STRUCTURE OF AEROSOL "CAPS" OVER INDUSTRIAL CENTERS

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The vertical and horizontal distribution of atmospheric aerosol over some cities of Russia and Kazakhstan is analyzed based on the data of airborne sounding. It is shown that the aerosol cap over industrial center is 400 *to* 600 m *thick and is completely determined by the height of the internal mixing layer. Its horizontal length seems to be tens of kilometers. The aerosol mass concentration in the cap is* 2 *to* 8 *times greater than that in the near-ground layer. The enrichment in same components inside the cap can reach* 350 *times. Chemical composition of aerosol essentially differs from the near-ground one. Then, it is an individual element for each city, which is characteristic of it. In addition to the peculiarities of chemical composition of emissions, such an element can be a certificate of the industrial center, when studying the transboundary transport of air pollution.*

Modern industrial center with a lot of sleeping blocks and technological objects concentrated on its territory is an island of heat because significant amount of heat is emitted to the air on a limited area. Nonuniform distribution of temperature over it, together with orography and, as a rule, the presence of big reservoirs, leads to the formation of local circulation of air over the city. That favors the accumulation of admixtures and the appearance of the "cap" of pollution over it.[1,2]

Because of apparent simplicity of this phenomenon (sometimes the cap is seen visually) it is poorly studied. However, without considering the peculiarities in formation of the cap of admixtures, the forecast of the level of air pollution in a city is low justified, and it is difficult to establish the suburb area affected by emissions of a city, and the environmental protection measures may be low effective. There are more questions than answers in not so numerous papers devoted to the study of the admixtures distribution over industrial centers.[3–6] In particular, it is not known to date what are the horizontal and vertical size of the caps of admixtures, what is their chemical composition, what is the enrichment in the elements in the cap in comparison with the background or near-ground air in the city, how admixtures are distributed in vertical and horizontal directions, etc. Moreover, specialists from environmental protection agencies very often make a mistake being sure that the composition of air pollution in the cap has only low effect on the level of air pollution in the near ground air.

Nevertheless, even general information shows that the role of aerosol cap is very important. First, the layer of admixtures over city attenuates the inflow of solar radiation thus changing the radiation balance and, hence, thermal regime on its territory. Second, the spectrum of solar radiation can be essentially changed depending on the composition of air pollution in the cap.[7,8] Then, as a rule, the ultraviolet radiation, active in photosynthesis, is strongly absorbed. Third, the photochemical processes usually are intense in the cap. The secondary, more toxic than the initial, emissions are produced from the primary ones as a result of these processes.[9] Fourth, large aerosol particles and gases with great molecular weight fall out from the cap on the territory of a city, and they directly affect the composition of air near the ground.[10,11]

Naturally, the contributions from each of the above factors vary over different cities and depend on industrial enterprises on their territory, on the size of cities and their orography, climatic characteristics of the zones where they are situated, the presence of big reservoirs near cities, etc.

This paper presents some results of the study of the aerosol cap structure over the cities in Russia and Kazakhstan (Table I). The data were collected with the airborne laboratory AN-30 OPTIK-E[12] using the techniques described in Ref. 13.

Since mainly the analysis was carried out using relative characteristics (all vertical distributions were normalized to the near-ground value of concentrations), Table II shows the near-ground concentrations of chemical components, what makes it possible to pass to the absolute values.

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TABLE I.

City	Period	Number of samples at the altitudes							
		0	200	400	600	800	1000	1200	1400
Pavlodar	13–20.03.1990	24	27	29	32	16	16	11	4
Ust'-Kamenogorsk	03–10.04.1990	38	6	42	38	12	11	5	2
Khabarovsk	20–30.06.1990	18	40	41	38	21	9	8	6
	10–19.12.1990	16	32	28	31	12	7	6	5
Nizhnii Tagil	01–06.11.1990	25	28	29	16	7	4	2	2
Komsomol'sk-on-Amur	10–19.12.1990	24	12	37	36	19	8	6	4
Nizhnevartovsk	19–29.08.1991	37	42	43	48	37	23	8	6
	24.11–03.12.1991	26	32	30	35	27	14	6	5

Let us first consider the mean vertical distribution of the aerosol mass concentration over some cities. As is seen from Fig. 1, the concentration can exceed the near-ground concentration by 2.6 (Ust'-Kamenogorsk) to 10.8 times (Komsomol'sk-on-Amur). The altitude of the maximum concentration varies from 200 to 400 m. The above threshold of the cap is quite well seen and is at the altitude of 400 to 600 m, varying within this limits over different cities. Then, the aerosol cap over the city follows the dynamics of the internal mixing layer, which, according to the data obtained earlier,[14] is within these limits.

The sounding over cities of Khabarovsk and Nizhnevartovsk was carried out twice, in winter and in summer. That makes it possible to estimate the seasonal dynamics of the aerosol cap. As is seen from Fig. 2, the altitude of the maximum concentration increases from winter to summer, as the ratio of the near-ground concentration to that in the maximum changes. Thus, the maximum over the city of Khabarovsk rises from 200 to 400 m, and over Nizhnevartovsk it rises from 600 to 800 m. The ratio of concentration in the cap to the near-ground one in Khabarovsk decreases from winter to summer, while in Nizhnevartovsk this ratio increases, and the secondary maximum appears at 400 m altitude.

Such a picture reflects the geographical peculiarities of the positions of these cities. In particular, the oil deposits Megion and Samotlor are situated near the city of Nizhnevartorsk, with their plumes burning the hydrocarbons accompanying the oil extraction.

According to the existing conceptions, the aerosol cap is formed as a result of superposition of several mechanisms. They are the emissions from the motor transport and low-altitude sources within the internal mixing layer; the aerosol-forming substances evaporated from the ground surface, which are transformed into particles in the atmosphere; and, finally, the plumes from sources at high altitudes (higher than 100 m) such as power plants, plant stacks, etc., that emit gases and aerosol directly to the cap. And, if over all cities the increase in aerosol concentration begins from the ground surface, Figs. 1 and 2), then in Nizhnevartovsk the concentration is less at the altitude of 200 m than near the surface and in the cap of pollution both in winter and in summer (Fig. 2*b*). This is indicative of the fact that elevated sources, among which are the oil deposit plumes, have higher power than sources situated on the city territory, and that the cap is formed here in a different way.

In general, although the aerosol cap changes its structure from winter to summer, it remains in the above altitude range.

According to the data from Ref. 15, the following elements are emitted into the atmosphere when burning coal: up to 10% of Al, Co, Fe, Mn, and Sc; up to 30% of Cr, Cu, Ni, and V; and up to 100% of As, Br, Hg, Sb, and Se. The gas component of these elements in the emissions of metallurgical plants is up to 30% for Cr, Se, Br, and Cd; up to 70% for As; and up to 100% for Hg. The emission of Pb is mainly from the motor transport.

It is seen from Fig. 3 that, similarly to the mass concentration, the maxima in the content of the majority of chemical compounds are also observed in the cap of pollution within the layer from 200 to 600 m. The exception is the city of Nizhnevartovsk in summer, where the maxima of the ratios of concentration that reach the values of 53 and 62 for SO_4^{2-} and Hg^{2+}, respectively, are observed at the altitude of 800 m.

One should pay attention to the fact of high values of the enrichment coefficients of some ingredients in the cap, in comparison with the mass concentration. The value C_H/C_0 for the mass concentration is in the range 2 to 8, while it is 113, 53, 62, and 350 for Br^-, SO_4^{2-}, Hg^{2+}, and Cd^{2+}, respectively, in the city of Nizhnevartovsk in summer; 31, 32, 44, 86, and 32 for Br , Hg^{2+}, Si, Mn, and Ti, respectively, in the city of Pavlodar; 26 for Fe in Khabarovsk in summer; and 35 for Br^- in Komsomol'sk-on-Amur.

Thus, the data on vertical distribution of aerosol chemical components in the cap of pollution are in a good agreement with the conclusions drawn in Ref. 15.

However, there are some deviations from the general picture mentioned above. Some of them are shown in Fig. 4.

Table II. Mean concentration of aerosol chemical components (μg/m³) near the ground (C_0), to which the values at other altitudes (C_H) were normalized.

City and season	Na^+	K^+	Cl^-	Br^-	NO_3^-	NH_4^+	SO_4^{2-}	Hg^{2+}	As^{5+}	Zn^{2+}	Cd^{2+}	F^-	Si	Fe	Mn	Mg	Pb	Cr	Ni	Al	Ti	Ca	Cu	V
Pavlodar, spring	2.58	0.97	3.60	0.30	0.79	1.78	1.17	0.035	1.230	0.030	–	–	0.27	0.46	0.006	0.060	–	0.057	0.020	5.70	0.044	0.80	0.050	0.001
Ust'-Kamenogorsk, spring	1.84	1.85	5.82	1.58	0.53	1.30	–	1.270	0.040	0.039	–	0.65	3.56	0.70	0.003	0.003	0.005	0.007	–	–	0.780	0.45	0.014	0.006
Khabarovsk, summer	2.75	3.87	6.15	2.42	0.66	4.59	1.90	0.090	0.163	0.125	0.020	1.08	4.57	0.79	0.042	0.340	0.004	0.025	0.012	2.53	0.473	1.99	0.050	0.129
Khabarovsk, winter	1.50	0.57	13.20	1.20	0.86	1.20	0.91	0.010	0.070	0.070	0.006	8.20	29.20	9.80	0.130	0.670	0.006	0.100	0.100	4.71	0.180	7.26	0.240	0.001
Nizhnii Tagil, fall	1.90	1.04	5.83	8.14	2.98	0.59	–	0.100	0.460	0.060	0.040	0.12	–	4.90	0.090	0.610	–	0.100	0.060	1.40	0.140	26.63	0.040	0.003
Komsomol'sk-on-Amur, winter	2.17	0.42	2.85	3.24	0.25	–	2.68	–	0.100	0.138	0.046	1.15	29.20	1.08	0.150	0.400	0.001	0.098	0.273	1.61	0.100	3.15	0.238	0.003
Nizhnevartovsk, summer	1.07	2.68	11.70	1.10	0.40	0.10	0.80	0.004	0.010	0.200	0.002	2.09	17.32	6.7	0.300	20.400	0.080	0.270	0.010	10.10	0.280	26.09	0.070	0.007
Nizhnevartovsk, winter	0.63	0.57	22.75	1.20	0.43	–	0.16	–	–	0.060	0.008	3.10	0.41	0.74	0.003	0.48	0.030	0.110	0.010	0.82	0.300	5.42	0.050	0.006

Note: Dash means that the element was not determined or its concentration was less than the detection threshold.

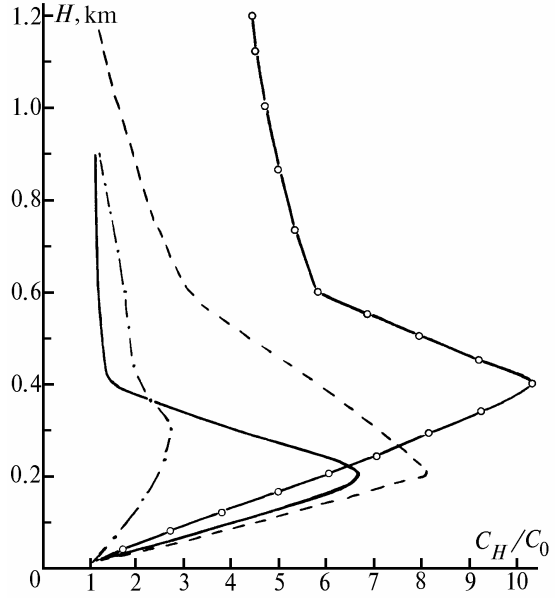

FIG. 1 Normalized vertical distribution of aerosol over: Khabarovsk (winter) (–); Pavlodar (spring) (– –); Ust'-Kamenogorsk (spring) (– • –); Komsomol'sk-on-Amur (winter) (°––– °).

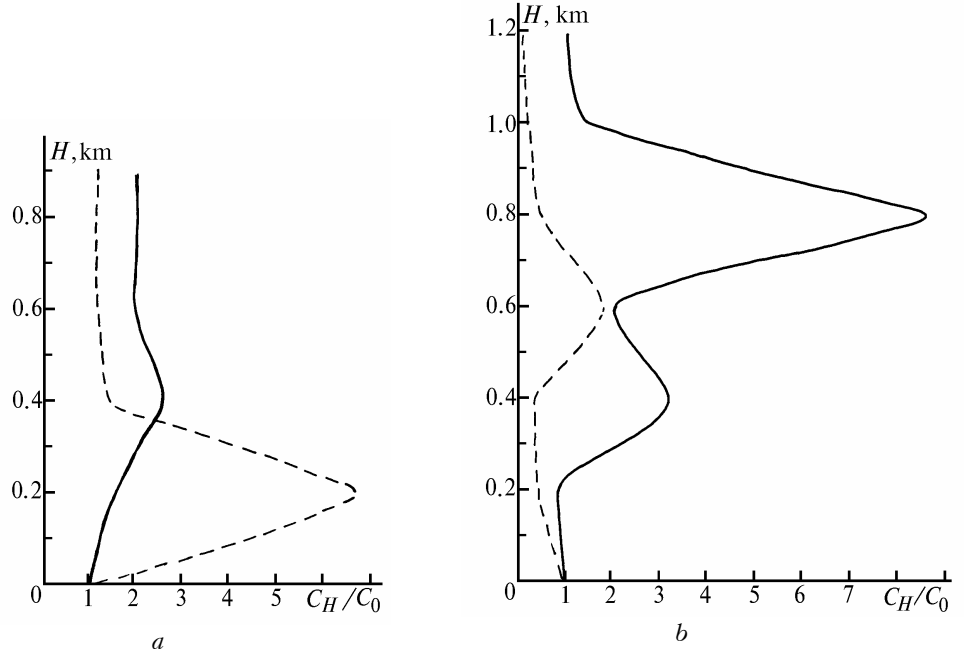

FIG. 2. Normalized vertical distribution of aerosol over: Khabarovsk (a); Nizhnevartovsk (b); summer (–), winter (– –).

The first deviation is caused by the presence of "foreignB plumes over all cities, over which the flights were performed. These are the plumes coming to the territory of the city from outside. They give the additional source of chemical components that are not detected in the local emissions of the city. This source contributes to the cap formation from the above. Such a vertical distribution is shown in Fig. 4. Figure 4*a* shows the presence of both "ownB sources of As^{5+}, K^+, Cu, Cr, and Ni forming the cap over the cities of Khabarovsk, Nizhnevartovsk, and Komsomol'sk-on-Amur, and the foreign ones, coming from outside. Figure 4*b* gives the evidence of the fact that sometimes the external source is so powerful that it creates an inverse distribution of elements over altitude. Such a distribution was observed in Khabarovsk, Komsomol'sk-on-Amur, and Ust'-Kamenogorsk for Cd^{2+}, Cl^-, Cu, F^-, and NH_4^+. The trajectory analysis has shown that the plume came to Komsomol'sk-on-Amur from Amursk, to Khabarovsk from the China Republic, and to Ust'-Kamenogorsk from Semipalatinsk and Pavlodar.

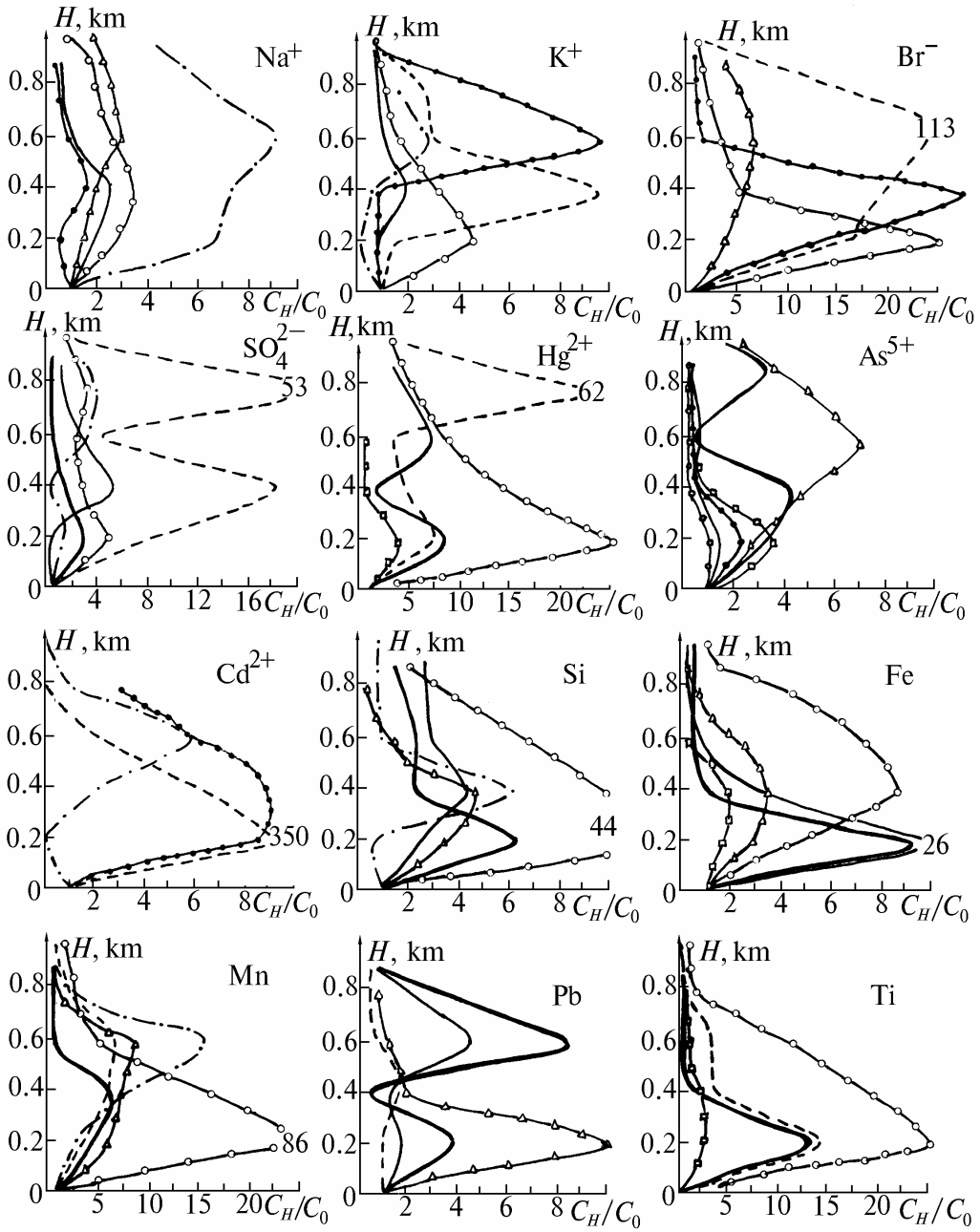

FIG. 3. Normalized vertical distribution of aerosol over: Khabarovsk (summer) (—); Khabarovsk (winter) (– – –); Nizhnevartovsk (summer) (– – –);Nizhnevartovsk (winter) (–·–); Ust'-Kamenogorsk (Δ–– Δ); Pavlodar (°–– °); Komsomol'sk-on-Amur (•––•); Nizhnii Tagil (⌒––⌒).

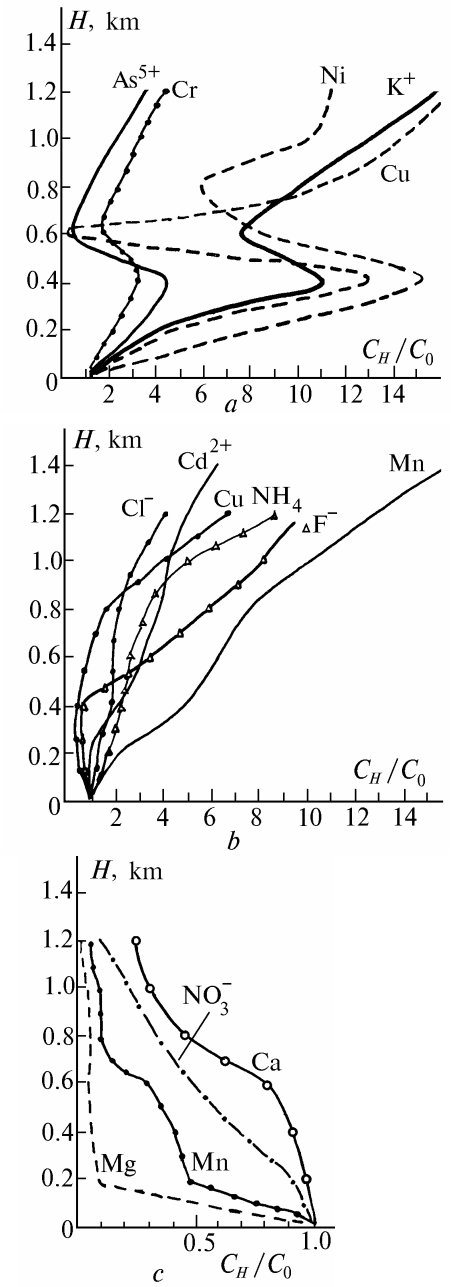

FIG. 4. Designations are the same as in Fig. 3.

In our opinion, the data presented in Figs. 4*a* and *b* also answer the question that often arises in mind of specialists when carrying out the near-ground monitoring. The question is from where the elements, that are not present in local emissions, appear in the composition of suspended substances.

And, finally, Fig. 4*c* shows the situation when such chemical components as Ca, Mg, Mn, and NO_3^- in Pavlodar, Nizhnevartovsk, and Komsomol'sk-on-Amur are mainly generated near ground, and the power of near-ground emissions of these substances here is much greater then the intensity of the sources situated above. Let us emphasize that there are only four cases among all cities and substances listed in Tables I and II, when the concentration of elements near the ground surface is greater than that in the cap of pollution.

Thus, based on the data shown in Figs. 1–4 one can unambiguously conclude that at least periodic control of admixtures is necessary just over a city.

It follows from the aforementioned that the aerosol mass concentration and the concentration of some chemical components in the cap is higher than that in the near-ground air. Regularly the question arises on what is the relation between the chemical composition of particles in the cap and in the near-ground air. Such data (%) are shown in Fig. 5 for five cities. The chemical composition of aerosol near the ground surface (H = 0) is compared with that at the level of the maximum concentration determined from Fig. 1. The sector with the sum of elements listed in Table II and having the concentration of each less than 1% is hatched.

It is seen from Fig. 5 that the water-soluble photochemical fraction in the near-ground layer of Khabarovsk (F^- and CL^-) is about one quarter of the total mass, and the terrigenic (soil) fraction is about three quarters. This ratio changes at the level of the maximum concentration. The photochemical component disappears almost entirely at the altitude of 200 m. Terrigenic component is about 90%. Such a result is a little bit unexpected, because it is the city where the greatest intensity of photochemical processes[9] is observed at the altitude of 200 m. Two explanations of this fact are possible. First, the products of photochemical reactions quickly fall out on the surface, that was noted in Ref. 9. Second, the principal sources of emissions to this height burnt the coal with high content of ashes. According to the data from Refs. 16–18, it can lead to the fact that the mass of mineral fraction will dominate in the total concentration of the emission.

The situation similar to the described is observed in the city of Pavlodar (Fig. 5). The difference is that the soil fraction is represented here by aluminum. Its contribution is about 25% near the surface, and about 50% at the altitude of 200 m. In contrast, the photochemical component near the ground is about three quarters of the total mass, and at the altitude of 200 m it is slightly greater than one quarter. Let us note that the enterprises using coal with great ashes content in Pavlodar also give the principal mass of elevated emissions.

The variation of aerosol chemical composition with increasing altitude up to the altitude of the maximum concentration in the cap of pollution over Komsomol'sk-on-Amur, Nizhnevartovsk and Nizhnii Tagil has an opposite behavior in comparison with those in Pavlodar and Khabarovsk (Fig. 5). The photochemical component near the ground surface is one quarter, and in the cap its contribution increases up to 60–70%. It is explained by the fact that there are big metallurgical and chemical plants on the territory of these cities, that emit the basic substances for the photochemical component.

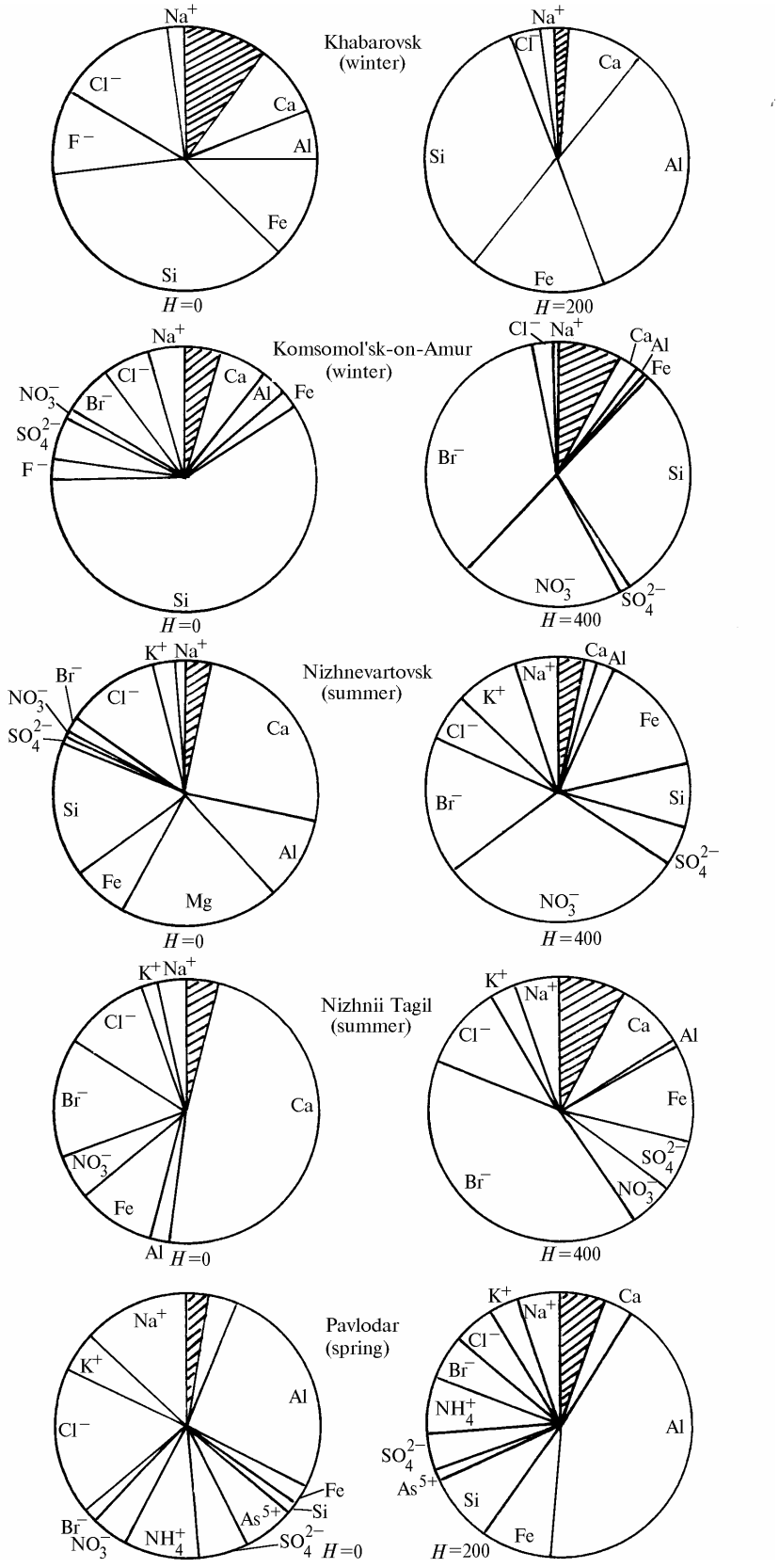

FIG. 5. Aerosol chemical composition (%) near the ground surface and in the maximum of concentration in the cap.

One also should pay attention to the fact that follows from the analysis of Fig. 5. As a rule, there is an element of terrigenic origin, the contribution of which to the total mass is dominating, in the composition of near-ground aerosol in each city. It is Si for Khabarovsk and Komsomol'sk-on-Amur, Ca for Nizhnii Tagil, and Al for Pavlodar. Obviously, such a monoelement picture reflects the peculiarities of the soil chemical composition characteristic of the specific city. It is seen from Fig. 5 that the content of this element can decrease or increase in the cap of admixtures over city. Nevertheless, in addition to the data on chemical composition of emissions, separation of such a monoelement can be a certificate of a city when studying the transboundary transport of pollution.

Comparison of the data presented in Figs. 3–5 shows that the chemical composition of aerosol in the cap and in the near-ground layer is different not only quantitatively, but also qualitatively. It additionally strengthens the thesis on the necessity of monitoring the admixtures in the cap over the city. The matter is not only in high toxicity of many components observed in the caps of cities.

According to the data obtained earlier[9–11] these components fall out to the near-ground layer over the territory of the city. In addition, the peculiarities of air circulation in the vicinities of the city favors the fact that the air pollution is not transported from its territory, but is accumulated over it.[1,2,13] So, only the near-ground monitoring will not give a complete information on air quality in different parts of the city.

Data on the horizontal structure of aerosol cap over city are very contradictory. The numerical simulation[1,2] shows that it is significantly greater than the territory of a city. It was noted in Ref. 3 that the aircraft passed through the cap during 50 to 60 sec. The size of a cap can be 4 to 12 km. The investigation we have carried out confirms that it is really greater than the city territory.[9–11,19,20] However, without knowing the results of Ref. 2 we could not plan the experiment[9–11,9,20] correctly. So the maps we have drawn show only the core, but not the whole aerosol cap. Analysis of these maps shows that the horizontal structure of the cap is, as a rule, inhomogeneous. The size of inhomogeneities is approximately within the range mentioned in Ref. 3. They vary from 0.8 to 9 km depending on a city. The plumes are excluded from the sample here. Then in this paper we do not present the data on horizontal structure of aerosol caps. On the one hand, the maps of horizontal distributions of aerosol number density in the cap core are already published.[9–11,13,14,19,20] On the other hand, total horizontal size of these caps is unknown.

The horizontal inhomogeneity of the distribution of aerosol chemical components out of the cap and in its core is shown in Fig. 6 that presents an idea of enrichment in the elements composing the particles over Nizhnii Tagil.

Figure 6 used the data of two flights. One of them was performed on the windward side of the city, in clear air, at the altitude where the aerosol concentration in the cap was maximum. The second flight crossed the core of the cap of pollution at the same altitude. The column "exitB is the difference between the concentration of each element and the total one obtained from the results along both paths. The data are presented in absolute units, because the majority of elements in air coming to the city had the concentration less than the detection threshold or at the trace level. So the normalizing similar to that done for the vertical distributions should give the infinite value for some substances.

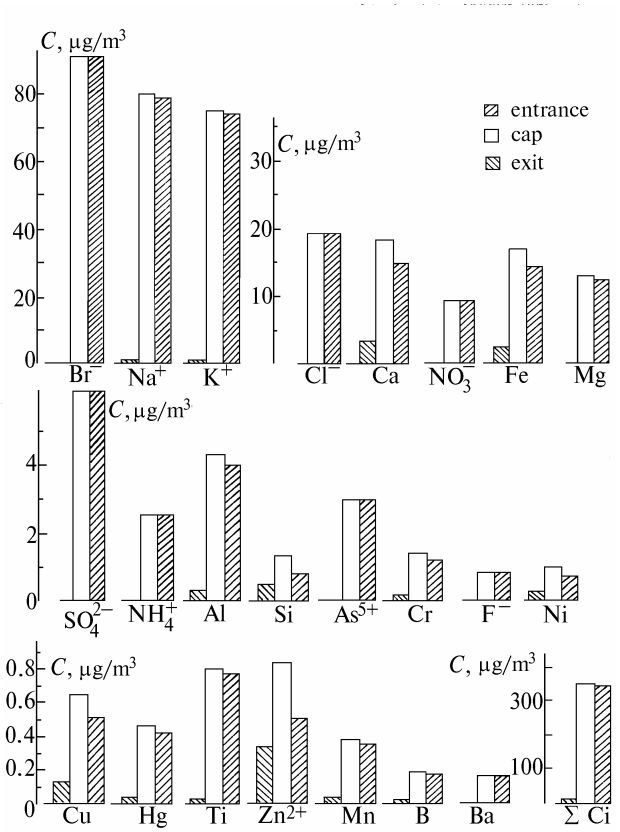

FIG. 6. Concentration of aerosol chemical components on the windward side of Nizhnii Tagil, in the cap core, and their difference.

It is seen from Fig. 6 that aerosol mass concentration in the cap core is 150 times greater than that at the windward side of the city. The enrichment in the ions Br^-, Na^+, and K^+ in the cap reaches 700 to 1200 times. Much lower enrichment is characteristic of only terrigenic elements Ca, Fe, Al, Si, Cu, and Zn.

Thus, the horizontal contrasts of aerosol concentration between the cap over city and the clean air is much greater than along the vertical direction, that is understandable. The particles are generated by the low sources in the near-ground layer on the city territory, and the aerosol aged in the cap falls out here. The vertical contrasts decrease resulting from the action of both mechanisms.

In conclusion let us sum up the results. The aerosol cap over city that appears due to superposition of emissions in the near-ground layer and from the elevated sources has the vertical size of several hundred meters (400 to 600 m) and the horizontal size of several ten kilometers. Its altitude and dynamics are determined by the level of internal mixing layer. It has inhomogeneous structure along the vertical and horizontal directions and is essentially different from the near-ground aerosol in its chemical composition.

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