Physical and chemical properties of atmospheric aerosols in Moscow and its suburb for climate assessments

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Abstract. The paper presents results of experimental study of the physicochemical properties of surface aerosols in the Moscow region in the summer of 2019. Microphysical parameters, mass concentration and elemental composition of submicron and micron aerosol particles are considered, and their morphological structure is described. The features of variability of aerosol characteristics under atypical weather conditions prevailing in June-July 2019 are shown. The spatial distribution of the elemental composition of aerosols is established, a significant degree of their enrichment with heavy metals and metalloids is revealed. Geochemical analysis of aerosols and soils in Moscow was performed, and the coefficients of local aerosol concentration were calculated. The obtained experimental data will be useful for the refinement and verification of climate models.

1. Introduction

The uniqueness of the properties of aerosol particles determines their active role in atmospheric processes leading to changes in the Earth’s climate and the state of the environment. Microphysical parameters dynamically change in space and time, the chemical composition and morphology of aerosols affect the optical characteristics of the atmosphere and the heterogeneous processes occurring in it, as well as the thermal regime of the atmosphere and the Earth’s surface. However, the contribution of aerosols to climate formation is still not well defined partly due to the lack of experimental studies of atmospheric aerosols, their distribution and chemical transformations. Strong heterogeneities in the chemical and dispersed composition and morphological structure of aerosol particles, depending on the region and local sources, can significantly affect local weather conditions, chemical composition, and atmospheric circulation [1-3].

Due to the strong spatiotemporal variability of aerosol parameters and insufficient experimental data on dynamic processes, modeling of the structure of atmospheric aerosols and their effect on the climate is difficult; the assessment of their emission, chemical transformation, and transport in the atmosphere is not quite correct, which causes significant errors in the accuracy of climatic models and ecosystem prediction.
In connection with the ongoing urbanization of the territory of Russia, the release of aerosols of anthropogenic origin into the atmosphere has increased significantly, which exacerbated the problem of their impact on the climate. Solving this problem requires the development of experimental approaches to observing the characteristics of aerosols which are necessary for modeling aerosol processes and structures. The most promising seems to be an integrated approach combining terrestrial and remote aerosol research methods, the use of long-term series of experimental data and numerical simulation results.

This methodological approach is used by the authors in the study of the multicomponent composition of atmospheric aerosols in the surface layer of the atmosphere of urbanized zones. This paper shows the first results of studying the physicochemical properties of surface aerosols in the atmosphere of the Moscow region.

2. Subjects, conditions, methods, and tools of experimental studies

The subjects of research were fine- and medium-dispersed microparticles of aerosols in the surface layer of the atmosphere of the Moscow region in the summer from 10.06. until July 10, 2019. Observation points were selected taking into account the degree of urbanization and potential sources of aerosols and were located in a zone of dense development - in the Central Administrative District of Moscow (IAP RAS main building, Pyzhevsky per., 3, and site near Podsosensky per., 18/5), in the green urban zone of moderate development and park landscapes - in the Western administrative district of Moscow (Moscow State University, 1M zone, Vorobevy Gory), and in the semi-background suburban zone - at the Zvenigorod Scientific Station (ZSS) of the IAP RAS (Odintsovo District, Moscow Region).

For study the multicomponent structure of surface aerosols we use a geochemical approach based on a complex analysis of the geochemical composition of aerosol particles and soils of the natural and anthropogenic landscapes when the observation points are placed along the main mass transport [4].

The landscapes of the Moscow metropolis are formed on the paleoglacial deposits of the Russian Plain, experiencing strong exogenous processes (water erosion and accumulation, karst). A moderate continental climate with a strong influence of the marine Atlantic and with a pronounced seasonality influences the transformation of the surface of soils. In the temperate climate of the East European Plain, waves of heat and cold brought by various air masses freely propagate, contributing to large fluctuations in air temperature and rainfall. The annual rainfall is 600-800 mm with a maximum in the summer, and the average annual humidity is 75%.

Due to the dense development, wind speeds are insignificant - 2-3 m / s. The air regime of Moscow depends on the terrain, where air flows from the elevated periphery to the lower center, exacerbating the condition of the air basin of the city. In recent years, the influence of the heat island above the Moscow megalopolis, which is being formed under anthropogenic influence, has increased with general global warming.

Natural vegetation belongs to the East European subboreal forest region of the Central Russian province. Natural soil cover is represented by sod-podzolic soils, which are mostly destroyed. Most of the territory of the Moscow metropolis is currently composed of urban soils with artificial introduced vegetation. Significant areas are sealed with pavement and urban development, unsealed areas in the center of Moscow from 3 to 5%. Dynamically complex natural-anthropogenic and technogenic landscape-geochemical systems are actively forming on the territory of Moscow.

The observation period (June 10 - July 10, 2019), depending on the prevailing meteorological conditions, can be divided into several time intervals. The first interval (10.06 - 12.06) was characterized by anticyclonic activity, moderately hot weather, calm or a quiet wind of the dominant western direction. The change of air masses to the north and north-east (13.06 - 14.06) caused a decrease in air temperature and purification of the atmosphere. Cyclonic activity with prevailing winds of the north-west, north and north-east directions (22.06 – 30.06) led to a gradual significant decrease in temperature and partial leaching of aerosols from the surface layer. The first decade of July was characterized by a quiet and light wind of predominantly western direction, cool weather with a short-term increase in air temperature on certain days and almost daily rainfall.
Analysis of the synoptic and meteorological conditions of observations using the method of backward trajectories of air masses calculated by the NOAA HYSPLIT model [5], revealed the dominance of Arctic, north-western and northern air masses over a considerable period of experimental studies in the Moscow region.

The combination of these factors led to the formation of atypical state of the atmosphere for the summer and the dynamic processes occurring in it, favorable for the purification of the surface air of the Moscow region from contaminants.

The microphysical parameters of aerosol particles were measured using the aerosol complex described in [6] and based on the laser spectroscopy principles.

The mass concentration of aerosols was determined by the gravimetric method, as well as by the numerical method using the measured counting concentration of microparticles.

Aerosol samples were taken by suction sampling devices for subsequent gravimetric and chemical analysis using AFA-HA-20 analytical filters. The sampling time at the observation points of the IAP RAS and ZSS of the IAP RAS was 24 hours, at the observation points “Podosensky, d. 18/5” and “Moscow State University, zone 1M” - 24 hours or more, depending on weather conditions.

To determine the mass fractional and elemental composition of aerosol particles, a 6-cascade impactor was used. The sizes of the particles deposited on the cascades of the impactor are: 1 -> 6.5 microns; 2 - 4.0-6.5 microns; 3 - 2.5-4.0 microns; 4 - 1.5-2.5 microns; 5 - 0.5-1.5 microns; 6 - <0.5 μm.

Laboratory analysis of aerosol samples for the study of elemental composition was carried out by inductively coupled plasma atomic emission spectrometry (ICP-AES) [7, 8].

To study the morphological structure of individual aerosol microparticles, samples were taken on fluoroplastic membrane filters of the MPFC type. The morphology of the objects was determined by scanning electron microscopy [9]. The studies were carried out in a high resolution scanning electron microscope with a cold emission cathode JSM-7500F company JEOL (Japan).

The clarks of the elements concentration in the soil (KK) of the studied area were calculated for landscape-geochemical analysis. They characterize the deviation of the content of a chemical element in the soil from clark [4,10]:

$$KK = S/C,$$

where $S$ is the content of the chemical element in the soil, $C$ is the clark of this element in the granite layer of the continental crust [4,11,12].

To assess the selective accumulation of chemical elements in aerosol particles, we used the coefficients of aerosol concentration (accumulation) $KK_a$ [4,10]:

$$KK_a = A/C,$$

where $A$ is the content of the chemical element in aerosols.

To assess the atmospheric pollution of aerosol particles from urban soils and soils of the natural landscapes, the coefficients of local aerosol concentration $KK_u$ and $KK_l$ were calculated:

$$KK_u = A/US, \quad KK_l = A/L,$$

where $US$ is the content of the chemical element in urban soils, and $L$ is the content of the chemical element in the soils of natural landscapes.

3. Results

Temporal variability of microphysical parameters and mass concentration of surface aerosols

The time course of the counted and mass concentrations of submicron (up to 1 μm) and micron (1-5 μm) fractions of aerosol particles in Moscow in June 2019, obtained from aerosol spectrometer measurements are presented on graphs in Figure 1. For comparison, the temporal variability of meteorological parameters is shown. The main contribution to the total calculated and mass concentration of surface aerosols was made by particles of the submicron fraction, the amount of which increased with growing relative humidity. Variations in the concentration values of larger,
Figure 1. The time course of the calculated (a) and mass (b) concentrations of aerosol particles and meteorological parameters (c) in the near-surface air of Moscow in June 2019.
micron particles were insignificant. The most significant decrease in the calculated and mass concentration of large aerosol particles was observed at the end of June 2019. This period was characterized by strong cyclonic activity, accompanied by the invasion of cold Arctic masses, rainfall and decrease in air temperature. This led to leaching of large fractions of aerosol particles from the surface layer of the atmosphere.

The strong influence of meteorological and synoptic factors on the pattern of changes in the microphysical parameters of surface aerosols is also confirmed by Figure 2, which shows the variations in the daily average mass concentrations of submicron and micron particles determined by various methods. The gravimetric method allows to determine the total mass concentration of particles of all fractions contained in the surface layer of the atmosphere. Under cleaned atmosphere conditions in June-July 2019, the total mass concentration averaged 20-40 μg/m³ in the urban air of the Moscow metropolis. This exceeded the total mass concentration of aerosols in the suburban natural zone (ZSS IAP RAS) by 30-50%. This indicates that under similar synoptic-meteorological conditions and landscape-geochemical parameters of the observation region, the main role in atmospheric pollution is played by local anthropogenic sources. Figure 2 compares the curve of the change in the average daily concentration of aerosol particles PM$_{2.5}$, determined according to the data of aerosol spectrometers, and also shows the mass distribution diagram for the fractions of surface aerosols according to the data of the 6-stage impactor (upper right part of the figure). As can be seen from Figure 2, the average daily mass concentration of PM$_{2.5}$ particles, including fine fractions, is on average an order of magnitude lower than the total mass concentration of particles determined by the gravimetric method, and is mainly 1.5–8 μg/m³. This is due not only to the significant contribution of larger particles to the total mass, but also to the specific conditions prevailing in June-July 2019. This is confirmed, in particular, by our earlier studies of the variability of the surface concentration of PM$_{2.5}$ aerosols in Moscow. They showed that in more typical weather conditions for the summer period, the average daily mass concentration of PM$_{2.5}$ particles was 15–30 μg/m³ [13].
and large particles in the atmosphere. Under conditions of high humidity and cyclonic activity, the disperse composition and mass concentration of larger particles (> 2.5 μm) are subject to significant dynamic changes due to processes such as coagulation, flooding, leaching of particles. According to the data obtained using the impactor, it can be concluded that the main contribution to the mass distribution of surface aerosols of the Moscow metropolis in June 2019 was made by particles larger than 2.5 μm. This factor influenced the dynamics of changes in microphysical parameters and mass concentration of aerosols in the surface layer of the atmosphere of Moscow.

The different nature of the variations in the mass concentration of aerosols in Moscow and in the semi-background suburban zone (ZSS IAP RAS), with other similar conditions, may be associated with short-term local synoptic processes and meteorological phenomena, as well as with the specifics of local anthropogenic sources, but this requires more detailed study.

The elemental composition of surface aerosols in the Moscow region

The results of elemental analysis of aerosol samples showed a strong accumulation of some elements in aerosols of the Moscow megalopolis. In particular, high enrichment of heavy metals (Cd, Cu, Zn, Mo, W, Ti, Au, Hg, Pb, Ag, Mn, Fe, Co, As), metalloids (Bi, Sb, B, P, As, Sn) and sulfur are present. Moscow aerosols have elements Ca, Na, K, Fe, Al, Mg. Table 1 shows the elemental fractional composition (the percentage mass content of the chemical element in the particle fraction). Only those elements are indicated whose content in aerosol particles is significant. The elements of global distribution of Ca, Na, K, Fe, Al, Mg are distributed over all fractions, most of which are concentrated in larger particles. These elements are mainly of mineral origin. Heavy metals and metalloids of anthropogenic nature, as a rule, are concentrated in submicron and micron particles. Sulfur is an indicator of sulfate aerosols formed in the atmosphere during the oxidation of sulfur. Sulfur-containing aerosols are particles of nano- and submicron size, long-lived and subject to chemical transformation and long-distance transport. Such particles are most dangerous for human life and are actively involved in climate-forming processes [1,2]. Sulfur increased content in surface aerosols is observed in urbanized and natural zones of various regions of Russia because of the physicochemical properties of sulfur-containing aerosol particles and the wide distribution of sulfur sources (automobile exhaust gases, industrial fuel combustion products, natural gas) [4]. Aerosol particles with a high sulfur content were also found in the atmosphere of the Moscow region (see Table 1). They concentrated in the particles of fine fractions of PM$_{2.5}$.

| Elements | Impactor cascades (μm) |
|----------|------------------------|
|          | > 6.5 | 4.0-6.5 | 2.5-4.0 | 1.5-2.5 | 0.5-1.5 | < 0.5 |
| Ca       | 19.04 | 10.68   | 27.38   | 18.95   | 5.61    | 0.50  |
| Fe       | 7.57  | 6.11    | 16.60   | 17.74   | 5.56    | 0.84  |
| S        | 1.27  | 0.92    | 2.93    | 6.49    | 10.98   | 3.35  |
| Al       | 4.72  | 2.90    | 6.81    | 5.67    | 1.95    | 0.36  |
| K        | 1.94  | 1.10    | 2.72    | 3.39    | 1.56    | 0.19  |
| Na       | 1.79  | 0.87    | 2.65    | 3.79    | 1.64    | 0.34  |
| Mg       | 1.93  | 1.25    | 2.80    | 2.53    | 0.84    | 0.08  |
| Cu       | 0.43  | 0.17    | 0.58    | 0.85    | 0.74    | 0.10  |
| P        | 0.48  | 0.22    | 0.76    | 0.63    | 0.37    | 0.06  |
| Zn       | 0.33  | 0.15    | 0.39    | 0.65    | 0.64    | 0.19  |
| Ti       | 0.37  | 0.19    | 0.40    | 0.50    | not     | not   |
| Ba       | 0.13  | 0.14    | 0.43    | 0.53    | 0.14    | 0.02  |
| Mn       | 0.14  | 0.10    | 0.27    | 0.36    | 0.24    | 0.04  |
| Pb       | 0.05  | 0.02    | 0.08    | 0.29    | 0.32    | 0.10  |
| B        | not   | not     | 0.40    | not     | not     | 0.02  |
Figure 3 shows a diagram of the spatial variability of the gross percentage of certain chemical elements (heavy metals, metalloids, and sulfur) in the Moscow region in June 2019. The obtained data on the elemental composition of surface aerosols in urbanized and natural zones confirm high sulfur content in all areas. Moreover, the degree of its accumulation, as well as phosphorus in aerosols of the suburban area, exceeds the degree of accumulation in aerosols of the Moscow metropolis. This testifies to the existence of local sources of sulfur and phosphorus in Zvenigorod. Elements of global distribution Ca, Na, K, Fe, Al and Mg have an increased percentage and a lithogeny. An elevated content of anthropogenic elements tin, antimony, and nickel was found in the park zone of the MSU.

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A significant degree of accumulation of heavy metals and metalloids in surface aerosols was revealed at all observation points (see Figure 3). A comparative analysis with the data on the chemical composition of road dust in Moscow published in [14], and the values of the calculated coefficients of local aerosol concentration $K_K$ and $K_K$, showed that the surface aerosols of the Moscow metropolis have high concentrations of heavy metals and metalloids not only in relation to the natural soils, but also relatively polluted urban soils and road dust (Table 2). This indicates that surface aerosols are one of the most vulnerable and dynamic components in geosystems, which depends on anthropogenic pressure and affecting the environment and climate.

*Morphological structure of aerosol particles*
Aerosol microparticles are actively involved in atmospheric heterogeneous processes leading to changes in the state of the atmosphere and climate of the Earth. The nature of the course and kinetic parameters of such processes strongly depend not only on the chemical composition of the aerosol particles, but also on their morphological properties (size, shape, area and surface roughness) [1,15]. Therefore, when describing the kinetics of heterogeneous chemical processes in global and regional climate models, it is necessary to take into account the morphological factor of aerosol particles. The results of laboratory analytical studies of aerosol samples obtained during field observations in the Moscow megalopolis revealed various morphological types of aerosol particles, classified according to their structure, composition and origin. Figure 4 presents electronic microimages of some morphological types of aerosol particles contained in the surface layer of the atmosphere of the Moscow metropolis.
Table 2. The content of heavy metals and metalloids in aerosols and soils of Moscow

| Elements | Urban soil, mg/kg | Natural landscapes, mg/kg | Aerosols, mg/kg | $KK_I$ | $KK_u$ |
|----------|-------------------|---------------------------|-----------------|--------|--------|
| Cd       | 1.8               | 0.09                      | 10.76           | 110.89 | 5.54   |
| W        | 4.3               | 2.03                      | 17.13           | 8.70   | 4.11   |
| Bi       | 0.69              | 0.23                      | 9.96            | 41.69  | 13.90  |
| Zn       | 190               | 75                        | 2410.18         | 33.27  | 13.13  |
| As       | 8.3               | 5.6                       | 5.73            | 1.11   | 0.75   |
| Cr       | 98                | 92                        | 258.96          | 2.69   | 2.53   |
| Sb       | 1.3               | 0.81                      | 90.01           | 101.85 | 63.46  |
| Pb       | 50                | 17                        | 353.83          | 19.95  | 6.78   |
| Cu       | 61                | 27                        | 1030.13         | 38.62  | 17.10  |
| Ni       | 27                | 50                        | 236.64          | 4.18   | 7.74   |
| Sn       | 10.1              | 2.5                       | 198.93          | 70.02  | 17.33  |
| Co       | 7.2               | 15                        | 21.58           | 1.40   | 2.91   |
| V        | 55                | 106                       | 75.32           | 0.74   | 1.42   |
| Mo       | 1.2               | 1.1                       | 27.76           | 24.52  | 22.48  |

Figure 4. Electronic microimages of aerosol particles with different morphological structures: ab) agglomerates of layered nanoparticles, c) large solid soil particles, d) aluminosilicate soil particles of irregular shape with the addition of anthropogenic elements, e) round particles of thermocondensation type with traces of metals, f) soil particles of layered salt-mixed structures

These include:
- agglomerates of nanoparticles with a loose or layered structure (PM$_1$ particles);
- aluminosilicate soil particles of regular round shape mixed with carbon and nitrogen / or mixed with metal salts (PM$_{2.5}$, PM$_5$);
- aluminosilicate soil particles of irregular shape with the addition of anthropogenic elements (with an admixture of sulfur, salts and traces of metals / or with an admixture of sulfur, carbon and metal salts) (PM$_{2.5}$, PM$_5$, PM$_{10}$);
- sphere particles of thermocondensation type with traces of metals (PM$_3$);
- silicate particles with traces of metals of anthropogenic origin (iron, nickel, copper, zinc, molybdenum, manganese, chromium, etc.) (PM$_3$);
- soil particles of a layered structure with an admixture of salts (PM$_{10}$);
- large solid soil particles with impurities of anthropogenic elements (metal salts and lead) (> 10 μm).

The obtained data correspond to the classification of aerosols according to morphological parameters given in [16, 17].

4. Conclusions

An experimental study of the physicochemical properties of surface aerosols in the Moscow region in June-July 2019 made it possible to obtain data on the multicomponent structure of atmospheric aerosol particles, which is necessary for the refinement and verification of climate models describing dynamic atmospheric and climate-forming processes involving aerosols.

Meteorological and synoptic factors had a strong influence on the variability of parameters of atmospheric aerosols. The specifics of local sources of aerosols and atypical weather conditions prevailing in June-July 2019 in the Moscow region, caused the purification of the atmosphere from polluting impurities. An abnormally low mass concentration of PM$_{2.5}$ particles (1.8-8 μg / m$^3$) was recorded compared with long-term average values (15-30 μg / m$^3$). In these conditions, micron particles with a size> 2.5 μm are subject to the greatest dynamics of change.

A comprehensive analysis of the morphological structure and elemental composition of surface aerosols showed the presence of solid and loose mineral particles, mineral particles with an admixture of anthropogenic elements (sulfur, salts and traces of metals), particles or agglomerates of particles of anthropogenic origin (soot, carbon nanoparticles, thermocondensation particles with traces of metals) in the atmosphere of the Moscow metropolis in the summer of 2019. Atmospheric aerosols in the Moscow region have high concentration of heavy metals, metalloids and sulfur, independence from the degree of urbanization of the observation zone. Moreover, the accumulation of these elements in aerosols significantly exceeds the accumulation in road dust and contaminated urban soils. The results of elemental analysis of aerosols have confirmed that road transport is currently the main pollution source in the Moscow region. In particular, particles of almost all the revealed morphological types contain Cu, Pb, Sr, Fe, and Zn. The presence of carbon nanoparticles and their agglomerates in the urban air of Moscow may be associated with the combustion of natural gas, as well as with various anthropogenic sources, including the burning of hydrocarbons. Soil particles with different morphological structures have high concentration of aluminosilicates, as evidenced by the significant presence of Al and Si.

Acknowledgments

We are grateful to Yu. I. Obvintsev, V.A. Lebedev and L.O. Maksimenkov for assistance in arranging, supporting, and carrying out field studies of the physicochemical parameters of surface aerosols in the Moscow metropolis.

The reported study was funded by RFBR, projects ## 19-05-00352 and 19-05-50088; by the Ministry of Science and Higher Education within the State assignment FSRC «Crystallography and Photonics» RAS.

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