Investigation of chemical composition of atmospheric aerosol in Siberian towns. Part 1. Results of investigation of the near-surface aerosol in towns of the Irkutsk industrial region

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In February 2004, air pollution fields of East-Siberian towns, situated between Irkutsk and Tomsk, were studied using AKV-2 mobile station, designed at the Institute of Atmospheric Optics. Twenty two samples were collected and then used to follow the behavior of the inorganic component (total mass concentration of elements and ions) of the near-ground aerosol in nine towns of the region under study. The scatter in the concentration of the inorganic fraction turned to be large: from 22 μg/m^3 in Kansk suburbs to 0.5–0.6 mg/m^3 in the Usol'e-Sibirskoe industrial zone. In some cases, the concentration varied by an order of magnitude within the same town (Achinsk, Irkutsk). The synoptic situation at the sampling sites during the experiment has been analyzed. For towns of the Irkutsk industrial region (Irkutsk, Angarsk, Usol'e-Sibirskoe), the detailed aerosol-chemical matrix is presented, which includes both the inorganic composition and the contents of polycyclic aromatic hydrocarbons (PAHs) in public green spaces and industrial zones of these towns. The significant inhomogeneity of the inorganic component, in particular, within the towns of the same industrial zone, is caused, on the one hand, by the composition of emissions from local sources and by the prehistory of the air mass and, on the other hand, by the screening effect of the local circulation. The level of PAH concentrations, as well as their percentage more depend on the aerosol sampling site and its closeness to pollution sources comparative to the inorganic composition. Therefore, these components, being of definitely anthropogenic origin, can serve as tracers in the transport of aerosol between different town districts.

Introduction

Study of chemical composition of the atmospheric aerosol is very important for solving of a wide range of problems of atmospheric physics and optics, as well as for ecology. The chemical composition of particles influences their index of refraction and, correspondingly, regularities of solar radiation scattering and absorption in the atmosphere. Aerosol often contains heavy metals and polycyclic aromatic hydrocarbons (PAHs) with carcinogenic and mutagenic properties that makes study of the chemical composition of atmospheric aerosol very important for ecology. Dilution of sulfate-containing particles in cloud droplets triggers acid rains, which cause the degradation of biota, water, and soils. Therefore, the chemical composition of atmospheric aerosol is under active study for last decades.[1]

In this field, the study of anthropogenic aerosol is of special interest, particularly, for industrially urbanized agglomerations, i.e., modern towns, small in size, but having hazardous deviations of atmospheric parameters comparative to open nature. Accumulation of buildings and industrial enterprises, characterizing modern towns, results in significant environmental emission of all kinds of impurities, which are not observed in nature. Already first measurements have shown a large (to dozens of times) exceeding of concentration of some urban aerosol components in comparison with non-agglomerated environment.[2–4] Data of our previous measurements in several towns of the former USSR verified facts of multiple enrichment of urban aerosol with many chemical components.[5,6]

Traditional and stable concepts of urban aeration prevailing for the recent 15–20 years and based on the influence of the local climatic wind rose on urban landscape, fail to explain the impurity accumulation, which is observed not only in the surface layer, but sometimes in the whole boundary atmospheric layer above a town. Modern theories of formation and transformation of urban air pollution fields, rapidly developed for the last 10–15 years,[7–9] made the character of urban aeration much more clear. Nevertheless, sometimes, there is a discrepancy between the observed distribution of impurities and theoretically predicted special local circulation determining their behavior in the urban atmosphere. Therefore, the *in-situ* measurements of pollutant concentrations in industrial urban centers are of great importance. From this point of view, the aerosol component is of special interest because of complexity of its formation and its existence in the polluted urban atmosphere. Many recognized authorities unambiguously classify the aerosol, "living" in a town, as a special type, namely, urban aerosol.[10,11]

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Technique and conditions of the experiment

Our experiment was carried out with the use of a mobile station AKV-2, intended for air monitoring. It was produced at the Institute of Atmospheric Optics on the base of the Gas-66 vehicle. The station performs free-running measurements of meteo- and radiation parameters of the near-ground atmosphere, concentrations of trace gases, and disperse aerosol composition. A detailed description of the AKV-2 station is given in Ref. 12.

To study the ion-element composition, aerosol was sampled on AFA-KhP-20 filters via an outboard filter-aspirated system based on gas counters of SGB G4-1 type. Besides the inorganic aerosol fraction, polycyclic aromatic hydrocarbons (PAHs), which are microcomponents of aerosol and superecotoxicants of 1st class of hazard,[13] were of interest. To study them, aerosol from Irkutsk–Cheremkhovo industrial zone (Irkutsk, Angarsk, Usol'e-Sibirskoe) was sampled on fibrous glass filters "Whatman 41."

Chemical quantitative analysis for ion-element composition and PAH content was performed in laboratories of the Irkutsk Limnological Institute (PAH, hydrocarbonate-anion) and the Tomsk State University (elements and other ions) upon the expedition finishing. The analytical techniques, detection thresholds and errors are presented in Table 1.

Aerosol was filter-sampled in nine towns of Eastern Siberia, located along the federal route M53 between Tomsk and Irkutsk; in addition to Irkutsk, they were: Angarsk, Usol'e-Sibirskoe, Tulun, Hizhneudinsk, Taishet, Kansk, Krasnoyarsk, and Achinsk. The expedition started in Irkutsk on February 25 and finished in Tomsk on February 29, 2004. Sampling was carried out at the entrance or (and) exit of every town and near the town center. Sampling sites and time were planned for each town in such a way as to receive (under similar weather conditions, if possible) representative information on aerosol composition both from the industrial zone and from the conditionally reference zone, characteristic of residential part of town and its outskirts, which are the least industrially contaminated.[14]

The aerosol filter-sampling was conducted at an air aspiration rate of 6 m^3/h per filter during 20–40 min depending on the atmospheric pollution intensity. Time gaps between samplings within one town were as short as possible to increase the probability of similarity of weather patterns for all measurement sites. Further analysis of synoptic maps has shown that this condition was not satisfied only for sampling sites in the first town – Irkutsk (Table 2).

Table 1. Characteristics of techniques used in analysis of atmospheric aerosol samples

Component	Detection technique	Detection threshold, ng in a sample	Error, %
NO_3^-, SO_4^{2-}	Ion chromatography	600	5
HCO_3^-	Ion chromatography	500	10
Cl^-	Ion chromatography	100	12
F^-, NH_4^+, NO_3^-	Ionometry	200	10
Na^+, K^+	Atomic absorption	200	5
Al, Co, Cr, Mo, Ni, Ti, Zn, B, Si	Atomic emission spectroscopy	20	20
Ag, Ba, Cu, Pb, Sn, V, Mg, Mn	Atomic emission spectroscopy	10	20
Fe, Ga, W	Atomic emission spectroscopy	100	20
Ca, Cd	Atomic emission spectroscopy	200	20
PAH	Chromatography-mass spectrometry	0.001	10

Table 2. Sampling time and sites; attendant weather conditions

Date	Time (GMT)	Town, sampling site	Pressure pattern	Air mass
02.25.04	08:00	Irkutsk, airport district (town outskirts)	Trough center, CAF rear	CA
02.25.04	09:00	Irkutsk, Znamenskii convent (center)	Trough center, WAF rear	CT
02.26.04	05:40	Irkutsk, Novo-Lenino (industrial zone)	Cyclone south-east	CS
02.26.04	08:10	Angarsk, ORP, TPP-9 (industrial zone)	Cyclone south	CS
02.26.04	09:20	Angarsk, Potapov – Chaikovskii str.	Cyclone south	CS
02.26.04	12:10	Usol'e-Sibirskoe, south-east outskirts	Cyclone south-west	CS
02.26.04	13:00	Usol'e-Sibirskoe, railway station (center)	Cyclone south-west	CS
02.26.04	13:40	Usol'e-Sibirskoe, Chem-pharm distr. (industrial zone)	Cyclone south-west	CS
02.27.04	02:00	Tulun, distillery (industrial zone)	Cyclone west, CAF rear	CA
02.27.04	03:00	Tulun, exit, windward side (town outskirts)	Cyclone west	CA
02.27.04	06:50	Nizhneudinsk, entrance, leeward side	Ridge rear	CA
02.27.04	07:50	Nizhneudinsk, "Uda" hotel, central market	Ridge rear	CA
02.27.04	08:40	Nizhneudinsk, exit (town outskirts)	Ridge rear	CA
02.28.04	02:50	Taishet, center, railway station	Anticyclone south-east	CA
02.28.04	09:20	Kansk, entrance	Anticyclone south-east	CA
02.28.04	10:10	Kansk, center	Anticyclone south-east	CA
02.28.04	11:10	Kansk, exit	Anticyclone south-east	CA
02.28.04	18:30	Krasnoyarsk, center, Semaphornaya str.	Anticyclone center	CA
02.28.04	20:00	Krasnoyarsk, Sovetskii distr., exit	Anticyclone center	CA
02.29.04	01:30	Achinsk, entrance	Anticyclone north-west	CA
02.29.04	04:20	Achinsk, center	Anticyclone north-west	CA
02.29.04	05:25	Achinsk, exit	Anticyclone north-west	CA

Describe the weather patterns in measurement sites during the experiment.

On February 25, the weather near Irkutsk was determined by a trough formed by two powerful irregular cyclones centered on Middle Siberian Platean between rivers Nizhnyaya and Podkamennaya Tunguska and eastward Kamchatka Peninsula, as well as two anticyclones centered over Arctic Ocean and the Irtysh river upstream. Against the background of the trough, a polar front passed, whose cold and then warm branches affected the weather and temperature–humidity conditions in the region. The Arctic air mass was substituted with a temperate one.

On February 26, the center of the first cyclone was dislocated to the region of Vanavara village, therefore, the south-east part of the cyclone determined the weather in Irkutsk. To the beginning of measurements, a warm polar front passed through the town and its rear also influenced the weather, while temperature–humidity conditions were determined by the air mass from subtropical regions of Central Asia.

Measurements in Angarsk were carried out from 8 a.m. to 10 a.m. (Greenwich time), and the local weather was determined by the southern part of the same cyclone. Due to a streamflow of 100–120 km/h speed at the steering flow height, the cyclone moved south-east at a speed of 70 km/h near the Earth surface; when measuring from 12 to 14 o'clock, its south-west part was observed near Usol'e-Sibirskoe, while its central part – near Ust'-Kut. The subtropical air mass was observed throughout all measurement time.

On February 27 from 2 to 3 p.m., the weather near Tulun was determined by the cyclone rear still moving south-east. At about 2 p.m., a cold arctic front has passed through Tulun and from this moment on, the arctic air mass determined temperature–humidity conditions of the region.

A closing anticyclone has been developed in the cyclone rear. To the moment of measurements, the weather near Nizhneudinsk was determined by the rear part of the ridge connected with that formation. Against the ridge background, surface cold fronts passed and caused wind intensification resulting in a snowstorm.

On February 28, the anticyclone, centered to this time near Imbatskoe village, deepened and decelerated, so, to 3 p.m., its south-east rear determined the weather near Taishet. The same weather pattern was observed for all measurement runs in Kansk.

The deepening anticyclone moved south and its central part determined the regional weather when measuring in Krasnoyarsk.

On February 29, the anticyclone continued its moving south, so, to 2 p.m. (the beginning of measurements in Achinsk) its north-west rear determined the regional weather and its western part was observed to the end of the measurements.

Besides different (on weather pattern) sampling conditions in Irkutsk, it is necessary to note an unstable weather in Tulun, where the front passage and air mass change took place during the sampling time, disturbing the stable aerosol stratification.[15,16]

Discussion

Figure 1 shows a general behavior of the surface aerosol inorganic fraction in the East-Siberian towns. It should be noted that the "periphery/industrial zone" division is quite conditional for small towns with undefined districts; it is based there on windward/leeward location of sampling sites.

Consideration of the relative chemical composition of the aerosol inorganic fraction in the three first towns (Fig. 2) allows us to believe that it strongly depends on the air mass prehistory. This was especially pronounced in Irkutsk and Usol'e-Sibirskoe (after entering there the subtropical air mass from Central Asia).

Occurrence of silicon in aerosol composition after the air mass replacement on February 26, most likely, was caused by its long-distance transport than by local sources. At the same time, this phenomenon was not so pronounced in Angarsk, which can be explained by the town location in the center of the industrial zone common for several towns. A common pollution cap, including thermal emissions, which is formed above this zone, favors a local circulation with a common column over Irkutsk–Cheremkhovo industrial region and creates a screening effect over the column's center due to the main flow passing around it.

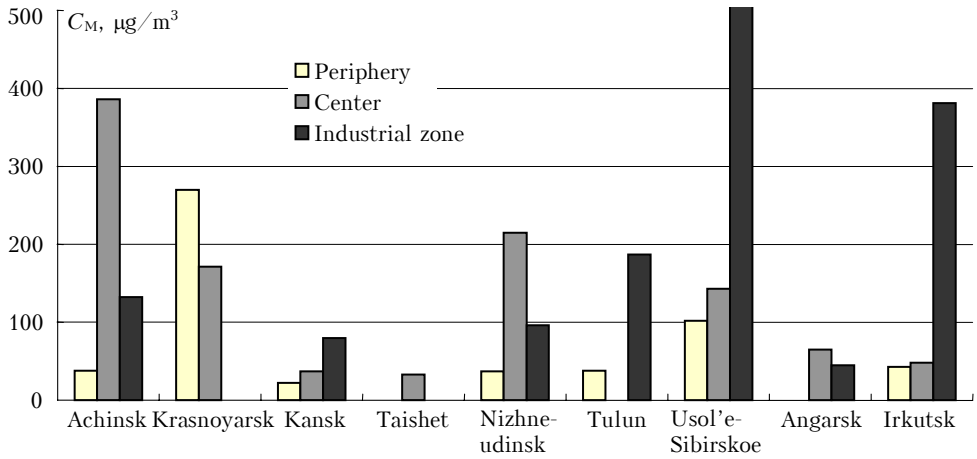

Fig. 1. Total concentration of the surface aerosol inorganic fraction in East-Siberian towns.

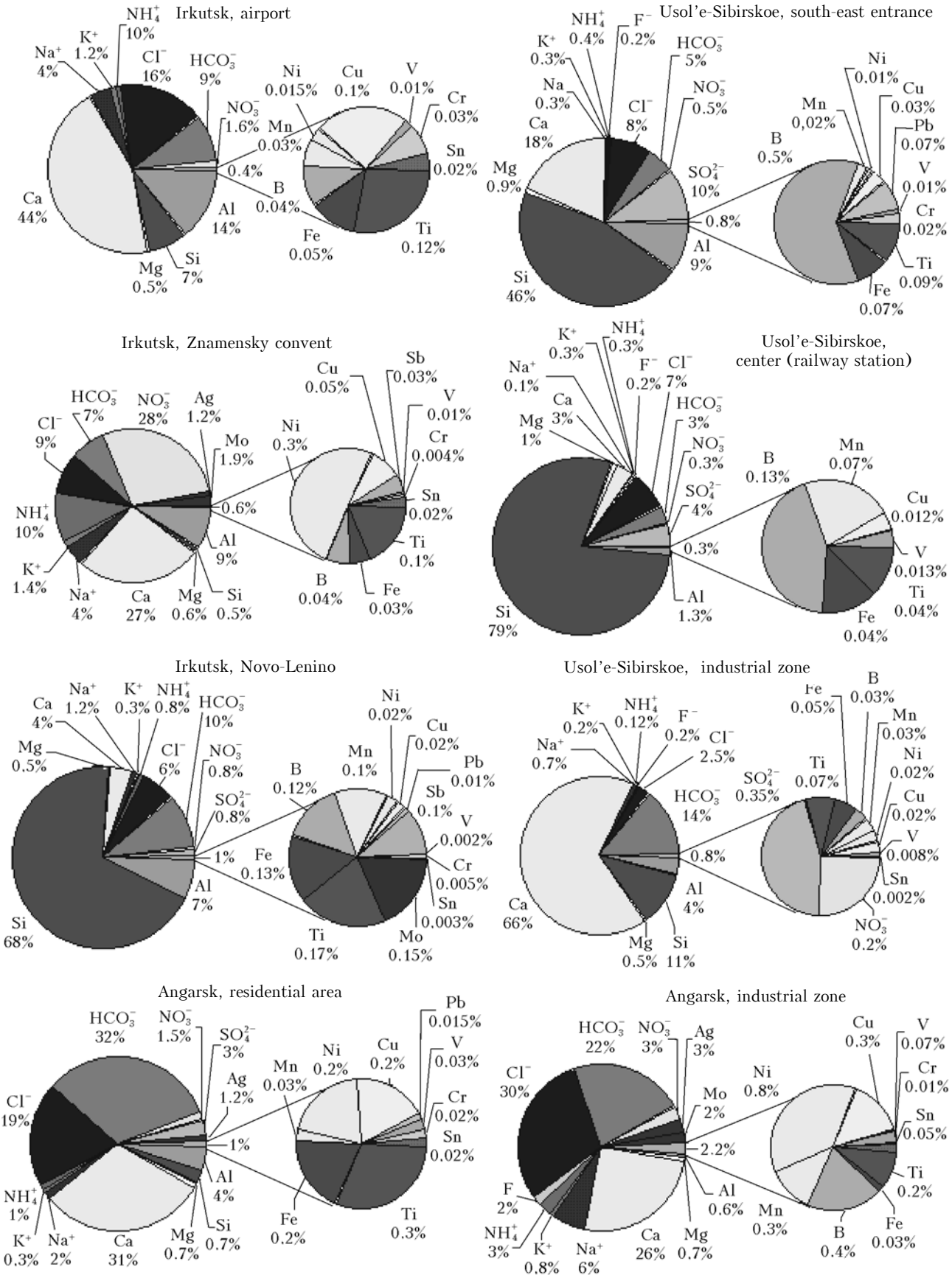

Fig. 2. Relative composition of the aerosol inorganic fraction in different districts of towns of Irkutsk industrial zone.

The column's ends, apparently, actively interact with the arriving air mass in the boundary layer, which results in aerosol fallout. The tendency of decreasing of enrichment factors for many aerosol microelements and ions (Table 3) in aerosol of industrial zone of Irkutsk and Usol'e-Sibirskoe can well be connected with their dilution due to desert aerosol fallout from the steering flow to the near-ground layer.

Enriching aerosol with microelements (V, Sn, Cl, F, Ag, Mo) is several times, if not orders, higher in the industrial zone of Angarsk than in the residential zone and in sampling sites of other towns. This manifests itself against the background of comparatively moderate (50 $\mu g/m^3$) total mass concentration of the aerosol inorganic fraction in the Angarsk industrial zone, although emissions of pollutants into its atmosphere are much stronger than in other towns of the region, including Irkutsk.[17] The total inorganic content also turned out to be 1.5 times higher (70 $\mu g/m^3$) in the Angarsk residential area. At the same time, total PAH concentration levels in aerosol samples taken from different town districts have an inverse ratio (Table 4).

In public green spaces, total concentrations of the priority PAHs varied from 20 to 30 ng/m^3. The benzo(a)pyrene concentration did not exceed MPC (1 ng/m^3). Elevated concentrations of PAH sum in aerosol were found in industrial zones of Irkutsk (Novo-Lenino) and Usol'e-Sibirskoe, where the benzo(a)pyrene concentration exceeded MPC by a factor of 7 and 2.5, respectively. Earlier studies[13] in Irkutsk have shown the total concentration of 12 PAHs to vary from 25 to 300 ng/m^3 in different districts of the city in winter; the benzo(a)pyrene concentration changed from 0.8 to 30 ng/m^3.

Table 3. Enrichment factors (by Al, Fe, Ti) of aerosol components in samples from the Irkutsk–Cheremkhovo industrial zone

Element	Irkutsk			Angarsk		Usol'e-Sibirskoe		
	Periphery	Center	Industrial zone	Center	Industrial zone	Periphery	Center	Industrial zone
Si	1.2	$1.2 \cdot 10^{-1}$	8.6	$3.0 \cdot 10^{-1}$	–	8.1	$4.3 \cdot 10$	3.4
Ca	$1.1 \cdot 10^{2}$	$8.9 \cdot 10$	7.1	$5.1 \cdot 10$	$1.8 \cdot 10^{2}$	$5.0 \cdot 10$	$2.1 \cdot 10$	$3.1 \cdot 10^{2}$
Al	8.6	7.3	3.5	1.7	1.0	6.4	2.8	4.3
Mg	2.1	3.4	1.7	1.8	7.9	4.0	$1.4 \cdot 10$	4.2
Ti	1.7	2.2	2.0	3.3	9.2	1.5	1.9	1.9
Fe	$1.0 \cdot 10^{-1}$	$1.0 \cdot 10^{-1}$	$1.4 \cdot 10^{-1}$	$1.8 \cdot 10^{-1}$	$1.0 \cdot 10^{-1}$	$1.1 \cdot 10^{-1}$	$1.9 \cdot 10^{-1}$	$1.2 \cdot 10^{-1}$
Mo	–	$9.7 \cdot 10^{4}$	$4.6 \cdot 10^{3}$	–	$2.0 \cdot 10^{5}$	–	–	–
Ag	–	$9.0 \cdot 10^{5}$	–	$4.3 \cdot 10^{5}$	$4.2 \cdot 10^{6}$	–	–	–
Mn	2.0	–	5.7	1.4	$5.4 \cdot 10$	1.8	$1.6 \cdot 10$	4.0
B	$2.1 \cdot 10^{2}$	$2.7 \cdot 10^{2}$	$4.6 \cdot 10^{2}$	–	$6.0 \cdot 10^{3}$	$2.8 \cdot 10^{3}$	$2.1 \cdot 10^{3}$	$2.6 \cdot 10^{2}$
Cu	$2.2 \cdot 10^{2}$	$1.6 \cdot 10^{2}$	$3.4 \cdot 10$	$2.9 \cdot 10^{2}$	$2.1 \cdot 10^{3}$	$7.8 \cdot 10$	$9.4 \cdot 10$	$1.0 \cdot 10^{2}$
Ni	$2.7 \cdot 10$	$7.6 \cdot 10^{2}$	$3.4 \cdot 10$	$2.6 \cdot 10^{2}$	$4.4 \cdot 10^{3}$	$2.6 \cdot 10$	–	$8.2 \cdot 10$
Sb	–	$9.0 \cdot 10^{3}$	$1.8 \cdot 10^{4}$	–	–	–	–	–
Pb	–	–	$1.9 \cdot 10$	$3.1 \cdot 10$	–	$2.3 \cdot 10^{2}$	–	–
V	5.6	5.9	$7.8 \cdot 10^{-1}$	$1.2 \cdot 10$	$1.3 \cdot 10^{2}$	6.9	$2.9 \cdot 10$	9.9
Cr	$4.8 \cdot 10$	8.0	5.7	$1.9 \cdot 10$	$3.3 \cdot 10$	$3.4 \cdot 10$	–	–
Sn	$3.4 \cdot 10^{2}$	$4.3 \cdot 10^{2}$	$5.0 \cdot 10$	$2.9 \cdot 10^{2}$	$2.5 \cdot 10^{3}$	–	–	$7.9 \cdot 10$
Cl^-	$4.6 \cdot 10^{3}$	$3.4 \cdot 10^{3}$	$1.4 \cdot 10^{3}$	$3.7 \cdot 10^{3}$	$2.4 \cdot 10^{4}$	$2.5 \cdot 10^{3}$	$7.0 \cdot 10^{3}$	$1.4 \cdot 10^{3}$
SO_4^{2-}	–	–	$2.5 \cdot 10$	$8.1 \cdot 10$	–	$4.5 \cdot 10^{2}$	$5.4 \cdot 10^{2}$	$2.7 \cdot 10$
NO_3^-	$6.4 \cdot 10^{2}$	$1.6 \cdot 10^{4}$	$2.7 \cdot 10^{2}$	$4.1 \cdot 10^{2}$	$3.5 \cdot 10^{3}$	$2.2 \cdot 10^{2}$	$4.2 \cdot 10^{2}$	$1.5 \cdot 10^{2}$
Na^+	8.3	$1.2 \cdot 10$	2.1	2.4	$4.1 \cdot 10$	$7.9 \cdot 10^{-1}$	$6.5 \cdot 10^{-1}$	3.2
NH_4^+	$1.5 \cdot 10^{3}$	$2.1 \cdot 10^{4}$	$9.0 \cdot 10^{2}$	$1.0 \cdot 10^{3}$	$1.3 \cdot 10^{4}$	$5.9 \cdot 10^{2}$	$1.7 \cdot 10^{3}$	$3.5 \cdot 10^{2}$
K^+	2.1	3.3	$3.9 \cdot 10^{-1}$	$3.6 \cdot 10^{-1}$	4.2	$6.4 \cdot 10^{-1}$	1.6	$5.5 \cdot 10^{-1}$
F^-	–	–	–	–	$3.2 \cdot 10^{2}$	$1.9 \cdot 10$	$5.1 \cdot 10$	$2.1 \cdot 10$

Table 4. PAH concentration levels in urban aerosol (ng/m^3) sampled on 02.26.2004 in Irkutsk Region

PAH	Irkutsk, Akademgorodok	Irkutsk, Novo-Lenino	Angarsk, industrial zone	Angarsk, residential area	Usol'e-Sibirskoe, industrial zone
Phenanthrene	1.1	15.0	15.8	12.6	16.8
Anthracene	0.14	1.4	< 0.001	0.53	1.2
Fluoranthene	3.1	10.2	14.4	6.7	23.9
Pyrene	2.4	7.3	7.9	3.8	13.8
Benz(a)anthracene	1.2	4.7	0.7	< 0.001	2.3
Chrysene	2.3	4.8	1.6	< 0.001	4.6
Benzo(b)fluoranthene	2.6	5.7	1.2	1.6	4.2
Benzo(k)fluoranthene	0.21	7.3	0.94	< 0.001	3435
Benzo(e)pyrene	2.2	4.0	0.87	0.75	2.6
Benzo(a)pyrene	**0.31**	**7.2**	**0.58**	**0.53**	**2.7**
Perylene	0.29	1.3	< 0.001	< 0.001	<0.001
Indeno(1,2,3-c,d)pyrene	2.8	9.7	1.3	1.2	4.0
Dibenzo(a,h)anthracene	0.6	< 0.001	< 0.001	< 0.001	< 0.001
Benzo(g,h,i)perylene	2.4	8.9	1.4	1.7	3.3
Total PAH	**21.6**	**87.5**	**46.7**	**29.4**	**82.7**

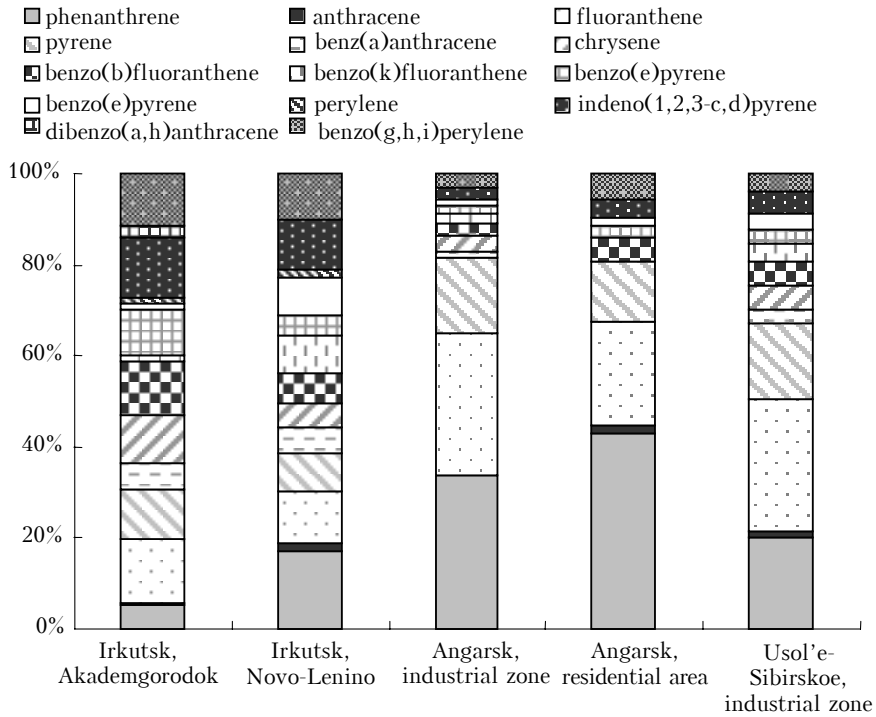

Fig. 3. Percentage of individual PAHs in urban aerosol sampled in Irkutsk region.

According to Roshydromet data,[18] annual average benzo(a)pyrene values exceeded MPC by a factor of 6.5 in Irkutsk, 6 in Angarsk, and 2.8 in Usol'e-Sibirskoe in 1996, while in 2002 they increased in Irkutsk and Usol'e-Sibirskoe up to 10.4 and 6.2 of MPC, respectively. Only in Angarsk they decreased down to 2.8 of MPC.[17] Such dynamics well agrees with our measurement data.

The PAH concentrations and the percentage depend on sampling sites, i.e., location of pollutant sources. Among the identified PAHs in Angarsk, phenanthrene, fluoranthene, and pyrene prevail in public green spaces and industrial zones; their total amount reaches 80% of the detected PAHs amount (Fig. 3). Similarity in the PAH percentage in aerosol samples from these zones points out to pollutant transport from the industrial zone to the residential one. Akademgorodok and Novo-Lenino in Irkutsk are characterized by a high content of benzo(g,h,i)perylene and indeno(1,2,3-c,d)pyrene, which are indicators of air pollution by vehicles.[19] In Novo-Lenino, the main PAH sources are medium- and low-capacity heating stations using coal spreader stokers, as well as private houses with stove heating.[20] High benzo(a)pyrene content in aerosol samples from this district indicates an incomplete burning of fuel.

Conclusion

The performed investigations show that formation of aerosol fields in towns of a large industrial region is of a complicated character and is a superposition of several transport mechanisms and sources both distant and local. The ratio of their contribution can vary significantly in different parts of such a zone.

For the present, a lack of parallel measurement data on different aerosol fractions in hand makes impossible an in-depth analysis of correlations between them and, as a consequence, an overall characterization of urban anthropogenic aerosol.

However, the comparison of total concentrations of aerosol macro- (inorganic fraction) and microcomponents (PAHs) (see Fig. 1 and Table 4) shows a significant correlation. Aerosol macrocomponents can well serve as a collecting matrix for the organic fraction. Being of a definitely industrial anthropogenic origin, microcomponents are evidently a peculiar kind of a tracer in aerosol transport between different districts.

Hence, our complex mobile measurements show themselves as highly valuable and useful in rapid and simultaneous acquisition of data on chemical composition and other parameters of pollutants, which can serve a foundation for model investigations in cities.

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