC per unit area during the period of snow cover, mg/dm^2; C – the concentration of the CO in the melt water, mg/L; V – the volume of the melt water in the sample, dm; S – the area of the internal transverse pipe cross-sections, dm^2; N – the number of snow cores taken at one point.

The annual deposition fluxes of CO are calculated by Eq. (3):

$$\mathrm{F} = \mathrm{C} \cdot \mathrm{H}, \tag{3}$$

where F – a flux deposition for OC, ton/km^2; C – the VWM concentration OC, mg/L; H – the amount of precipitation over a timed period, mm.

Results and discussions

During the study period, 432 samples of wet atmospheric precipitation (rain, snow) were analyzed. The results of the concentration of organic carbon in the atmospheric precipitation of the city of Barnaul in comparison with the data of other researchers are presented in Table 1. As can be seen from Table 1, anthropogenic factor affects the concentration of organic carbon in atmospheric precipitation. The organic carbon content is

Table 1: Concentration of organic carbon (CO), mg/L in atmospheric precipitation.

	Location		OC	Mean ±(SD)	Range	Author
Europe	Schauinsland, Germany	Background	DOC	1.49 ± 1.29	0.18–5.07	Cerqueira et al. [35]
	Aveiro, Portugal			0.44 ± 0.19	0.20–0.80	
	K-Puszta, Hungary			1.35 ± 0.67	0.57–2.76	
	Leon, Spain	Urban	DOC		0.40–1.75	Oduber et al. [36]
	Gdynia, Poland	Urban	DOC	2.9 ± 2.4	0.1–13.6	Witkowska and Lewandowska [28]
	Turin, Italy	Urban	TOC		0.42–2.38	Albinet et al. [37]
	Thessaloniki, Greece	Urban	DOC	4.82	0.46–21.3	Balla et al. [38]
	Poznan, Poland	Urban	TOC		3.56–23.19	Dąbrowska and Nawrocki [24]
	Poznan, Poland	Urban	DOC	5.10 ± 7.46	0.18–48.7	Siudek et al. [27]
	Jeziory, Poland	Suburban	DOC	4.72 ± 4.21	1.59–19.53	
	A Coruna, Spain	Suburban	TOC	15.3	0.88–116	Moreda-Piñeiro et al. [39]
	Zagreb, Croatia.	Urban	TOC	2.05 ± 1.49	0.10–6.44	Orlović-Leko et al. [40]
	Syktyvkar, Russia	Suburban	DOC		0.69–4.86	Kuznetsova and Archekhova [25]
			TOC		7.3–25.30	
South America	Uberlandia, Brazil	Suburban	DOC	2.6		Ciglasch et al. [41]
	Mexico, Mexico	Urban	DOC		2.4–19.5	Montero-Martínez et al. [42]
	Araraquara, Brazil	Suburban	DOC	4.06 ± 0.48.		Coelho et al. [43]
	Ribeirao Preto, Brazil	Suburban	DOC	3.26 ± 0.26.		
	Pena Roja, Colombia	Forest	DOC	3.94 ± 1.62		Tobon et al. [44]
	Loja, Ecuador	Forest	TOC	4	2.2–5.5	Wilcke et al. [45]
			DOC	0.25	0.15–0.35	
North America	Bondville, USA	Suburban	DOC	1.7	0.55–3.88	Torres et al. [46]
	Westwood, USA	Urban	TOC	9.8 ± 5.4	2.0–18.6	Kawamura et al. [23]
	Wilmington, USA	Urban	DOC		0.04–1.48	Mullaugh et al. [47]
	Green Lakes Vall., USA	Suburban	DOC	1.12 ± 0.19		Mladenov et al. [48]
	Mont St. Hilaire, Canada	Suburban	DOC	2.0		Dalva and Moore [49]
Asia	Shenzhen, China	Urban	TOC	2.21	0.118–25.9	Huang et al. [50]
	Yangjuangou, China	Suburban	DOC		0.56–28.71	Wang et al. [51]
	Yulong, China	Suburban	DOC	1.11 ± 0.07		Niu et al. [52]
	Khun Sathan, Thailand	Suburban	DOC	1.7 ± 0.2		Möller et al. [53]
	Baoding, China	Urban	DOC	3.1 ± 4	1.1–6.3	Pan et al. [26]
	Beijing, China		DOC	3.9 ± 0.8	1.2–7.6	
	Tianjin, China		DOC	2.7 ± 0.5	0.6–6.5	
	Tangshan, China		DOC	2.4 ± 0.3	0.8–4.0	
	Cangzhou, China	Suburban	DOC	3.3 ± 0.7	0.3–5.5	
	Launcheng, China		DOC	2.7 ± 0.5	0.6–3.8	
	Yangfanf, China		DOC	3.9 ± 1.0	1.3–4.5	
	Qingdao, China	Urban	DOC	2.60	0.34–7.39	Wang et al. [54]
	Yantai, China		DOC	3.19	0.84–6.77	
	Taian, China	Suburban	DOC	0.768	0.36–1.02	
	Guandaushi, Taiwan	Suburban	DOC	4.7 ± 2.9		Liu and Sheu [55]
	Seoul, South Korea	Urban	DOC	1.1	0.18–9.36	Yan and Kim [56]
	Dhaka, Bangladesh	Urban	DOC		0.2–37.9	Nahin Mostofa et al. [57]
	Vladivostok, Russia	Urban	DOC		1.9–4.2	Kondrat'ev et al. [58]
	Barnaul, Russia	Urban	TOC	7.2 ± 0.6	0.75–96.75	This study
			DOC	4.2 ± 0.4	0.75–74.25	

noticeably higher locating in urban sampling points. In urban on the continent, the concentration of organic carbon is higher than in urban located on the coast. This is because in continental regions, in contrast to coastal regions, chemical reactions for the formation of secondary pollutants are more active [22].

Our data show what, the variation in the concentration of organic carbon in individual samples was significant: 0.75–96.75 mg/L for TOC and 0.75–74.25 mg/L for DOC. The weighted average values of the concentrations of TOC and DOC were 7.2 ± 0.6 and 4.2 ± 0.4 mg/L, respectively, which significantly exceeds the average values of the global atmospheric content of DOC, equal to 2.64 ± 1.9 mg/L [10].

The organic carbon is of very much scientific interest researchers of the world. But there are very few data on the content of the TOC in the research literature. However, our data are well comparable with the results of the TOC in the atmospheric precipitation of continental cities such as Westwood (USA) [23], Poznan (Poland) [24] and Syktyvkar (Russia) [25]. And our data are well comparable with the average results of the DOC content in the urban atmospheric precipitation of Asia 3.9 ± 0.8 mg/L (Beijing, China), 3.1 ± 4 mg/L (Baoding, China) [26] and Europe 5.10 ± 7.46 mg/L (Poznan, Poland) [27].

There is no reliable seasonal dynamics of TOC carbon contents in atmospheric precipitation in Barnaul (warm period 7.3 mg/L; cold period 6.9 mg/L), while DOC contents are characterized by an increase in precipitation of the warm period (4.7 mg/L) as opposed to the cold one (3.3 mg/L). The last is confirmed by the data of other researchers [28, 29] and the results of studying the composition of atmospheric aerosol [30–32].

The results of our work showed that the values of OC concentrations are inversely proportional to the amount of precipitation, which was also noted by other researchers [33, 34]. At the same time, the values of OC concentration were below the detection limit for only 7 % of getting samples. These samples were collected at the next day after intense precipitation with low values of organic carbon concentrations. This shows that the removal of organic matter from the atmosphere occurs after the first portions of heavy precipitation.

Depending on the location water-soluble organic compounds make up a significant part (10–90 %) of the OC content in environmental aerosols [4]. For example, in northern China the part of dissolved organic carbon is 79 % [26], in Poland 87–91 % [27]. Based on the data of our study, the content of DOC in the total volume of OC was on average 58 % during the period 2016–2020.

The annual flux of OC deposition with atmospheric precipitation on the territory of Barnaul in 2016–2020 was 2.4–3.9 t/km^2 for TOC and 1.4–2.1 t/km^2 for DOC. Seasonal flux variations were as follows: for TOC 0.9–2.2 t/km^2 during the warm period and 0.7–1.0 t/km^2 during the cold period; for DOC 0.4–1.6 t/km^2 in the warm period and 0.3–0.5 t/km^2 in the cold one.

In contrast to the limited possibilities of stationary sampling of atmospheric precipitation, the snow cover makes it possible to assess the irregularity of the substances deposition from the atmosphere to different parts of the study area, which makes it possible not only to indirectly assess the quality of the ambient air during the cold period, but also to determine the sources of pollution and compare their intensity [59]. The use of geostatistical analysis methods (in our case, simple kriging) allows us to obtain a spatial characteristic of the distribution of pollutants in the study area and to make conclusions and assessments.

The geostatistical model of the distribution of organic carbon in the snow cover of the city of Barnaul is shown in Fig. 2.

In winter, emissions of organic carbon from vegetation are very low, so its input is mainly associated with anthropogenic sources (car exhaust and combustion of fossil solid fuels). According to Fig. 2, the deposition of organic carbon from the atmosphere is irregular and its maximum values are recorded in areas adjacent to motorways (car exhaust), as well as thermal power plants and the private sector (solid fuel combustion).

Conclusions

The studies carried out have shown a significant content of organic carbon in atmospheric precipitation in the city of Barnaul. The concentration of DOC is subject to seasonal changes, while the content of TOC does not depend on the season. The values of the weighted average concentrations of OC were 7.2 ± 0.6 and 4.2 ± 0.4 mg/L for TOC and DOC, respectively. The annual fluxes of organic carbon with atmospheric

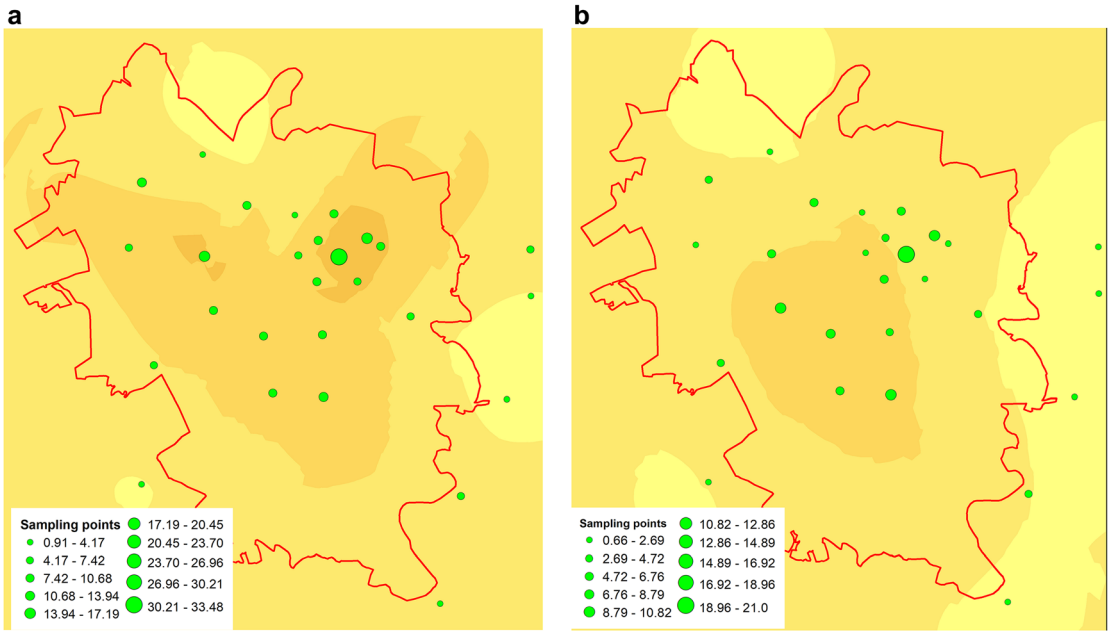

Fig. 2: The spread of OC in the snow cover in and around of Barnaul in 2019: TOC (a), DOC (b), mg/dm^2.

precipitation into the study area in 2016–2020 amounted to 2.4–3.9 t/km^2 for TOC and 1.4–2.1 t/km^2 for DOC. The geostatistical model showed the heterogeneity of the areal distribution of organic carbon in the snow cover, depending on the location of local pollution sources.

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