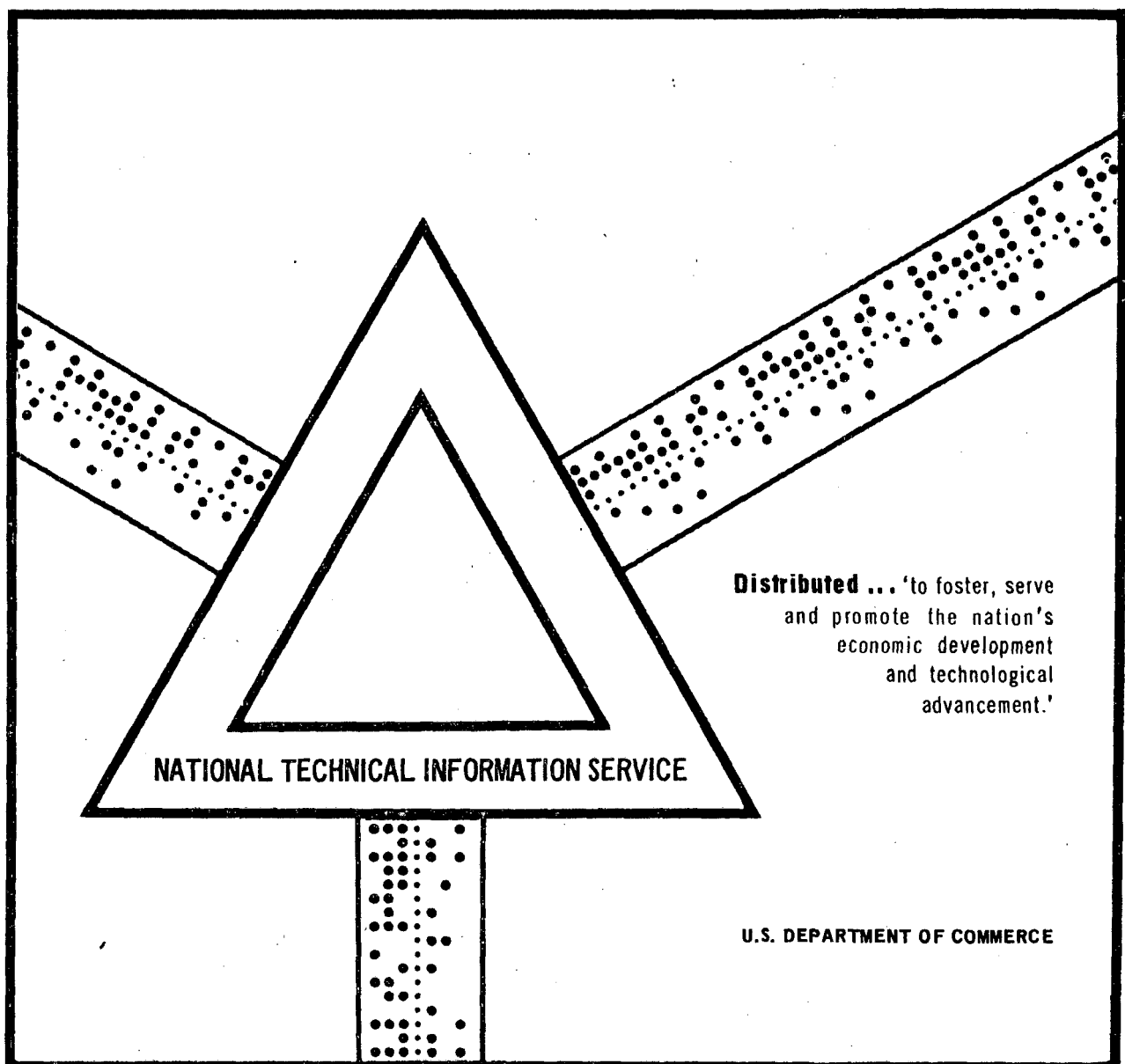


AICE SURVEY OF USSR AIR POLLUTION LITERATURE.  
VOLUME IV. METEOROLOGICAL AND CHEMICAL  
ASPECTS OF AIR POLLUTION; PROPAGATION AND DIS-  
PERSAL OF AIR POLLUTANTS IN A NUMBER OF AREAS  
IN THE SOVIET UNION

M. Y. Nuttonson

American Institute of Crop Ecology  
Silver Spring, Maryland

December 1969



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A RESEARCH ORGANIZATION OF THE PROBLEMS OF  
FARM PRODUCTION AND PROTECTION

AIR SURVEY OF U.S. AIR POLLUTION LITERATURE

Volume IV

METEOROLOGICAL AND CHEMICAL ASPECTS OF AIR POLLUTION  
PROPAGATION AND DISPERSAL OF AIR POLLUTANTS IN A NUMBER OF AREAS  
OF THE SOVIET UNION

edited by

M. Y. Nudberg

The material presented here is part of a survey of  
USSR literature on air pollution,  
conducted by the Air Pollution Section  
AMERICAN INSTITUTE OF CROP SCIENCE

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## PREFACE

Much of the background material presented in the prefaces to the preceding volumes of this series is repeated here in view of its relevance to the present volume.

Contamination of the natural environment constitutes a major problem in all industrial regions of the Union of Soviet Socialist Republics (USSR). The country's industry and transport are continually bringing about massive qualitative changes in the habitat of man and vegetation through an ever increasing pollution of air, soil, and streams. In recent years there has been a greater awareness of the immense problems of air and water pollution on the part of the urban and rural administrative agencies as well as on the part of various research institutes of the USSR. There is a mounting demand there to maintain a high quality physical environment. Protective measures against the pollution threat are gradually taking shape. Much relevant air pollution research data are being developed and are apparently put to good use in some parts of this vast and diverse country.

The behavior of atmospheric contaminants, notably gases and fine particles discharged into the air, is similar to that of the air masses near the surface of the earth -- the distribution of the contaminants being influenced by atmospheric stability, wind, precipitation, and topographic features of a given area or region. The most outstanding and dominant characteristic of the atmosphere is its unceasing change, a change resulting from variations of temperature, wind, and precipitation. These meteorological conditions vary widely as a function of latitude, season, and topography. Seasonal as well as diurnal temperature gradients, horizontal and vertical, affect the speed of the wind flow. Generally, the greater the wind velocity the more rapid is the dispersion of pollutants in the atmosphere. In continental areas the temperature gradients and the consequent wind flow increase during the winter season and during the daytime periods, the latter being usually subject to more turbulent winds of higher velocity than those that prevail during night hours that are typically characterized by low-level stability with a minimum dispersal and dilution of the pollutants.

Studies of atmospheric diffusion and air pollution constitute a rapidly developing area of meteorological sciences in the USSR. Determination and analysis of the complex set of meteorological factors causing the processes of atmospheric diffusion are being extensively developed there in conjunction with theoretical and experimental studies of the pattern of propagation of contaminants in the atmosphere.

Most of the reports of investigations brought together in this volume deal with meteorological conditions and relief as factors in propagation and dispersal of air pollutants in a number of areas in the Soviet Union. A number of the papers of this volume deal also with the chemical composition of air pollution and methods for determination of some toxic impurities

of the air. A considerable number of these investigations have been conducted in various industrial regions of the USSR, regions that are geographically far apart from each other and subject to distinctly different natural and man-made environmental conditions.

Some of the material presented here deals with various noxious pollutants emitted to the atmosphere in high concentration at or near ground level and with the exposure of these pollutants to the continuous mixing, diffusion, stirring, and dilution that take place between regions of the atmosphere as a result of air turbulence. A number of papers deal with the intensity and structure of air turbulence in relation to temperature and wind, which form the background of atmospheric diffusion and stirring. Other papers deal with the direction frequencies and intensities of wind, which differ markedly for stable and unstable conditions of atmosphere; with the extremely slow diffusion through an inversion; and with the general climatology of atmospheric turbulence, diffusion, and the dispersions of air pollutants in different parts of the country and during different seasons of the year. The effect of rain on air pollution levels is also discussed.

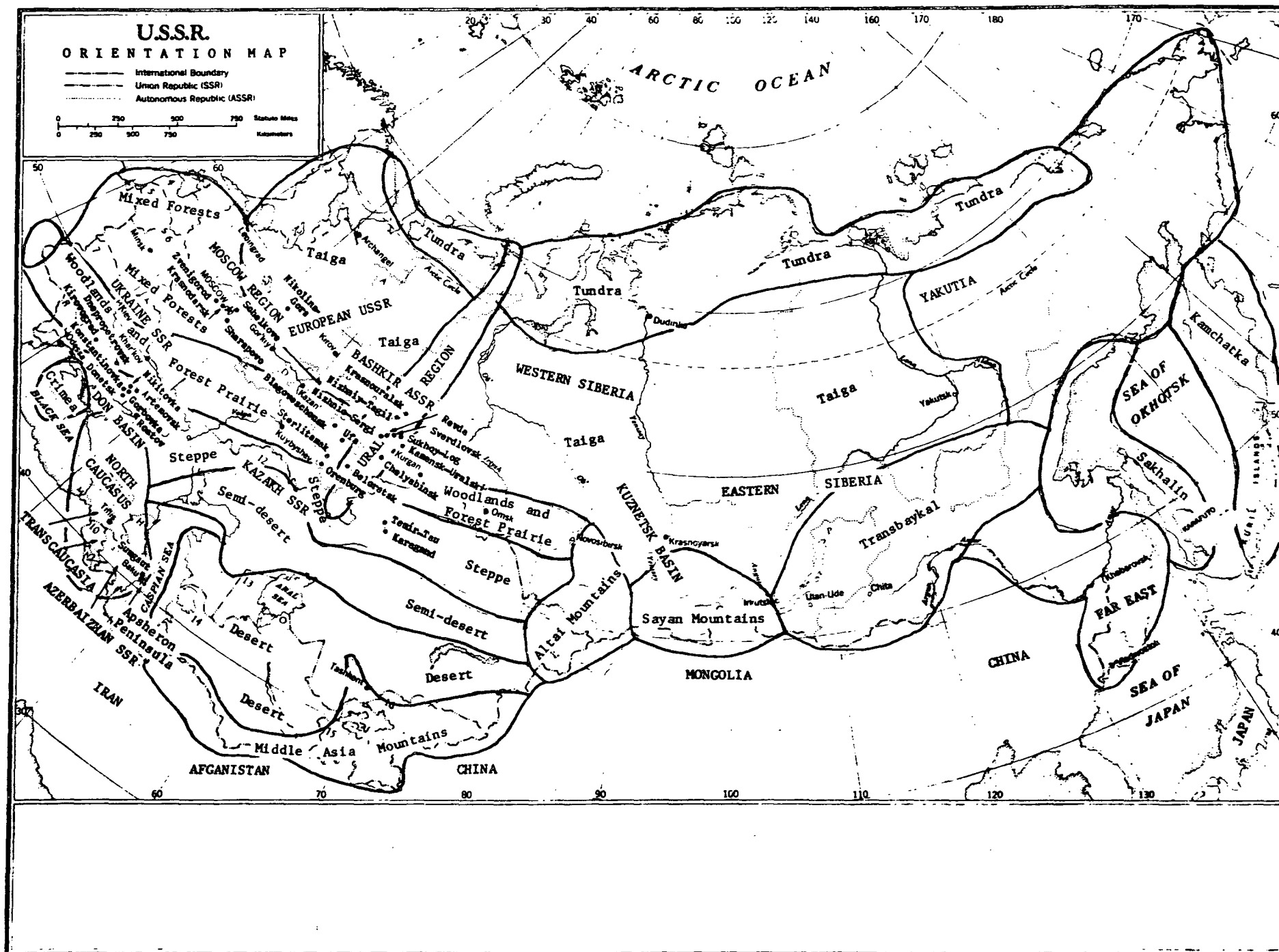
It must be borne in mind that the data presented in this volume relate to many diverse environments in a vast land area; that the USSR extends for about 7,000 miles from west to east and 3,000 miles from north to south; and that the country covers a wide range of climatic and relief conditions throughout much of its north-south and west-east extent. In this connection, a brief outline of the very general natural features of the USSR may be desirable. Lowlands and plains dominate the landscape of the major portion of the country. Its landscape can be roughly described as one consisting of broad latitudinal climatic belts of the lowlands and plains and of narrow, vertical climate zones of the highlands and mountains. Each of the broad latitudinal belts is distinct from the other in the major features of its climate, vegetation, and soils, though within each latitudinal belt there is a decrease in the annual precipitation as one proceeds from west to east. The latitudinal belts include the nearly barren and treeless tundra in the extreme north, where the winters are severe, the summers, short and cool, and where precipitation is very limited. There follow the belts of the taiga or coniferous forests, mixed forests, woodlands, forest prairie or forest steppe, the steppe, and the semi-desert. Finally in the extreme south, east of Caspian Sea, there are the dry deserts, hot in summer and cold in winter, and, along the southern reaches of the Black Sea in Transcaucasia, there is a relatively limited area, humid and more or less subtropical, which is subject to mild winters, hot summers, and heavy precipitation.

It is hoped that the papers selected for presentation in this volume will permit an assessment of some of the USSR studies of the meteorological, chemical, and topographic aspects of air pollution. As the editor of this

volume I wish to thank my co-workers in the Air Pollution Section of the Institute for their valuable assistance. Special thanks are due to Adam Peiperl who, as one of the principal translators, carried much of the load of this project.

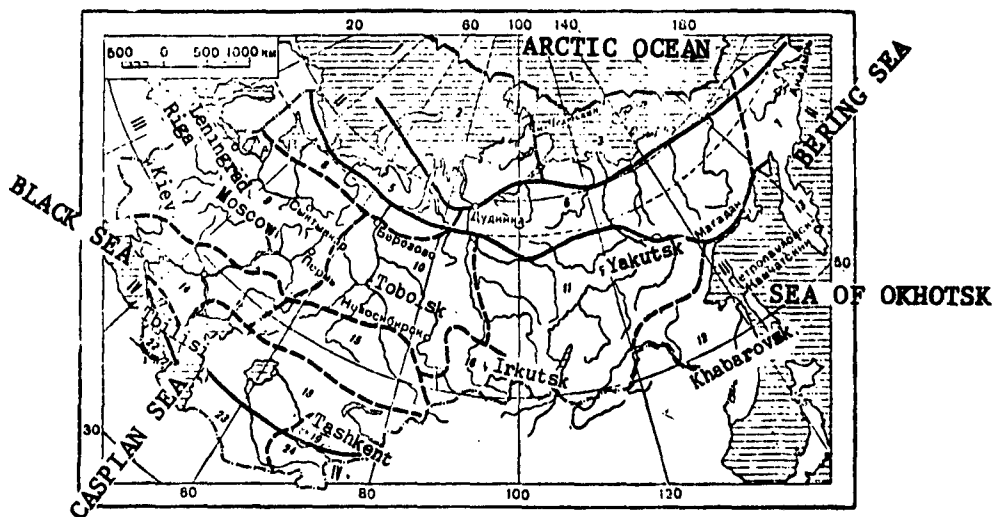
M. Y. Nuttonson

Silver Spring, Maryland  
September 1970





# CLIMATIC ZONES AND REGIONS\* OF THE USSR



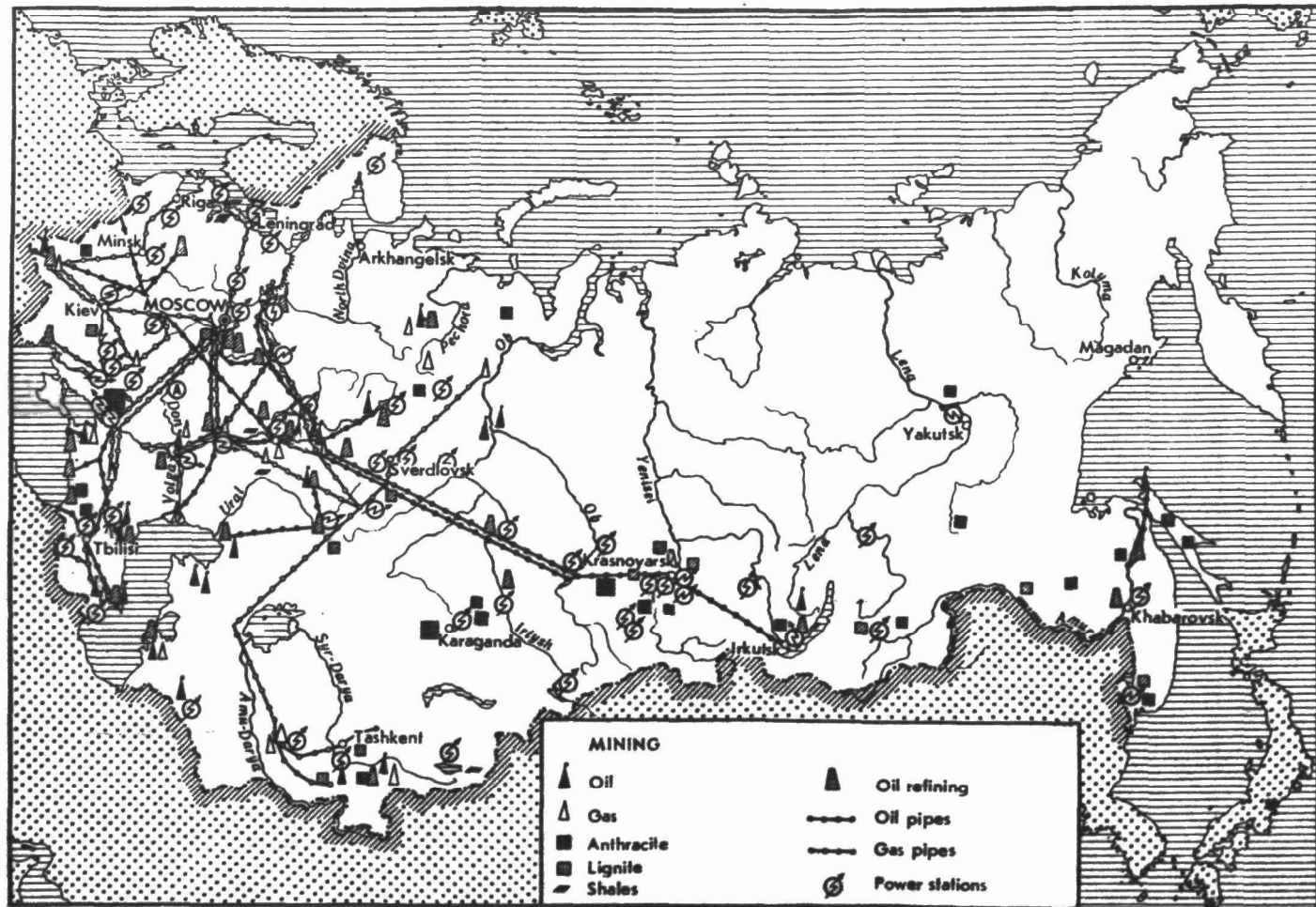
Zones: I-arctic, II-subarctic, III-temperate, IV-subtropical  
Regions: 1-polar, 2-Atlantic, 3-East Siberian, 4-Pacific, 5-Atlantic, 6-Siberian, 7-Pacific, 8-Atlantic-arctic, 9-Atlantic-continental forests, 10-continental forests West Siberian, 11-continental forests East Siberian, 12-monsoon forests, 13-Pacific forests, 14-Atlantic-continental steppe, 15-continental steppe West Siberian, 16-mountainous Altay and Sayan, 17-mountainous Northern Caucasus, 18-continental desert Central Asian, 19-mountainous Tyan-Shan, 20-western Transcaucasian, 21-eastern Transcaucasian, 22-mountainous Transcaucasian highlands, 23-desert south-Turanian, 24-mountainous Pamir-Alay

\* After B. P. Alisov



# THE MAIN MINING CENTERS OF THE USSR

11



(After A. Efimov, "Soviet Industry", Moscow 1968)



METEOROLOGICAL CONDITIONS IN RELATION TO THE FORMATION OF  
PERIODS OF HEAVY AIR POLLUTION IN CITIES

L. R. Son'kin and T. P. Denisova

From Trudy, Glavnaya Geofiz. Observat. im. A. I. Voeykova, No. 238  
p. 33-41, (1969)

Analysis of observational data on air pollution in a number of cities of the Soviet Union has shown that on certain days, a simultaneous rise in the impurity concentration is observed in different parts of a city. This effect has been discussed in general terms in our earlier studies [9-12] and also in some articles of other authors [15-17].

It is obvious that periods of simultaneous increase in air pollution above a considerable portion of a city are most dangerous from the standpoint of the influence on the health of the population, vegetation, materials, etc. Under such conditions, higher impurity concentrations are observed simultaneously over a large area. They are relatively stable in time. The level of the pollution of air with various impurities is raised. Thus, a combined action of a number of ingredients takes place, so that the danger is substantially increased. Obviously, an increase in the content of noxious impurities in air at most stationary points simultaneously may be explained chiefly by the influence of weather conditions. The study of periods of general heavy air pollution in a city is therefore of major importance in connection with meteorological measures designed to insure the purity of the atmosphere. The adoption of additional steps in the early prediction of such periods may lead to a lowering of the concentrations of noxious impurities in air in these cases and to a general decrease in the level of pollution of a city's air reservoir.

An objective method can be proposed for the identification of such periods. It consists in the fact that on the basis of data for a comparatively long period of time (a month, a season, half a year, etc.) a certain number of the highest concentrations is taken at each stationary point for each impurity being measured. As the criterion for referring a given measurement to the above-indicated cases, the condition  $q > 1.5 q_{av}$  is postulated, where  $q$  is any concentration, and  $q_{av}$  is the average concentration for the entire period considered. If in the course of one or several days  $m$  is the number of observations with  $q > 1.5 q_{av}$  and  $n$  is the total number of observations, the quantity  $P = \frac{m}{n}$  characterizes the degree of total air pollution for the city as a whole. Obviously,  $p$  varies from zero to 1. The values of  $p$  may be calculated both for individual impurities and for all the measured impurities taken together.

We have examined data on air pollution for January and July 1967 and also for the cold six months of the year (October 1967 - March 1968) for a number of cities. The values of  $p$  were calculated for all the cities for each

day. On the average, for different cities  $p \approx 0.2$ . Thus, at  $p > 0.2$ , the air pollution for the city as a whole may be considered high. On some days, values of  $p > 0.5$  were observed, when the accumulation of noxious impurities in the city air was most substantial.

Preliminary analysis has shown that cases with  $p > 0.5$  are usually observed under stagnant air conditions. In January 1967, a general heavy air pollution was noted in the course of several days almost simultaneously in Sverdlovsk and Magnitogorsk. On those days, both cities were located in a slow-moving crest of high pressure. Cases where  $p > 0.5$  are observed relatively seldom. Most frequently, periods of heavy air pollution in cities are characterized by the values  $p = 0.3-0.4$ .

It can be shown that the clustering of high values of impurity concentrations in the periods we have selected is not accidental. We are considering a purely binomial situation: each observed concentration is recorded only from the standpoint of whether it exceeds 1.5 average values for the entire period. For each period of time one can calculate the probability of random deviation from the average value of  $p$ . This is done most simply by assuming a normal probability distribution. In our case this is possible when the total number of observations  $n$  in the course of a period with heavy air pollution is no less than 60 [8]. Such a number of observations in the course of a day was carried out in Irkutsk, Novosibirsk, and Sverdlovsk. If, however, the period of heavy air pollution lasts two or three days in a row or longer,  $n > 60$  occurs in almost all of the cities considered. The calculations performed indicate that the probability of random formation of the observed periods is considerably below the 5% taken as the criterion [8]. In particular, at  $n = 100$  and  $p_{av} = 0.2$ , the probability that  $p = 0.3$  nonrandomly amounts to 99% and that  $p = 0.4$  to practically 100%.

Having calculated the parameter  $p$  for each day, we not only obtain the characteristics of air pollution for the city as a whole, but also substantially decrease the element of randomness present in single measurements as a result of the high variability of the concentration and a still inadequate quality of the observations.

A considerable portion of high impurity concentrations in city air noted in the month considered were observed in the course of a few days.

If in the course of the five days with the heaviest air pollution in January 1967 additional measures had been taken to reduce the noxious discharges, the amount of high impurity concentrations in the course of the month would be decreased by the following amounts: in Magnitogorsk, Leningrad, and Zaporozh'ye by a factor of 3-4; in Chita, Dnepropetrovsk, and Sverdlovsk by 2-2.5; and in the remaining cities considered by 1.5-2.

It is evident that in a large city, the simultaneous increase of air pollution by various impurities at different points is chiefly determined by the meteorological conditions. The present paper analyzes the weather conditions during periods with heavy air pollution of the city as a whole,

characterized by high values of the parameter  $p$ . The most general relationships are considered at first, then an attempt is made to refine them and define them concretely in order to apply them further to the prediction of air pollution.

Statistical relationships between the concentrations of noxious impurities in city air and meteorological factors are to a great extent well-known [1, 9, 10, 11, 12, 14, 18]. It is of interest to consider how these relationships are manifested in various cities differing in their character and total amount of discharges, location of the sources, etc. On the basis of data from nine cities for January and July 1967, the average values of meteorological parameters on days with heavy, and for comparison, with light air pollution were calculated. For this purpose, based on data for each city, five days per month were selected with the highest and five with the lowest values of  $p$ . The results for January are listed in Table 1.

Table 1

Average Values of Certain Meteorological Parameters During Periods of Heavy and Light Air Pollution in January 1967.

City	State of Air Pollution	Meteorological Parameters									
		$p$	$v_0$ , m/sec	$v_{500}$ , m/sec	$\Delta T^\circ$	$\Pi$ , %	$\Delta T_1^\circ$	$H_1$ , m	$\Pi$ , %	$H_2$ , m	$R$ , %
Leningrad	Heavy	0,61	0,9	8,0	0,8	56	6	510	44	460	0
	Light	0,00	1,3	11,1	1,5	50	5	700	50	740	0
Sverdlovsk	Heavy	0,66	0,1	3,8	-9,2	100	11	900	00	000	0
	Light	0,00	5,4	16,4	0,2	40	2	320	60	460	0
Novosibirsk	Heavy	0,36	2,2	9,2	-6,7	76	15	1510	24	530	10
	Light	0,15	5,3	15,3	0,5	42	7	1130	47	860	0
Chita	Heavy	0,40	1,0	7,0	-4,3	88	8	450	12	580	0
	Light	0,00	2,4	8,3	3,5	67	5	240	33	1360	0
Alma-Ata	Heavy	0,39	0,1	2,8	-2,3	88	5	320	12	80	13
	Light	0,08	0,4	2,6	3,5	43	5	300	57	520	27
Krivoy Rog	Heavy	0,35	4,6	8,2	-1,0	40	5	420	60	370	6
	Light	0,05	6,5	10,2	0,3	00	—	360	60	320	8
Irkutsk	Heavy	0,31	1,0	3,7	-1,8	94	10	540	6	50	39
	Light	0,08	5,3	8,1	-2,5	64	9	580	29	1180	00
Zaporozh'ye	Heavy	0,58	0,5	—	—	—	—	—	—	—	—
	Light	0,00	1,0	—	—	—	—	—	—	—	—
Dnepropetrovsk	Heavy	0,36	2,1	—	—	—	—	—	—	—	0
	Light	0,00	3,6	—	—	—	—	—	—	—	19

The following notation is used in Table 1:  $p$  - characteristics of air pollution for the city as a whole,  $v_0$  - wind velocity at the earth's surface,  $v_{500}$  - wind velocity at 500 m level,  $\Delta T$  - diurnal temperature difference near the ground and at the 500 m level,  $\Pi$  - frequency of ground inversions,  $\Delta T_1$  - intensity of ground inversions,  $H_1$  - height of upper boundary of ground inversions,  $\Pi_2$  - frequency of elevated inversions,  $H_2$  - height of the lower

boundary of elevated inversions,  $R$  - frequency of days with precipitation.

It is apparent from Table 1 that in winter, in all of the cities under consideration, during periods of heavy air pollution as compared with periods of light pollution, the wind velocity at the earth's surface and at the height of 500 m is substantially lower, the stratification of the boundary layer of the atmosphere is more stable, and the frequency of ground inversions and their average thickness and intensity are higher. This corresponds to the results obtained earlier, and also to the physical representations of the mechanism of general air pollution in cities. The conclusion drawn earlier concerning the effective purification of air due to the removal of impurities by precipitation was not confirmed. In many cases, heavy air pollution in the city as a whole was observed on days with precipitation. This is apparently due to the fact that in past studies [9, 11], we mainly considered the dust content of air, whereas in the present study consideration was given to the pollution of air with gaseous impurities, while only very few observations were made on the dust concentration.

The data of Table 1 indicate a stability of the known statistical relationships for the different cities. This confirms once again that the selected period of heavy air pollution in cities are nonrandom and that they are largely brought about by the weather conditions.

It thus appears possible to use the data of the study for a more precise and concrete definition of the relationship between the pollution of city air and the meteorological factors.

For the indicated cases, the weather conditions during periods of heavy air pollution of different intensities were examined. It was found useful to divide all the cases of heavy air pollution selected earlier (five days per month with the highest values of  $p$  for each city) into two types. The value  $p = 0.5$  was found to be convenient as the criterion. For the first type of periods  $p \geq 0.5$ . In this case, the air pollution over the entire city is particularly heavy. The second type of periods is characterized by the value  $0.5 > p \geq 0.3$ . As was noted earlier, these periods are statistically nonrandom. Also considered was a third type of periods which included all the cases of low  $p$  values selected earlier.

Because the statistical relationships were qualitatively identical for all the cities considered (see Table 1) and we are dealing with relative characteristics of air pollution, a combined consideration of data on the different cities is permissible as a first approximation. Thus, in studying the weather conditions of the total heavy pollution of city air of various intensities, the average values of the meteorological elements during the given periods were calculated together for all the cities considered. The results are given in Table 2. It is apparent from the Table that in the case where  $p \geq 0.5$  as compared to  $0.5 > p \geq 0.3$ , a further decrease of the wind and increase of the stability of the atmosphere are observed. In addition, it is characteristic that the parameters of the boundary layer in the two indicated types of heavy pollution differ on the average, only slightly, whereas the wind velocity at the earth's surface decreases sharply, practically approaching zero, when the air pollution is markedly heavy ( $p \geq 0.5$ ).



Table 2

Average Characteristics of Certain Meteorological Parameters for  
Different States of Air Pollution in Cities

State of Air Pollution	January 1967				July 1967			
	$v_0$ , m/sec	$v_{500}$ , m/sec	Cloudi- ness, points	$\Delta T^\circ$	$v_0$ , m/sec	$v_{500}$ , m/sec	Cloudi- ness, points	$\Delta T^\circ$
Markedly Heavy ( $p \geq 0.5$ )	0.5	5.4	3.5	-3.8	—	—	—	—
Heavy ( $0.5 > p \geq 0.3$ )	2.3	6.2	6.8	-3.2	2.7	5.8	7.5	4.4
Light	3.9	11.8	7.1	+0.9	3.1	6.5	7.1	8.8

No cases of heavy air pollution above the city as a whole ( $p \geq 0.5$ ) were noticeable in the summertime, despite the fact that on the average, in many cities the concentrations of noxious impurities in air in summer were higher than in winter [5, 10]. During the warm part of the year, the same general relationships prevail as during the cold part, although they are less pronounced; at the same time, it is most apparent that a heavy air pollution for the city as a whole takes place when during daytime hours the atmosphere is comparatively stable.

In the course of work on the refinement and concrete definition of the derived statistical relationships it was decided to narrow down the problem: subsequently, data on air pollution will be analyzed only for the cold part of the year.

Because of the necessity of having a large number of  $p$  values for individual cities, we are analyzing data on air pollution for cities where these values were calculated for half a year (Leningrad, Sverdlovsk, Chita and partly Novosibirsk, Omsk, Irkutsk). As noted above, the meteorological factors, which on the average determine the heavy air pollution, are known to some extent. In other words, while there are some considerable exceptions, we are generally aware of the meteorological conditions under which a dangerous accumulation of impurities in the air is observed with relative frequency, and under which such an accumulation occurs relatively seldom. The exceptions noted above may be due first of all, to the fact that the meteorological conditions variously affect the spreading of discharges of different types [2, 3, 4]. Secondly, the studies made earlier obviously have not permitted the consideration of all the factors determining such a complex process as the pollution of a city air reservoir. The present study offers an analysis of cases representing exceptions from the standpoint of known statistical relationships. An attempt is made to pass from general statistical relationships to specific ones between air pollution and meteorological conditions which would be manifested in individual cases.

It is well-known that a dangerous accumulation of impurities in the ground layer of air frequently takes place in the absence of wind. At the same time,

in the presence of a stable transport of air, no heavy air pollution above the city as a whole usually takes place. It is of interest to examine the manifestation of this relationship as a function of the nature of the temperature profiles of the lowest atmospheric layer. This was done on the basis of data for Leningrad (no such detailed analysis could be made for Sverdlovsk and Chita, since in these cities periods of calm were almost always associated with ground inversions). Days were considered when, during all the periods, the wind velocity did not exceed 1 m/sec. From these days, 17 cases with heavy air pollution ( $p > 0.25$ ) and 24 days with light air pollution ( $p < 0.20$ ) were selected. Simultaneously, from the group of cases where the average wind velocity was no less than 2 m/sec. 14 cases with heavy and 24 cases with light air pollution were selected. For each selection, the frequency of types of temperature profiles was calculated (Table 3).

It is evident from the table that heavy air pollution at a wind velocity of 0-1 m/sec is associated with ground inversions, whereas high impurity concentrations at a wind velocity of 2 m/sec and higher are characterized by elevated inversions. With light air pollution and a wind velocity of 0-1 m/sec., in the majority of cases elevated inversions are observed.

Similar results were obtained by E. Yu. Bezuglaya [1], who examined the relationship between air pollution and wind velocity in ground and elevated inversions. In ground inversions, the principal concentration maximum was found at a wind velocity of 0-1 m/sec, and in elevated ones, at 5-6 m/sec, as a result of heated discharges from large-capacity sources [3, 4].

Table 3

Frequency (%) of Types of Temperature Profiles for Heavy and Light Air Pollution in Leningrad During the Cold Part of the Year as a Function of the Wind Velocity.

State of Air Pollution	Gradations of Wind Velocity, m/sec	Temperature Profiles		
		Ground Inversions	Elevated Inversions $H < 1000$ m	Below 1000 m No Inversions Observed
Heavy ( $p > 0.5$ )	0-1	73	23	0
	$> 2$	7	64	29
Light ( $p < 0.25$ )	0-1	29	58	13
	$> 2$	13	47	40

This effect corresponds to the existing physical interpretation, i.e., elevated inversions are most unfavorable at a dangerous wind velocity [2, 3], which for sources located in cities ranges from 1 to 6-7 m/sec. The presence of low concentration in the region of a State Regional Electric Power Plant (SREPP) in the case where an elevated inversion is associated with a weak wind is indicated in the paper of B. B. Goroshko [7].

In a city where a substantial contribution to the air pollution is made by low discharges, a dangerous combination is that of a very slight wind and a ground inversion. However, even for such a combination, which we shall

arbitrarily refer to as air stagnation, a heavy pollution of the atmosphere is not always observed. We have given special attention to air pollution under stagnation conditions in several cities. Table 4 indicates the frequency of high impurity concentrations in the air under these conditions. By heavy air pollution for a city as a whole we shall mean cases where  $p$  is greater than the average, i. e.,  $p > 0.2$ .

Table 4  
Frequency of Days with Heavy Air Pollution Under Stagnation Conditions.

City	Number of Days		Frequency (%) of Days with $p > 0.2$
	With Stagnation	Of These, with $p > 0.2$	
Leningrad . . . . .	19	13	68
Sverdlovsk . . . . .	13	5	38
Novosibirsk . . . . .	10	4	40
Irkutsk . . . . .	23	19	83
Chita . . . . .	27	26	96
Total . . . . .	92	67	73

As is evident from Table 4, the frequency of heavy air pollution under stagnation conditions is different in different cities. In Chita and Irkutsk, the combination of wind with a velocity less than 1 m/sec and a ground inversion may be regarded as a prognostic sign for predicting a heavy accumulation of impurities near the ground. Apparently, the data listed in Table 4 reflect the fact that the nature of the air pollution during stagnation periods depends on the characteristics of the discharges in the city. It is well known that the danger of stagnation conditions is greater when the contribution of low discharges is large. Indeed, in accordance with the data obtained, the contribution of high discharges to the total air pollution in Sverdlovsk and Novosibirsk is substantially greater than in Chita and Irkutsk.

Table 5  
Frequency (%) of Winds of Different Directions for Heavy and Light  
Air Pollution in Cities as a Whole.

City	State of Air Pollution	Wind Direction								Number of Cases
		NE	E	SE	S	SW	W	NW	N	
Leningrad	Heavy	14	11	36	8	25	3	0	3	36
	Light	18	0	0	14	14	27	27	0	22
Sverdlovsk	Heavy	0	0	26	30	11	24	9	0	46
	Light	7	7	0	14	14	41	3	14	29

In the study of weather conditions during periods of heavy air pollution in a city as a whole, an important factor determining the distribution of noxious impurities is the wind direction. The character of the influence of the wind direction may be related to at least two factors: the predominance of the main sources of discharges in a region and the dependence of the

direction of movement of air on the general meteorological situation (type of air mass, advection of heat and cold, etc.).

Table 5 lists data on the frequency of certain wind directions in the case of heavy and light air pollution in Leningrad and Sverdlovsk in December, January and February. The most distinct relationship between the magnitude and direction of the wind was found in an analysis of the three middle winter months.

It is evident from the table that in both cities, a high air pollution is associated with winds of southern directions, the most dangerous being the southeastern wind. As an indication of a high impurity content in air, one can take the following: in Leningrad, eastern, southeastern, southern and southwestern winds, in Sverdlovsk, southeastern and southern winds. We selected cases when winds of these directions were observed for 24 hours. In Leningrad, out of 29 cases, a heavy air pollution ( $p > 0.2$ ) was observed 20 times (69%), and in Sverdlovsk, out of 18 cases, 13 times (72%). Exceptions were found to be associated with higher wind velocities.

In Leningrad, for the same wind directions and a velocity not more than 2 m/sec, a heavy air pollution was observed 16 times (89%), out of 18 cases, and in Sverdlovsk, in 5 cases out of 5.

The general air pollution in a city as a whole is also determined by a lag factor. Whereas at an individual point the concentrations of impurities may increase very rapidly, the value of  $p$  characterizing the accumulation of noxious impurities in the air of the city as a whole increases gradually. This is manifested particularly under conditions of air stagnation (Fig. 1). It is evident from Fig. 1 that under stagnation conditions a close relationship exists between the previous ( $p_{n-1}$ ) and current ( $p$ ) air pollution in the city. This conclusion is of great prognostic importance.

When the lag is taken into account, the reliability of the statistical relationships increases substantially, particularly under stagnation conditions. From data for Leningrad, Sverdlovsk and Chita, 35 cases were selected in which the wind velocity at the earth's surface was less than 1 m/sec and at the height of 500 m less than 10 m/sec, a ground inversion took place, and  $p$  for the previous day ( $p_{n-1}$ ) was greater than 0.15. Of these, heavy air pollution ( $p > 0.2$ ) was observed 34 times (97%).

Thus, data on air stagnation, taking into account the existing level and lag, make it possible to predict with substantial confidence a heavy air pollution during the cold time of the year.

As indicated earlier, in the absence of stagnation, a marked pollution of the atmosphere is associated with certain directions of wind of low velocities and with an increase in air temperature. It is characteristic that under these conditions, the role of the lag is less substantial than in stagnation conditions: in a number of cases, heavy air pollution arose after low impurity concentrations in the city during the previous day ( $p_{n-1} < 0.15$ ).

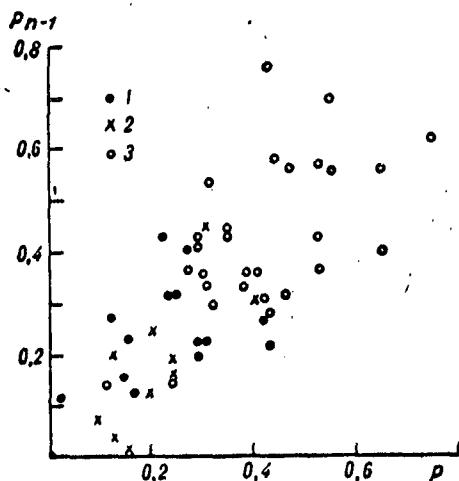


Fig. 1. Correlation between air pollution under stagnation conditions ( $p$ ) and the value of  $p$  during the previous day ( $p_{n-1}$ ).

1 - Leningrad, 2 - Sverdlovsk, 3 - Chita

Thus, the precision of statistical relationships and the identification of some new effects of the influence of meteorological conditions on the content of noxious impurities in cities, as well as the introduction of a new characterization of air pollution for a city as a whole ( $p$ ), in which the element of randomness has been markedly reduced, have permitted the formulation of certain prognostic indicators of great accuracy. These initial results of investigations in this direction show that they should be continued in order to work out a method for predicting air pollution in cities.

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## ATMOSPHERIC DIFFUSION OF IMPURITIES DURING A CALM

M. Ye. Berlyand and O. I. Kurenbin

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It is well-known that when the wind velocity dies down to a calm, high concentrations of impurities originating from sources can build up in the ground layer of air. And yet, the formulas for the calculation of impurity dispersal in the atmosphere used at the present time pertain to conditions in which the wind velocity is substantially different from zero.

As a basis for the derivation of such formulas, the solution of the equation of turbulent diffusion

$$u \frac{\partial q}{\partial x} + w \frac{\partial q}{\partial z} = \frac{\partial}{\partial z} k_z \frac{\partial q}{\partial z} + k_y \frac{\partial^2 q}{\partial y^2} \quad (1)$$

is widely employed with suitable boundary conditions specifying the presence of a source, the nature of the interaction of the impurity with the underlying surface, and a decrease of the concentration at a sufficiently large distance from the source.

Here  $q$  is the impurity concentration,  $u$  the wind velocity,  $w$  the vertical displacement velocity of the impurity, and  $k_z$  and  $k_y$  the vertical and horizontal components of the exchange coefficient. Axis  $x$  is directed along the mean wind, axis  $y$  is perpendicular to it in the horizontal plane, and axis  $z$  is directed along the vertical.

The form of equation (1) and the parabolic character of its solution are substantially determined by the fact that it contains the advective term  $u \frac{\partial q}{\partial x}$ . In particular, it follows from solution (1) that in the presence of a high source at some distance from it along the mean direction of the wind, a maximum of the ground concentration is reached. On the basis of a numerical solution of (1) it was found in [3, 4, etc.] that for a sufficiently general form of the coefficients in (1), characteristic of the real atmosphere,

$$q_m = \frac{a_1}{u_1^\alpha H^\beta} \quad (2)$$

Here  $H$  is the height of the source,  $u_1$  is the wind velocity in the ground layer of air at a fixed height  $z_1$  (usually  $z_1 = 1$  m), and  $\alpha$  and  $\beta$  are some positive constants whose values are given in the indicated papers. The coefficient  $a_1$  depends on the capacity of the source and the turbulent characteristics of the atmosphere.



Formulas of type (2) for certain values of  $\alpha$  and  $\beta$  are also given in other papers, where a solution of the equation of atmospheric diffusion was obtained for certain special expressions for the coefficients of this equation. From the formulas for the calculation of the impurity concentration, which are based mainly on statistical considerations, including the familiar Sutton formulas [15], there also follows a particular form of formula (2) for  $q_m$ :

$$q_m = \frac{a_2}{uH^2}. \quad (3)$$

It should be noted, however, that in this case the problem of the level above the underlying surface to which the velocity  $u$  should be referred remains unsolved.

It follows from expressions (2) and (3) that  $q_m$  is inversely proportional to the wind velocity and formally should increase indefinitely as the latter falls off to zero. This makes it impossible to apply the working formulas to calm conditions.

An original attempt to extend the Sutton formula to the case of absence of wind was made by Fortak [17], who used the relationship between the components of the exchange coefficient and dispersion and also the general form of the solution of the equation of impurity diffusion. In deriving the steady-state concentration as a result of integration of the solution for an instantaneous source, Fortak postulated that the exchange coefficient and dispersion are functions of time alone and independent of the coordinates.

On the basis of general considerations, Fortak assumed that the maximum of the ground concentration from a high source is reached directly under the source, and he obtained an approximate formula for estimating this maximum. However, the formula obtained was not investigated, and no numerical calculations were made with it in Fortak's paper. It is thus difficult to gauge the actual potential of the applications of his results.

Of some interest in this connection is the paper of Nester [18], which discussed Fortak's results and gave examples of comparison of calculations of maximum concentration using Fortak's and Sutton's formulas for two special cases. In a physical sense, however, these examples do not give clear cut results.

A somewhat similar study was made by D. Yordanov [12], except for the fact that he assigned a value to the vertical component of the exchange coefficient.

The unlimited increase of the concentrations as the velocity decreases to zero causes difficulties in the solution of a number of problems. In this connection, the question of setting up norms for discharges into the atmosphere requires a special treatment. Thus, when planning and running enterprises and other facilities whose operation is associated with the expulsion of noxious substances into the atmosphere, it is necessary to determine the

amount of the discharges, their height, and other parameters so that the impurity concentrations do not exceed the maximum permissible values under any meteorological conditions. For a fixed source height, this problem may appear insoluble if  $q_m \rightarrow \infty$  when  $u_1 \rightarrow 0$ , and it follows further that the most unsafe conditions should always exist during a calm. However, in considering industrial discharges, the situation changes because the stack gases usually have an initial vertical escape velocity and are frequently overheated relative to the surrounding atmosphere. Consequently, one must introduce some initial ascent  $\Delta H$ , and thus the effective level of the discharge turns out to be higher than the actual height of the source. It is significant that  $\Delta H$  depends on the wind velocity, and since as  $u$  decreases it increases, and so does  $q_m$ , according to (2), there exists a so-called "dangerous wind velocity"  $u_M$ , at which  $q_m$  reaches its highest value. According to the calculations made in [4, 7], for heat power plants and high-capacity industrial enterprises  $u_M$  3-5 m/sec. At low velocities, low impurity concentrations are observed in these cases in the ground layer of air. This is also indicated by data of experimental observations. As an example, Fig. 1 shows values of  $q_m$  as a function of the wind velocity based on data of expeditionary studies in the region of the Shchekino State Regional Electric Power Plant (SREPP). In the case of cool discharges (when the temperature difference between them and the surrounding medium is close to zero) it turns out that the dangerous velocity decreases, but it is also different from zero.

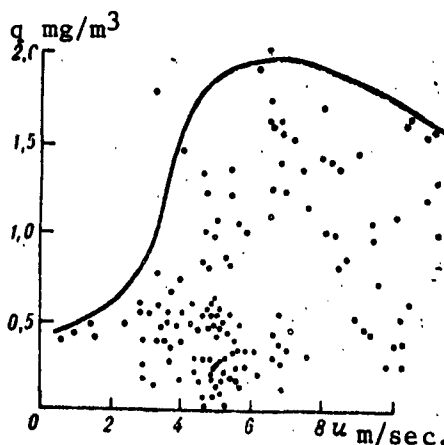


Fig. 1.

The formulas for calculating  $\Delta H$  as a function of  $u$ , given in [4, 5], pertain to the most frequently observed meteorological conditions, in particular, when the temperature drops with the height. However, they are inapplicable under certain anomalous conditions. Such "anomalous" cases are usually related to the presence of a temperature inversion above the source. Refs. [3, 6] give a numerical solution of the problem for cases where the coefficients of turbulent exchange decline sharply above the source as a result of a rise of temperature with the height. The source height was assumed constant. It was found that the ground concentration in the presence of such an "intercepting layer" increased, and did so to a higher degree the lower this layer was above the

source. Under conditions where the temperature inversion begins immediately above the source, the ground concentration maximum increases by a factor of 1.5-1.7, and sometimes more than 2.

In [6] it was noted that in the presence of an inversion above the source, it is necessary to consider its influence on the initial ascent of the impurity. In order to obtain a limiting estimate of the latter, a formula was obtained in [6] for calculating  $\Delta H$  as a function of the gradient of temperature inversion during a calm. One should bear in mind that in the presence of the wind, the value of  $\Delta H$  will of course be lower. It was found that for these conditions there exists some "ceiling"  $z_M$  above which the impurity from the source cannot rise. Hence,  $\Delta H$  does not increase indefinitely, as when the temperature decreases with the height, but takes on a certain final value. For large heat sources, the height  $z_M$  is substantial, and the impurities usually "pierce" the temperature inversion, rising above it. In the case of cool impurities from relatively weak sources,  $z_M$  turns out to be small, on the order of a few tens of meters, and this markedly alters the problem of dangerous wind velocity. Since  $\Delta H$  turns out to be limited, it follows again from relation (2), if the latter applies, that  $q_m$  grows indefinitely as the wind velocity drops to zero.

Hence, it may be concluded that the presence of dangerous conditions is connected with the calm and the temperature inversion above the source. However, as already noted, the use of (2) in this case does not permit one to set up norms for the discharges. Furthermore, it is clear from physical considerations that the concentration cannot increase indefinitely. Equation (1), from which formula (2) was derived, is actually approximate. It does not take into account the term describing the turbulent diffusion of the impurity along axis  $x$ . This approximation is valid in the presence of a wind velocity, when the advective transport of the impurity along the direction of the wind substantially exceeds its diffusive displacement. However, when  $u$  decreases to zero, this approximation is obviously invalid.

As was noted earlier, dangerous conditions may include the presence of a temperature inversion above the source of impurity. In addition, the elevation and lowering of the temperature with the height in the subjacent layer of air must be distinguished. In the first case, as the wind velocity drops to zero, turbulent exchange also disappears for all practical purposes. Essentially, therefore, there are no factors causing the impurity to spread from the sources. In practice, the problem amounts exclusively to the molecular diffusion of the impurity, whose laws are known. Conversely, under conditions where the temperature drops with the height in the lower layer of air down to the level of the source, convective motions may develop in this layer. As we know, in this case turbulent exchange is very substantial even in the absence of wind [14, etc.].

Consequently, it is of interest to consider the conditions of diffusion of an impurity in the absence of the wind, but during the development of turbulent exchange. In so doing, it is important to estimate the limiting value of the concentration during a calm, and this will permit one to set up norms for the discharges into the atmosphere. The present paper is devoted precisely to the finding and investigation of the solution of the equation of

atmospheric diffusion, taking into consideration the turbulent mixing in all directions in the case of absence of the wind.

We can confine ourselves to the absence of the vertical displacement velocity as well. The equation being sought then assumes the form

$$\frac{\partial}{\partial z} k_z \frac{\partial q}{\partial z} + \frac{\partial}{\partial y} k_y \frac{\partial q}{\partial y} + \frac{\partial}{\partial x} k_x \frac{\partial q}{\partial x} + M \delta(x) \delta(y) \delta(z - H) = 0. \quad (4)$$

Here the last term expresses the presence of a source,  $M$  being its capacity and  $\delta(\xi)$  being the delta function. It is useful to obtain the solution of this equation for values of the components of the exchange coefficient at which it would be the limiting expression for the solution of equation (1) obtained in [4, 5, 7]. The values of these components should be selected so that they also are the limiting expressions for their values in the presence of a wind velocity. For the vertical component of the exchange coefficient we shall assume, as in the above-mentioned references, that it depends on the height, i. e.,  $k_z = k_1 f_1(z)$ , where  $f_1(z)$  is some function of the height. With respect to the horizontal component  $k_y$  in [4, 5, etc.], it is assumed, according to [2], to increase with the height in proportion to the wind velocity, i. e.,  $k_y = k_0 u$ . In addition, the averaging of  $q$  was carried out by allowing for the fluctuation of the wind direction with time, whose distribution  $\omega(\varphi)$  is described in the general case by the standard law.

$$\omega(\varphi) = \frac{1}{\varphi_0 \sqrt{2\pi}} e^{-\frac{\varphi^2}{\varphi_0^2}},$$

where  $\varphi_0$  is the average dispersion during the time period considered. As a result, according to [4, 5], the function  $k_0(x) = k_0 + x\varphi_0^2$  should be introduced in place of  $k_0$ , or, for practical purposes,  $k_0(x) \approx x\varphi_0^2$ , is sufficient. As far as the other horizontal component  $k_x$  is concerned, it may be assumed equal to  $k_y$ , this being valid because of symmetry in the absence of the wind. Further, it is necessary to consider the limiting dependences of the components of the exchange coefficient on the wind velocity at small  $u$  values.

In the general case one can write that for the dependence on the height  $z$ ,  $u = u_1 f_2(z)$ , where  $u_1$  is the wind velocity at a fixed level, for example, at a height of  $l_m$ , and  $f_2(z)$  is some function of the height  $z$ . As already noted, during convection in the case of absence of the mean wind,  $k_y$  and  $k_x$ , like  $k_z$ , assume values different from zero. Therefore, from considerations of dimensionality one can postulate that as the wind velocity decreases, the quantity  $\beta^2 = \varphi_0^2 u$  retains a certain value. Hence, at low wind velocities,  $\varphi_0$  changes in accordance with the equality  $\varphi_0 = \frac{\beta}{\sqrt{u_1}}$ . Then  $k_y = \beta^2 x f_2(z)$ .

Similarly one can write  $k_1 = \frac{k}{u_1}$  and assume at low wind velocities that

$$\frac{k_1}{u_1} = \frac{\alpha}{u_1}, \text{ where } \alpha \text{ is a constant.}$$

Let us now turn to the integration of equation (4), and first write it in the cylindrical coordinate system

$$\frac{\partial}{\partial z} k_z \frac{\partial q}{\partial z} + \frac{1}{r} \frac{\partial}{\partial r} \left( r k_r \frac{\partial q}{\partial r} \right) + \frac{M}{2\pi r} \delta(r) \delta(z-H) = 0, \quad (5)$$

where  $k_z = k_1 f_1(z)$  and  $k_r = \beta^2 r f_2(z)$ .

As the boundary conditions we shall take as usual

$$\begin{aligned} \text{for } z=0 \quad k_z \frac{\partial q}{\partial z} &= 0, \\ \text{for } r^2 + z^2 \rightarrow \infty \quad q &\rightarrow 0, \end{aligned} \quad (6)$$

and by virtue of the symmetry of the horizontal concentration field we shall also set for  $r=0$ ,  $k_r \frac{\partial q}{\partial r} = 0$ .

The solution of this problem can be obtained numerically in the same expressions for the functions  $f_1(z)$  and  $f_2(z)$ , which are adopted in refs. [4, 5, 7 etc.]. However, at this stage, when it is necessary to obtain the first estimates and to get some idea of the laws of distribution of the concentrations during a calm, it is useful to obtain an analytical solution of the problem by confining ourselves to some particular form of the functions  $f_1(z)$  and  $f_2(z)$ .

Making use of the fact that the homogeneous part of equation (5) allows the separation of variables, we shall represent its solution in the form

$$q = q_1(r) q_2(z),$$

where  $q_1$  satisfies the equation

$$\frac{d}{dr} r^2 \frac{dq_1}{dr} + \omega^2 r q_1(r) = 0, \quad (7)$$

and hence (cf. [13]),

$$q_1 = \frac{1}{\sqrt{r}} J_1(2\omega\sqrt{r}). \quad (8)$$

Here  $\omega^2$  is some constant (spectral parameter)

To integrate (5), it is desirable to use, as is sometimes done (see for example [9, 16]), an integral transform of the form

$$\bar{q}(\omega, z) = \int_0^\infty q(r, z) q_1(r, \omega) r dr \quad (9)$$

or, in accordance with (8),

$$\bar{q}(\omega, z) = \int_0^\infty q(r, z) \sqrt{r} J_1(2\omega\sqrt{r}) dr, \quad (10)$$

which actually corresponds to the well-known Hankel transform.

We shall subject all the functions contained in equation (5) and boundary condition (6) to this transformation.

On the basis of equation (7) and conditions (6)

$$\int_0^\infty \frac{J_1(2\omega\sqrt{r})}{\sqrt{r}} \frac{\partial}{\partial r} r^2 \frac{\partial q}{\partial r} dr = -\omega^2 \bar{q}.$$

Since at low  $r$  values asymptotically  $J_1(2\omega\sqrt{r}) \approx \omega\sqrt{r}$ , then

$$\int_0^\infty \frac{1}{\sqrt{r}} J_1(2\omega\sqrt{r}) \delta(r) dr = \omega.$$

Thus, (5) for the transform function will assume the form

$$\frac{d}{dz} k_1 f_1(z) \frac{d\bar{q}}{dz} - \beta^2(z) \omega^2 \bar{q} + \frac{M}{2\pi} \omega \delta(z-H) = 0 \quad (11)$$

with the boundary conditions at  $z = 0$ ,  $k_z \frac{d\bar{q}}{dz} = 0$ , при  $z \rightarrow \infty \bar{q} \rightarrow 0$ .

Equation (11) will be integrated by using the Green function  $G(z, \zeta)$  satisfying the homogeneous part of (11), the boundary conditions, and also the continuity conditions of  $G(z, \zeta)$ , and of the jump  $\frac{dG}{dz}$  at  $z = \zeta$ , equal to  $\frac{1}{k_1 f_1(z)}$ .

According to the properties of the Green function, the solution of equation (11) is related to it by the expression

$$\bar{q} = \frac{M\omega}{2\pi} \int_0^\infty G(z, \zeta) \delta(\zeta - H) d\zeta,$$

or

$$\bar{q} = \begin{cases} \frac{M\omega}{2\pi} G(z, H) & \text{при } z < H \\ \frac{M\omega}{2\pi} G(H, z) & \text{при } z > H \end{cases} \quad (12)$$

We introduce for consideration two fundamental solutions of the homogeneous part of equation (11),  $\tau_1$  and  $\tau_2$ . Then according to [1] we obtain

$$G(z, \zeta) = \frac{\tau_1(z)}{\left(k_1 f_1(z) \frac{d\tau_1}{dz}\right)_{z=0} \cdot \omega} \left[ \left(k_1 f_1 \frac{d\tau_2}{dz}\right)_{z=0} \tau_1(\zeta) - \right. \\ \left. - \left(k_1 f_1 \frac{d\tau_1}{dz}\right)_{z=0} \tau_2(\zeta) \right], \quad (13)$$

where  $\omega = k_1 f_1 \left(\tau_1 \frac{d\tau_2}{dz} - \tau_2 \frac{d\tau_1}{dz}\right)_{z=0}$  is the Wronskian of the functions  $\tau_1$  and  $\tau_2$ .

Since the change of the wind velocity and of the exchange coefficient in the ground layer of air with the height is approximately described by power functions, we can set

$$f_1(z) = z^m, \quad f_2(z) = z^n,$$

where  $m$  and  $n$  are positive constants, and  $m \approx 1$ ,  $n \approx 0.1-0.3$ . Then (see [13])

$$\tau_1 = (\eta \omega)^\mu K_\mu(\eta \omega) \quad \text{и} \quad \tau_2 = (\eta \omega)^\mu I_\mu(\eta \omega), \quad (14)$$

where

$$\mu = \frac{1-m}{2+n-m}, \quad \eta = \frac{2\beta}{(2+n-m)\sqrt{k_1}} z^{-\frac{2+n-m}{2}}, \quad (15)$$

$K_\mu(\eta \omega)$  are MacDonald functions and  $I_\mu(\eta \omega)$  is the Bessel function of an imaginary argument.

We thus obtain an expression for the Green function in accordance with (13) and (12)

$$\bar{q}(\omega, z) = \frac{M \omega}{\pi k_1(2+n-m)} (zH)^{\frac{1-m}{2}} K_{-\mu}(\eta_H \omega) I_{-\mu}(\eta \omega) \quad (16)$$

for  $z \leq H$  and

$$\bar{q}(\omega, z) = \frac{M \omega}{\pi k_1(2+n-m)} (zH)^{\frac{1-m}{2}} K_{-\mu}(\eta \omega) I_{-\mu}(\eta_H \omega) \quad (17)$$

for  $z \geq H$ , where  $\eta_H = |\eta|_{z=H}$ .



It is now necessary to turn from  $\bar{q}(\omega, z)$  to the function  $q(r, z)$ , i. e., to perform a transformation which is the opposite of (10). This can be done by using the general theory of expansion in eigenfunctions (see for example [16]). However, in our specific case, after some simple operations involving the substitution of variables which reduce (10) to a standard Handel transform [9], we obtain

$$q(r, z) = \frac{2}{\sqrt{r}} \int_0^\infty L_1(2\omega\sqrt{r}) \omega \bar{q}(\omega, z) d\omega. \quad (18)$$

Substitution of (16) into (18) gives

$$q(r, z) = \frac{2M}{\pi k_1 \sqrt{r}} \frac{1}{2+n-m} (zH)^{\frac{1-m}{2}} \int_0^\infty \omega^2 J_1(2\omega\sqrt{r}) \times \quad (19)$$

$$\times K_{-\mu}(\eta_H \omega) I_{-\mu}(\eta \omega) d\omega$$

for  $z \leq H$  and

$$q(r, z) = \frac{2M}{\pi k_1 \sqrt{r}} \frac{1}{2+n-m} (zH)^{\frac{1-m}{2}} \int_0^\infty \omega^2 J_1(2\omega\sqrt{r}) \times$$

$$\times K_{-\mu}(\eta \omega) I_{-\mu}(\eta_H \omega) d\omega$$

for  $z > H$ .

To determine the integrals in (19), we use the relation (see [13], formula 6.578<sub>11</sub>)

$$\int_0^\infty x^{\nu+1} K_\mu(ax) I_\mu(bx) J_\nu(cx) dx = \frac{(ab)^{-\nu-1} c^\nu e^{-(\nu+\frac{1}{2})\pi} Q_{\mu-\frac{1}{2}}^{\nu+\frac{1}{2}}(u)}{\sqrt{2\pi}(\nu^2-1)^{\frac{1}{2}\nu+\frac{1}{4}}},$$

where  $2abu = a^2 + b^2 + c^2$ ,  $[\operatorname{Re} a > |\operatorname{Re} b|]$ ,  $c > 0$ ,  $\operatorname{Re} \nu - 1$ ,  $\operatorname{Re}(\nu + \frac{1}{2}) > -1$ ,  $Q_\mu^\nu(u)$  — is a spherical function of the second kind.

Then

$$q(r, z) = \frac{M}{k_1} \left(\frac{2}{\pi}\right)^{1/2} (zH)^{\frac{1-m}{2}} \frac{IQ_{-\mu-\frac{1}{2}}^{1/2}(s)}{(\eta \eta_H)^2 (s-1)^{1/2}}, \quad (20)$$

where

$$s = \frac{\eta_H^2 + \eta^2 + 4r}{2\eta \eta_H},$$

and  $\eta$  and  $\eta_H$  are determined in accordance with (15) and (17).

By virtue of the relation (cf. [9], formula 8.723<sub>2</sub>)

$$Q_\nu^\mu(\text{ch } \alpha) = e^{i\pi\mu} 2^\mu \sqrt{\pi} \frac{\Gamma(\nu + \mu + 1) e^{-(\nu + \mu + 1)}}{\Gamma\left(\nu + \frac{3}{2}\right) (1 - e^{-2\alpha})^{\mu + \frac{1}{2}}} \text{ch } \alpha \times \\ \times F\left(\mu + \frac{1}{2}, -\mu + \frac{1}{2}, \nu + \frac{3}{2}; \frac{1}{1 - e^{-2\alpha}}\right) \\ \left[\mu + \nu + 1 \neq 0, -1, -2, \dots; \alpha > \frac{1}{2} \ln 2\right],$$

where  $F(\alpha, \beta, \gamma; z)$  is a hypergeometric function.

We transform (20) into the form

$$q(r, z) = \frac{8M}{\pi} \frac{1}{k_1} (zH)^{\frac{1-m}{2}} \frac{(1-\mu) e^{-(2-\mu)\alpha}}{(1 - e^{-2\alpha})^2} F\left(2; -1; \mu + 1; \frac{1}{1 - e^{-2\alpha}}\right),$$

where  $\cosh \alpha = s$ .

In this case, the second index of the hypergeometric function is equal to -1, and this function can therefore be represented (cf. [9], formula 9.100) as follows:

$$F\left(2; -1; -\mu + 1; \frac{1}{1 - e^{-2\alpha}}\right) = 1 + \frac{1}{1-\mu} \frac{1}{\text{sh } \alpha} \frac{1}{\text{ch } \alpha + \text{sh } \alpha}.$$

As a result, after some algebraic operations, we obtain

$$q(r, z) = \frac{M}{\pi} \frac{2^{3(1-\mu)}}{(2+n-m)^{1-2\mu}} \frac{1}{k_1^{1-\mu} \beta^{2\mu}} \times \\ \times \left[ \frac{D^2}{T^2(T+R)^{1-\mu}} + \frac{(1-\mu)(T+R)\mu}{T^2} \right], \quad (21)$$

where

$$D = 2\eta\eta_H; R = \eta^2 + \eta_H^2 + 4r; T = \sqrt{R^2 - D^2}.$$

For a linear increase of the exchange coefficient with the height, i. e.,  $m = 1$ , which corresponds to  $\mu = 0$ , we obtain

$$q = \frac{M}{2\pi k_1(1+n)} \frac{\left(\frac{1}{1+n}\right)^2 \frac{\beta^2}{k_1} (H^{1+n} + z^{1+n}) + r}{\left[\left(\frac{1}{1+n}\right)^2 \frac{\beta^2}{k_1} \left(H^{\frac{1+n}{2}} - z^{\frac{1+n}{2}}\right)^2 + r\right]^{\eta_1}} \times$$

$$\times \frac{1}{\left[\left(\frac{1}{1+n}\right)^2 \frac{\beta^2}{k_1} \left(H^{\frac{1+n}{2}} + z^{\frac{1+n}{2}}\right)^2 + r\right]^{\eta_0}}. \quad (22)$$

For the ground concentration  $q_0 = q_{z=0}$ , expression (21) becomes simplified and assumes the form

$$q_0 = \frac{M}{2\pi} \frac{1-\mu}{k_1^{1-\mu} \beta^{2\mu}} \frac{(2+n-m)^{2\mu-1}}{\left[\left(\frac{1}{2+n-m}\right)^2 \frac{\beta^2}{k_1} H^{2+n-m} + r\right]^{2-\mu}}. \quad (23)$$

From the formulas obtained it is apparent that the maximum ground concentration is reached at  $r = 0$ , i. e., under the source, and is equal to

$$q_m = \frac{M}{2\pi} (1-\mu)(2+n-m)^2 \frac{k_1}{\beta^4} \frac{1}{H^{2+n-m}}, \quad (24)$$

and when  $m = 1$

$$q_m = \frac{M}{2\pi} (1+n)^2 \frac{k_1}{\beta^4} \frac{1}{H^{2(1+n)}}. \quad (25)$$

We can introduce a certain distance  $r = r_{1/2}$  at which the concentration is reduced by one-half relative to the maximum value. It can be readily seen that

$$r_{1/2} = \frac{0.41 \beta^2}{k_1(2+n-m)^2} H^{2+n-m}. \quad (26)$$

From (23) it follows that  $\frac{q_0}{q_m}$  is a function of  $\frac{r}{r_{1/2}}$  only. A graph of this function is shown in Fig. 2.

It is interesting to compare the results obtained with the solution of an analogous problem in the presence of wind velocity with the same models for  $k_y$  and  $k_z$ .

According to (4), for the maximum ground concentration we have in this case (for  $m = 1$ ),

$$c_m = \frac{0.216 M k_1 |u_{+0}| (1+n)^2}{\varphi_0 u_1^2 H^{2(1+n)}}. \quad (27)$$

Then from (25) and (27) for  $\gamma = \frac{c_m}{q_m}$  it follows that

$$\gamma = \frac{0,432 \pi \beta^4}{\varphi_0 u_1^2} \quad (28)$$

In addition, it was assumed that (27) is determined for comparatively low velocities, so that the coefficient  $k_1$  in (25) and (27) is approximately the same.

The quantities  $\beta^2$  and  $\varphi_0$  are related to each other by the expression  $\beta^2 = \varphi_0^2 u_1$ , if comparatively small  $u_1$  values are considered. In the case of a gentle wind, as already noted, the quantities  $\beta$  and  $k_1$  and hence their ratio  $\frac{k_1}{\beta} = a$  remain approximately constant as  $u_1$  changes. Treatment of the observational data given in [8, 10] shows that  $a \approx 0,5 \frac{\text{m}^{1/2}}{\text{cek.}^{1/2}}$ . Therefore, it may

be approximately assumed that  $\beta \approx 2k_1$  (in the same system of units). Then, according to (28), the ratio  $\gamma = \frac{c_m}{q_m}$ , will take the form  $\gamma \approx 11 \left( \frac{k_1}{\sqrt{u_1}} \right)^3$ .

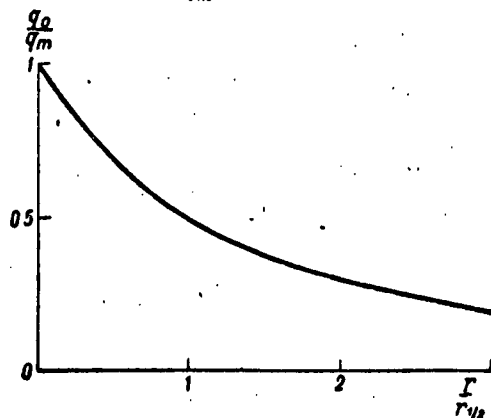


Fig. 2.

For a wind velocity  $u_1 = 1$  m/sec and the value  $k_1 \approx 0,15$  m/cek., characteristic of convection conditions,  $\gamma \approx 0,04$ , i. e., the concentrations at this velocity are approximately 25 times lower than during a calm. However, as the wind velocity decreases,  $\gamma$  increases rapidly. One can establish a certain wind velocity  $u_1 = u_{10}$  at which  $\gamma = 1$ , i. e., a limiting value of the maximum concentration is reached. Obviously, the calculation of the concentration  $c_m$  from formula (27) for wind velocities  $u_1 < u_{10}$  does not make sense. For the indicated values of  $k_1$  it was found that  $u_{10} \approx 0,1$  m/sec and  $r_{1/2} = 60$  m at a source height  $H = 120$  m.

On the basis of the formulas obtained for  $q_m$  during a calm and  $c_m$  for  $u \neq 0$ , a convenient form of the interpolation formula can be proposed:

$$q_u = \frac{0,216 M k_1 (1+n)^3}{(1,36 \beta^4 + \varphi_0 u_1^2) H^{2(1+n)}} \quad (29)$$

Fig. 3 shows the corresponding graph.

The use of this formula makes it possible to calculate the maximum ground concentration both in the presence and absence of the wind.

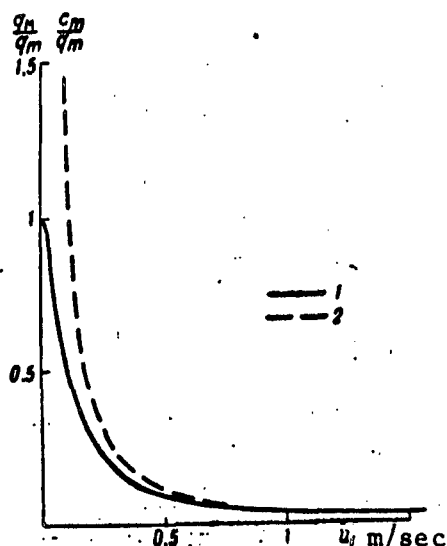


Fig. 3. Dependence of  $\frac{cm}{qm}$  (1) and  $\frac{cm}{qm}$  (2) on  $u_1$ .

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PATTERNS OF VARIATION OF THE TEMPERATURE GRADIENT  
IN THE GROUND LAYER OF AIR ON THE TERRITORY OF THE USSR

N. A. Kas'yan, T. A. Ogneva, and K. M. Terekhova

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p. 137-151, (1968).

The temperature gradient is one of the most important characteristics in the determination of turbulent exchange in the ground layer of air. The magnitude of this gradient can be characterized by the difference in air temperatures at two levels (0.5 and 2 m) near the underlying surface. The temperature gradient of the ground layer differs from the gradient in the free atmosphere in a considerable change of its magnitude and direction (change of sign). As a result, the temperature stratification and hence the turbulence coefficient also change. For this reason, the determination of the pattern of variation of the temperature gradient is of major interest in the problem of turbulent exchange and in practical problems where its indicators are used.

On the territory of the Soviet Union, systematic observations of the difference in air temperatures in the 0.5-2.0 m layer have been conducted at several dozen stations for a number of years. On the basis of these data, the present paper analyzes the character of the change of the temperature stratification in the ground layer of the atmosphere with time (daily and annual cycle) in different geographical areas and also under different weather conditions. Such studies are little known in the literature, especially as regards the analysis of mass observations.

The initial data used were obtained from systematic observations of the air temperature at levels of 0.5 and 2 m above the surface of the soil at 42 stations located in various geographical areas on the territory of the USSR (Fig. 1). The observations were made by means of suction psychrometers, and in the last few years, by using a method described in Handbook [1] and consisting in the following:

1) During the warm period of the year (at positive temperatures), the temperature measurements were made daily at 1 A.M., 7 A.M., 10 A.M., 1 P.M., 4 P.M., and 7 P.M. local mean solar time, and during the remaining time of the year, at 1 A.M. and 1 P.M.; 2) the suction psychrometers were mounted horizontally on the corresponding levels, and their bulbs (i.e., the reservoirs of the thermometers) were turned toward the wind; 3) during each of the periods, the thermometer readings at the 0.5 and 2 m levels were taken five times in the course of 10 to 13 min., and the mean temperature was calculated from five of its values during this period of time. It should be noted that this method was not kept the same during the entire period of systematic observations and had the following variations:

a) Up to 1965, the psychrometers at the two levels had different supports: at the 0.5 m level the instrument was suspended horizontally, and at the 2 m level, vertically;

b) Up to 1961, three thermometer readings were taken per day;

c) At some stations, no observations were made during the cold period of the year.

Among these deviations, the most interesting is the first, i. e., the possible errors in measurements of the temperature gradients, due to the different ways of suspending the psychrometers. This question was studied rather closely by V. V. Lazovskiy [2]; the errors were shown to be systematic in character and to lower the gradients at high values of the radiation balance. Comparison of factual data of gradient measurements for

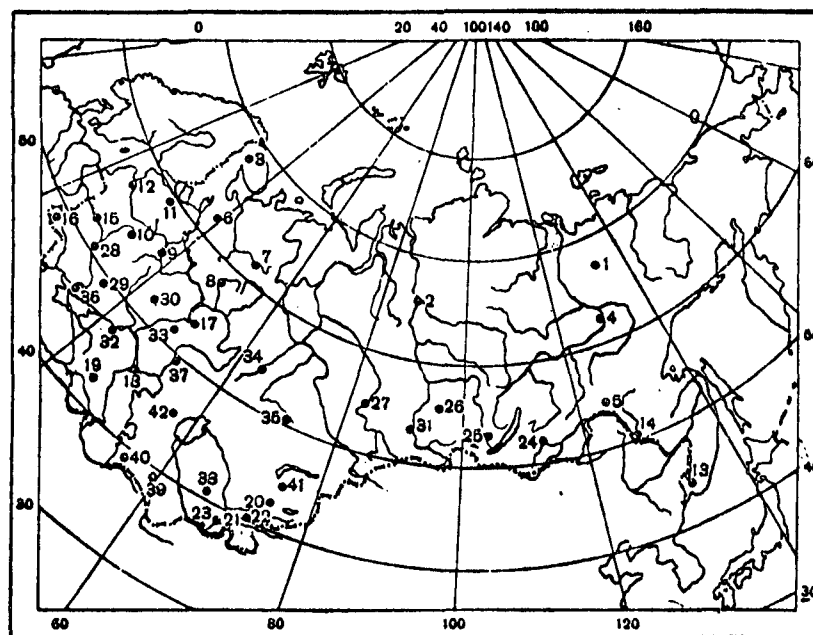


Fig. 1. Map of stations measuring temperature gradients.

different and for the same suspension of psychrometers carried out by T. A. Ogneva [3] for six stations confirmed this conclusion. An appreciable error is observed only at wind velocities higher than 2 m/sec; its absolute values increase with increasing radiation balance, particularly in areas of insufficient moisture. At a wind velocity of 2-4 m/sec and radiation balance values of 0.2-0.5 cal/cm<sup>2</sup> min, errors in the dry areas may amount to 0.2-0.3°C., and in areas of sufficient moisture, to 0.1-0.2°C. This must be taken into consideration in analyzing materials for different years. However, no appreciable jump in mean monthly values of the temperature gradient is observed as a result of switching to a different method, as demonstrated by Table 1. The latter gives mean monthly gradients ( $\Delta t$ ) for the different years for the time of highest radiation balance values (June, 1 P.M.). In Table 1 it is impossible to observe any substantial change

(namely, increase) of the temperature gradient beginning in 1965, when the method described in the Handbook [1] was introduced. On the whole, it is apparent that the existing series of observations can be used as a set of fully comparable data.

The first generalization of the observational material on temperature gradients for the network of stations was made by L. I. Prokof'yeva [4], but this pertained to a considerably smaller number of stations and volume of observations.

Table 1

Mean Monthly Values of the Temperature Gradient ( $\Delta t$ ) of Air at 13 h. in June

ZONE	STATION	YEAR							
		1960	1961	1962	1963	1964	1965	1966	1967
Forest	Nikolayevskoye	0,3	0,4	0,2	0,9	0,6	0,7	0,6	0,7
	Riga	0,3	0,3	0,3	0,4	0,2	0,6	0,7	0,7
	Yakutsk	1,2	0,6	0,6	0,5	0,8	0,7	0,8	1,2
	Beregovo	0,3	0,4	0,6	0,5	—	0,1	—	—
Steppe	Askaniya-Nova			0,8	1,1	0,5	1,2	—	0,4
	Gigant	0,5	0,4	1,1	0,4	0,2	0,6	0,4	
	Tselinograd		0,7	0,7	1,1	1,1	1,2	1,2	1,3
	Solyanka	0,2	0,5	0,4	0,4	0,8	0,8	0,4	0,8
	Poltava		0,6	0,3	0,8	0,4	0,8	0,5	0,7
Foothill Steppes	Kalmykovo	0,8	0,8	1,0	1,1	0,8	0,8	0,8	
	Dushanbe			0,4	0,4	0,8	0,8	0,4	0,2
Desert	Aydarly			1,5	0,8	1,5	1,2	1,3	1,3
	Beki-Bent				0,8	0,9	1,1	2,1	1,3
	Tamy			1,5	1,3	0,9	1,0	0,7	1,0

Table 2 gives mean values of the air temperature gradient in the 0.5-2 m layer for individual months of the year during the day (1 P.M.) and night (1 A.M.). These values were obtained by averaging the mean monthly values  $\Delta t$  at the given times over the existing period of observations, and the mean monthly  $\Delta t$ 's in turn were calculated by averaging single measurements in the course of each month for a specific time, 1 A.M. and 1 P.M.

Table 2 gives an idea of the characteristic values of the air temperature gradient in the lowest layer in different geographical zones from the desert to the conifer forests, in accord with the botanical classification based on the Geobotanical Map of the USSR [5]. The statistical reliability of the mean values  $\Delta t$  in the table varies; it depends on the total period of observations at any given station. Depending on the period of averaging, the characteristic values  $\Delta t$  themselves can change.

In order to explain possible differences in the average temperature gradients as a function of the period of averaging, we give Table 3, in which for stations having a series of observations of 8 to 13 years, there are average values obtained for observation periods of 5 years and 3 years. It was found that in the great majority of cases, differences do not exceed



Table 2

Mean Values of Temperature Gradient ( $\Delta t$ ) in the Daytime (1 P.M.) and at Night (1 A.M.) in Different Geographical Zones.

ZONE	STATION	NUMBER ON MAP	PERIOD OF OBSERVA- TIONS	$\Delta t$ at 1 A.M.						$\Delta t$ at 1 P.M.					
				V	VI	VII	VIII	IX	X	V	VI	VII	VIII	IX	X
Pretundra thin forest	Verkhoyansk	1	3	—	—	-0.6	-0.8	-0.3	—	—	—	0.9	0.7	0.4	—
Conifer forest	Turukhansk	2	3	—	-0.4	-0.6	-0.4	-0.2	—	—	0.4	0.5	0.4	0.2	—
	Khibiny	3	6	-0.3	-0.3	-0.3	-0.3	-0.3	-0.2	0.6	0.6	0.5	0.5	0.3	0.1
	Yakutsk	4	9	—	-0.8	-0.6	-0.5	-0.5	—	0.6	0.8	0.8	0.6	0.5	—
	Skovorodino	5	5	—	-0.3	-0.3	-0.4	-0.4	—	—	0.4	0.4	0.5	0.5	—
	Kargopol'	6	4	-0.2	-0.3	-0.3	-0.2	-0.2	-0.1	0.5	0.4	0.3	0.2	0.1	0.0
	Ust'-Vym'	7	4	—	-0.8	-0.9	-0.3	-0.3	—	—	0.6	0.3	0.3	0.2	—
	Nolinsk	8	6	-0.5	-0.6	-0.6	-0.6	-0.4	-0.2	0.2	0.3	0.4	0.3	0.2	0.1
	Sobakino	9	2	-0.5	-0.7	-0.4	-0.3	-0.4	—	0.4	0.4	0.4	0.3	0.1	—
Mixed forest	Smolensk	10	6	-0.5	-0.5	-0.4	-0.4	-0.4	-0.2	0.3	0.2	0.3	0.2	0.2	0.1
	Nikolayevskoye	11	9	-0.5	-0.5	-0.5	-0.4	-0.3	-0.2	0.4	0.5	0.5	0.4	0.3	0.1
	Riga	12	13	-0.5	-0.6	-0.5	0.4	-0.4	-0.2	0.4	0.4	0.4	0.3	0.2	0.2
	Primorskoye	13	3	-0.3	-0.1	-0.1	-0.1	-0.4	-0.3	0.5	0.5	0.5	0.4	0.4	0.4
Hardwood forest	Tolstovka	14	3	-0.5	-0.7	-0.4	-0.4	-0.2	—	0.7	0.7	0.6	0.4	0.5	—
	Pinsk	15	5	-0.4	-0.5	-0.6	-0.6	-0.4	-0.2	0.4	0.6	0.5	0.4	0.3	0.3
	Beregovo	16	8	-0.4	0.5	-0.5	-0.6	-0.6	-0.6	0.2	0.4	0.5	0.4	0.3	0.3
	Kuybyshev	17	8	0.6	0.8	0.8	-0.8	-0.6	-0.4	0.3	0.5	0.5	0.4	0.3	0.2
River flood plains	Astrakhan'	18	4	-0.4	-0.4	0.6	0.6	-0.4	-0.3	0.6	0.8	0.8	0.6	0.5	0.4
Steppe-covered meadows	Telavi	19	6	-0.4	0.4	-0.4	-0.4	0.3	-0.4	0.4	0.4	0.5	0.6	0.5	0.4

Table 2 (Cont'd)

ZONE	STATION	NUMBER ON MAP	PERIOD OF OBSERVA- TIONS	$\Delta t$ at 1 A.M.						$\Delta t$ at 1 P.M.					
				V	VI	VII	VIII	IX	X	V	VI	VII	VIII	IX	X
Foothill steppes	Frunze	20	4	-0.8	-0.8	-0.7	-0.7	-0.6	-0.5	0.7	0.9	1.0	1.1	0.9	0.6
	Dushanbe	21	6	-1.1	-1.4	-1.3	-1.2	-1.2	-1.0	0.4	0.4	0.7	0.8	1.1	0.9
	Fergana	22	4	-0.8	-1.1	-1.2	-0.9	-1.0	-0.7	0.9	0.6	0.6	0.8	0.9	0.9
Meadow steppes	Termez	23	7	-0.9	-0.8	-0.8	-0.9	-0.8	-0.8	0.5	0.6	0.9	1.1	0.9	0.6
	Chita	24	3	-0.3	-0.3	-0.2	-0.2	-0.3	-	1.0	1.1	0.8	0.8	0.9	-
	Khomutovo	25	5	-0.2	-0.2	-0.2	-0.1	-0.2	-	0.8	0.9	0.6	0.5	0.5	-
	Solyanka	26	9	-0.5	-0.5	-0.5	-0.5	-0.4	-	0.5	0.5	0.7	0.6	0.3	-
	Ogurtsovo	27	4	-0.4	-0.7	-0.9	-0.5	-0.5	-0.1	0.7	0.6	0.7	0.5	0.4	0.4
Typical steppes	Borispol'	28	8	-0.3	-0.4	-0.4	-0.3	-0.3	-0.2	0.6	0.5	0.6	0.4	0.4	0.4
	Poltava	29	8	-0.6	-0.6	-0.5	-0.6	-0.5	-0.4	0.5	0.6	0.5	0.5	0.4	0.3
	Kamemaya Step'	30	8	-0.6	-0.6	-0.6	-0.6	-0.5	-0.3	0.3	0.3	0.3	0.4	0.4	0.3
	Khakasskaya	31	7	-0.7	-0.6	-0.7	-0.5	-0.8	-0.5	0.7	0.8	0.7	0.7	0.6	0.3
	Gigant	32	8	-0.4	-0.4	-0.4	-0.4	-0.4	-0.3	0.4	0.5	0.8	0.7	0.6	0.3
Typical steppes	Yershov	33	2	-0.7	-0.5	-0.4	-0.5	-0.6	-0.5	0.8	1.1	0.8	1.0	0.8	0.6
	Rudnyy	34	4	-0.3	-0.3	-0.3	-0.3	-0.4	-0.2	0.7	0.9	0.8	0.8	0.7	0.3
	Tselinograd	35	6	-0.2	-0.2	-0.2	0.1	-0.2	-0.1	0.8	1.0	1.0	0.7	0.7	0.3
Steppe-covered deserts	Askaniya-Nova	36	10	-0.4	-0.4	-0.5	-0.4	-0.4	-0.3	0.8	0.9	1.1	1.0	1.0	0.5
	Kalmykovo	37	9	-0.1	-0.1	-0.2	-0.2	-0.2	-0.1	0.6	0.9	0.9	0.8	0.7	0.4
Deserts	Tandy	38	6	-0.5	-0.4	-0.5	-0.5	-0.6	-0.5	0.9	1.1	0.9	0.9	0.7	0.5
	Akmolla	39	2	-0.2	-0.2	-0.4	0.4	-0.3	-0.3	1.0	1.1	1.0	0.9	0.9	0.7
	Beki-Bent	40	4	-0.2	-0.4	-0.2	-0.3	-0.4	0.4	1.1	1.2	1.2	1.2	1.2	0.8
	Aydarly	41	5	-0.4	-0.6	-0.6	-0.7	-0.6	-0.4	1.2	1.3	1.2	1.1	1.0	0.7
	Churuk	42	2	-0.4	-0.4	-0.6	-0.4	-0.4	0.2	1.1	1.4	1.6	1.4	1.1	0.8

Table 3

Mean Values of Temperature Gradient ( $\Delta t$ ) for 5-Year and 3-Year Observation Periods.

Station	$\Delta t$ At 1 A.M.			$\Delta t$ At 1 P.M.		
	V	VII	IX	V	VII	IX
Mean $\Delta t$ for 5 Years						
Yakutsk	—	—0.6	—0.4	—	0.9	0.5
Riga	—0.5	—0.6	—0.4	0.4	0.4	0.2
Beregovo	—0.5	—0.5	—0.6	0.3	0.6	0.3
Solyanka	—0.5	—0.6	—0.4	0.5	0.8	0.5
Borispol'	—0.3	—0.4	—0.3	0.5	0.6	0.4
Poltava	—0.7	—0.5	—0.6	0.5	0.5	0.4
Gigant	—0.4	—0.4	—0.4	0.5	0.8	0.6
Askaniya-Nova	—0.4	—0.5	—0.4	0.7	1.0	1.1
Kalmykovo	—0.1	—0.1	—0.2	0.7	0.8	0.7
Kuybyshev	—0.6	—0.8	—0.6	0.3	0.4	0.2
Mean $\Delta t$ for 3 Years						
Yakutsk	—	—0.6	—0.4	—	0.9	0.4
Riga	—0.5	—0.6	—0.5	0.4	0.6	0.2
Beregovo	—0.5	—0.4	—0.5	0.3	0.6	0.3
Solyanka	—0.4	—0.6	—0.4	0.6	0.8	0.5
Borispol'	—0.3	—0.3	—0.3	0.6	0.7	0.6
Poltava	—0.6	—0.6	—0.5	0.5	0.5	0.4
Gigant	—0.4	—0.4	—0.4	0.5	0.8	0.8
Askaniya-Nova	—0.4	—0.4	—0.5	0.6	0.9	1.2
Kalmykovo	—0.1	—0.1	—0.2	0.8	0.8	0.6
Kuybyshev	—0.5	—0.8	—0.7	0.4	0.4	0.3

0.1°C. for averages for 5 years as well as averages for 3 years, particularly for the nighttime. In dry areas, during the day, the differences occasionally reach 0.2°C., but the absolute values of  $\Delta t$  are sufficiently high in these areas. It may be postulated that the three-year series of observations of air temperature gradients are sufficiently stable. On this basis, the data of Table 2 may be considered comparable to a certain extent, while the values of the temperature gradients are characteristic diurnal and nocturnal mean values during the warm time of the year for open areas of the natural surface of different landscape zones. (Measurements of gradients are made on sites of meteorological stations, on which the plant cover natural for the given zone is retained, and the sites themselves are usually located in places having little protection).

On the basis of an inspection of Table 2, we shall note the following pattern in the change of the temperature gradients. According to the sign of the radiation balance of the underlying surface, a difference of the sign of the gradients in the daytime and nighttime is observed.

In drier areas (steppes, deserts), an increase of the absolute values of diurnal gradients to 1-1.5°C. is observed as compared to the forest zone, where they essentially do not exceed 0.5°C. Occasionally, some

deviations are also observed. For example, in Eastern Siberia (Yakutsk, Verkhoyansk) and beyond the polar circle (Khibiny), the gradient values are somewhat higher than in the more southern parts of the conifer forests. This was noted earlier by N. I. Budyko [6]. It is due both to a latitudinal decrease of the air temperature and to the specific location of the ground station. (The Yakutsk station pertains, on the whole, to the zone of the middle taiga forests, but is situated in the subzone of steppe-covered meadows in combination with parts of forests.)

In the zone of meadow steppes of the European territory of the USSR (Borispol', Poltava, Kamennaya Step'), the gradients are somewhat reduced as compared with the analogous zone of the Asian part (Chita, Khomutovo, Solyanka, Ogurtsovo), and they are increased in southern regions of sufficient humidification (zone of steppe-covered subalpine meadows and foothill steppes). This characteristic is related to the higher values of the radiation balance on the European territory of the USSR.

At night, a certain decrease of the gradients is noted (in absolute value) in the steppe and desert zones as compared with the forest zone, owing to the higher wind velocities in these regions. The highest gradients at night are observed in the southern regions of sufficient humidification and at sheltered stations (Dushanbe, Fergana, Termez). In the remaining regions, the average values of the gradients basically do not exceed  $0.5^{\circ}\text{C}$ .

The annual cycle displays a definite variation of the average monthly values of daytime temperature gradients. The highest values are observed in June-July, and they decrease in autumn. This decrease begins for the forest zone in September, for the steppe zone in October, and for the deserts, still later. The variation of the night gradients in the annual cycle is less distinct. Only in autumn is a decrease of gradients observed in most regions, as compared with the summer months. In regions of irrigation farming (zone of foothill steppes) a shift of the highest gradient values (relative to the highest radiation balance values) to August-September is observed; it is due to a decrease of irrigation during this period and to the decisive importance of radiation factors as compared to the humidification of the soil.

Table 4 shows frequencies of different values of the gradients as a function of the cloud cover (clear, overcast, variable cloudiness) for five stations of different landscape zones. The frequencies were calculated on the basis of observations for three years (1965-1967). The "clear" gradation included cases where, during the observations, clouds were absent on the solar disk and in a zone of  $5^{\circ}$  around it, and the "overcast" gradation was one in which the solar disk was covered by a thick cloud layer.

The data of the table confirm a sufficient reliability of the average gradient values used in the study, and also show possible changes of these values because of weather conditions. For example, for the Nikolayevskoye station in the summertime, variable cloudiness is most typical (as demonstrated by the number of cases of observations compared with their number

Table 4

Frequency (%) of Gradients of Various Limits Under Various Weather Conditions

Month	Gradation of $\Delta t$ at 1 A.M.							Number of cases	Gradation of $\Delta t$ at 1 P.M.							Number of cases
	$<-2$	$0-2$ 10-1.1	$0-1$ 10-0.6	$0-0.5$ 10-0.2	$\pm 0.1$	$>0.1$	$<-0.1$		$\pm 0.1$	$0.2-0.5$	$0.6-1.0$	$1.1-2.0$	$>2$			
Yakutsk																
Clear																
VI	5	28	26	34	7		57			10	32	53	5	19		
VII	8	20	29	35	8		51				28	72		25		
VIII	3	22	39	25	8	3	36		10	10	80			10		
IX		13	14	44	24	5	37		25	12	63			8		
Variable cloudiness																
VI		24	24	34	6	12	17		2	10	51	35	2	51		
VII		4	29	30	37		27		2	15	33	50		46		
VIII			25	58	17		12			28	53	19		32		
IX				84	16		6		13	51	26	10		31		
Overcast																
VI			6	50	44		16		20	50	30			20		
VII				26	67	7	15		45	32	23			22		
VIII			7	21	65	7	14		35	60	5			20		
IX		6	12	18	52	12	17		47	48	5			21		
Nikolayevskoye																
Clear																
IV		26	26	48			23	80		20				10		
V	5	8	12	43	24	8	64			18	55	27		11		
VI		18	35	30	10	7	60		11	22	45	22		9		
VII		10	31	46	13		71			40	20	40		5		
VIII		5	23	42	14		64	33	67					3		
IX		9	20	41	28	2	54		43	57				7		
X		2	11	45	40	2	45		20	80				5		
XI			11	45	33	11	9	67		33				3		
Variable cloudiness																
IV				100			1	61	22	6	11			18		
V			31	38	31		16	2	11	37	30	20		56		
VI			17	39	44		18		4	26	32	36	2	57		
VII			12	41	41		17		12	21	46	21		67		
VIII				53	47		19	2	32	28	28	10		68		
IX				10	90		10	4	27	27	38	4		55		
X				50	50		14	9	36	40	15			33		
XI			10	20	70		10	15	70	15				13		
Overcast																
IV			17	33	50		6	50	50					2		
V		8	15	15	47	15	13		50	35	15			26		
VI		8		25	50	17	12		61	30	9			23		
VII				100			5	14	52	33	5			21		

Table 4 (Cont'd)

Month	Gradation of $\Delta t$ at 1 A.M.						Number of cases	Gradation of $\Delta t$ at 1 P.M.						Number of cases
	$<-2$	$0, -2$ $20-1, 1$	$0, -1$ $20-0, 6$	$0, -0, 5$ $20-0, 2$	$-0, 1$ $20-1, 1$	$>0, 1$		$<-0, 1$	$\pm 0, 1$	$0, 2-0, 5$	$0, 6-1, 0$	$1, 1-2, 0$	$>2$	
VIII				11	89		9	5	45	45	5			22
IX				16	80	4	25	4	65	21				28
X				29	71		34	4	74	22				55
XI					100		12		100					14

Solyanka														
Clear														
V	2	26	27	31	12	2	51			18	46	36		11
VI	3	23	30	36	8		60	8	8	15	44	15		13
VII	4	27	28	28	13		68				56	44		18
VIII	3	26	25	31	14	1	69		8	8	46	38		13
IX		23	32	34	11		53			14	65	21		14

Variable cloudiness														
V				50	40	10	10		14	20	31	35		51
VI		11		67	22		9	7	4	9	48	32		56
VII		9		36	55		11		6	11	38	45		55
VIII		20	20	40	20		5	2	8	23	50	15	2	52
IX				71	29		7	3	5	32	54	14		37

Overcast														
V			6	19	69	6	32		60	33	7			30
VI			5	19	76		21	5	47	24	24			21
VII				14	65	21	14		55	40	5			20
VIII	5			37	47	11	19		68	21	11			28
IX		3		13	81	3	30	3	71	23	3			39

Gigant														
Clear														
IV		2	11	65	15	7	46		7	15	70	8		14
V	1	9	23	53	14		70		13	32	43	22		23
VI		9	20	50	19	2	58		7	12	50	31		16
VII		5	32	45	13	5	75		6	20	40	30	4	30
VIII	1	4	15	47	33		73		4	17	33	46		24
IX	3	6	7	50	32	2	72		2	18	36	44		34
X	6	7	11	47	27	2	71		2	18	54	26		35
XI				14	81	5	22			62	38			8

Variable cloudiness														
IV			15	35	50		6		16	26	45	13		49
V		13	12	38	12	25	8		9	30	40	19	2	47
VI			7	50	43		14		13	38	36	13		52
VII		14	29	43	14		7			28	35	35	2	51
VIII			25	50	25		8		10	29	30	29	2	50
IX			14	29	57		7	5	14	35	36	10		49
X							3	2		37	51	10		43
XI							3	3		53	25			32

Table 4 (Cont'd)

Month	Gradation of $\Delta t$ at 1 A.M.						Number of cases	Gradation of $\Delta t$ at 2 P.M.						Number of cases
	$<-2$	or $-2$ to $-1.1$	or $-1$ to $-0.6$	or $-0.5$ to $-0.2$	$\pm 0.1$	$>0.1$		$<-0.1$	$\pm 0.1$	$0.2-0.5$	$0.6-1.0$	$1.1-2.0$	$>2$	
Overcast														
IV			6	26	60	8	38		70	27		3		27
V				33	67		15		45	50	5			18
VI			5	17	78		18	22	41	37				22
VII			18	46	36		11		42	58				12
VIII				33	58	8	12		22	45	33			9
IX				46	46	8	11		29	57	14			7
X				30	65	5	20		47	53				15
XI				6	89	5	35		35	65				20
Kalmykovo														
Clear														
IV		3	4	43	36	14	73	4	12	20	44	20		25
V		2	4	39	44	11	56			4	32	60	4	25
VI			4	42	41	13	46		22	11	11	45	11	9
VII		4		32	53	11	53		13		30	57		23
VIII	2		11	20	47	20	54		15	15	10	60		20
IX		2	4	55	37	2	46	12	24	12	18	34		17
X			12	26	50	12	42		6	12	63	19		16
Variable cloudiness														
IV				33	67		3		27	24	39	10		15
V					100		2		34	9	27	30		23
VI			17	50	33		6		16	14	26	44		43
VII				50	50		4		31	3	19	44		32
VIII				33	67		3	3	35	21	6	32	3	34
IX				25	75		8		27	3	43	21	6	33
X					80	20	5		14	32	40	14		28
Overcast														
IV					57	43	14		32	47	21			19
V				25	75		4		72	28				14
VI			12	88			8		88		12			8
VII				100			5		90		10			10
VIII				75	25	4			33	45	22			9
IX				100		6			50	30	20			10
X		13	13	61	13	15			55	28	17			18

in clear and overcast weather). During variable cloudiness, the probability of gradients from 0.2 to 1°C. is in excess of 60% during all the summer months, and the average value is 0.4-0.5°C. (see Table 2).

Table 4 also shows how the gradients change with the weather conditions and in the annual cycle. Thus, in clear weather at the Nikolayevskoye station in May-June, the probability of gradients above 0.6°C. during the day

is 60%, in the presence of variable cloudiness 50-70%, and in cloudy weather it does not exceed 15%; this pattern is observed to a greater or lesser degree at all the other stations regardless of the geographical zone.

The probability of formation of high gradients increases from spring to summer and decreases toward autumn, whereas the probability of low gradient values increases toward autumn. At night, the same pattern is observed as during the day: the frequency of high gradients (in absolute value) in clear weather is higher than in cloudy weather; in autumn, the frequency of low gradients is higher than that of high gradients under all weather conditions.

The frequency of gradients above  $\pm 2^{\circ}\text{C}$ . is slight; it amounts to a few percent and is observed only under clear weather or variable cloudiness conditions.

Table 5 lists maximum and minimum values of the temperature gradients  $\Delta t$ . The data were selected from observations in the last 3-5 years at 1 A.M. and 1 P.M. during all months of the year. Despite the "spottiness" of the extreme values, the following general features may be pointed out: 1) the gradients observed in the last five years do not exceed  $+4.5^{\circ}\text{C}$ . during the day and  $-4.7^{\circ}\text{C}$ . at night; 2) the extreme values of the gradients during the day and at night are chiefly observed during the summer months.

Table 5

Extreme Values of Temperature Gradient ( $\Delta t$ ) in Various Landscape Zones.

Station	Period of observations	Maximum		Minimum	
		$\Delta t$	Month	$\Delta t$	Month
Verkhoyansk	2	2.5	VII	-3.7	VIII
Turukhansk	3	2.7	VII	-4.3	VI
Khibiny	5	3.4	VIII	-3.6	VIII
Yakutsk	4	3.0	V	-3.4	VII
Skovorodino	5	2.3	VII	-3.2	X
Kargopol'	4	3.9	V	-2.9	IV
Nolinsk	3	2.2	VI	-3.6	VIII
Sobakino	5	2.5	VI	-3.5	X
Smolensk	5	1.7	IV	-3.6	VIII
Nikolayevskoye	5	3.3	VII	-3.7	VII
Riga	5	2.8	VI	-3.7	VI
Pinsk	5	1.9	IX	-3.6	IX
Beregovo	5	2.4	VIII	-3.5	VIII
Kuybyshev	3	1.9	V	-3.7	IX
Astrakhan'	3	3.2	VI	-3.4	VI
Telavi	5	3.3	VI	-3.2	IX
Frumze	4	2.9	V	-3.6	XI
Dushanbe	3	2.8	V	-4.6	V
Fergana	5	3.7	V	-4.0	VII
Chita	4	3.9	VII	-2.4	XI
Khomutovo	5	2.6	VII	-2.8	IX
Solyanka	5	3.7	VIII	-3.2	VII
Ogurtsovo	4	3.1	VII	-3.1	V
Borispol'	5	3.7	VI	-4.7	VII
Poltava	5	2.3	V	-3.4	V
Kamennaya Step'	5	3.1	VI	-4.0	IX
Khakasskaya	2	2.7	VI	-4.7	II
Yershov	3	3.8	VI	-3.8	V
Rudnyy	4	2.1	VII	-2.4	IV
Tselinograd	5	3.1	VIII	-1.4	IX
Tamdy	5	2.8	VII	-3.0	VIII
Beki-Bent	5	4.5	IV	-3.7	I
Aydarly	5	4.5	VIII	-4.3	I
Churuk	3	3.4	VIII	-2.9	VII



Table 6 shows the daily cycle of temperature gradients averaged over the observation period in different months of the year for five stations located in different landscape zones. The values for the 1 A.M. and 1 P.M. periods analyzed here actually correspond to the highest values of the gradients (in absolute value) for the daytime and nighttime hours; only in some cases, in autumn at stations with a sufficiently humidified surface (Dushanbe, Pinsk), and in winter in the desert (Beki-Bent), can the gradients at 7 P.M. be higher (in absolute value) than at 1 P.M. On the whole, the variation of gradients in the daily cycle follows the radiation balance cycle. In the summertime, positive gradients are observed during the 7 A.M. - 4 P.M. period in all the landscape zones, and only their absolute values change. In the forest zone (Riga, Pinsk) and in the presence of sufficient humidification of the surface (Dushanbe), the gradients at 4 P.M. do not exceed  $0.3^{\circ}\text{C}$ ., and in the steppe and desert zone (Askaniya-Nova, Beki-Bent) reach  $0.7-0.8^{\circ}\text{C}$ . In the wintertime, positive gradients at the Dushanbe and Beki-Bent stations are observed at 10 A.M. to 1 P.M., or 1 P.M. to 4 P.M., which are also determined by the possibilities of radiation influx of heat.

In general, the values of the air temperature gradients change with the relative proportion of the components of the heat balance. In a simplified manner, the proportion of the components may be characterized by the radiation balance (B), the state of humidification of the surface and the wind velocity. Fig. 2 shows graphs of the relationship of the temperature gradients to the radiation balance for three limits of wind velocity (less than 2 m/sec, from 2.1 to 4 m/sec, and above 4 m/sec.) and for four landscape zones - conifer and mixed forests, steppe, and desert. The graphs were plotted on the basis of average data from selections of single temperature gradients at the corresponding limits; the points correspond to mean  $\Delta t$  values at the given limit based on various stations, and the shaded area corresponds to the area of possible changes of mean gradients in any given group. The material used pertains chiefly to 1965, and the averages were obtained from data of dozens of single measurements.

The graphs give an idea of the mean temperature gradients above the natural surface of open stretches of dry land in the main landscape zones as a function of the wind velocity and magnitude of the radiation balance within  $0.1-0.2^{\circ}\text{C}$ . They may be useful for practical applications in evaluating the character of temperature stratification. The graphs show an obvious decrease of temperature gradients with increasing wind velocity at both positive and negative values of the radiation balance in the zone of forests and steppes; in the desert zone, the most positive gradients were found to arise at a wind velocity of 2-4 m/sec., and not at low velocities. It is evident from the graphs that the sign of the gradient changes to positive in the forest zone at higher values of the radiation balance than in the zone of steppes and deserts; as the wind velocity increases, the sign of the gradient changes at a high value of the radiation balance.

Table 6

Annual Cycle of Air Temperature Gradients in Different Months of the Year

Station	Period of Observation	Month	Time of observations, h.					
			1 A.M.	7 A.M.	10 A.M.	1 P.M.	4 P.M.	7 P.M.
Riga	1	IV	-0.1	0.0	0.2	0.5	0.3	0.0
	10	V	-0.5	0.2	0.4	0.4	0.3	-0.1
	12	VI	-0.6	0.2	0.4	0.4	0.2	-0.1
	12	VII	-0.5	0.2	0.4	0.3	0.2	0.0
	12	VIII	-0.4	0.1	0.3	0.3	0.2	-0.2
	11	IX	-0.4	0.0	0.3	0.2	0.1	-0.6
	10	X	-0.2	-0.1	0.1	0.2	0.0	-0.3
Pinsk	4	IV	-0.3	0.1	0.4	0.3	0.2	-0.3
	4	V	-0.4	0.0	0.3	0.4	0.0	-0.4
	4	VI	-0.6	0.2	0.6	0.6	0.3	-0.2
	4	VII	-0.6	0.2	0.5	0.5	0.2	-0.3
	3	VIII	-0.6	0.0	0.3	0.4	0.2	-0.1
	4	IX	-0.4	0.0	0.2	0.3	0.0	-0.6
	4	X	-0.2	-0.1	0.3	0.2	0.0	-0.3
Dushanbe	3	XI	-0.1	-0.1	0.0	0.1	-0.1	-0.2
	5	I	-1.0	-1.0	0.2	0.5	-0.3	-0.9
	5	II	-0.7	-0.6	0.2	0.5	0.1	-0.6
	5	III	-0.7	-0.5	0.5	0.6	0.1	-0.6
	5	IV	-0.9	-0.1	0.5	0.4	0.0	-0.8
	5	V	-1.2	0.1	0.5	0.5	0.0	-1.2
	5	VI	-1.4	0.1	0.6	0.5	-0.1	-1.7
Askaniya-Nova	6	VII	-1.3	0.1	0.7	0.7	0.2	-1.3
	5	VIII	-1.2	0.0	0.8	0.9	0.3	-1.4
	5	IX	-1.2	0.0	1.1	1.2	0.1	-1.3
	5	X	-1.0	-0.2	0.9	0.9	-0.1	-1.2
	5	XI	-0.9	-0.9	0.6	0.8	-0.4	-1.1
	4	XII	-1.0	-1.3	0.6	0.6	-0.4	-1.0
	7	IV	-0.3	0.2	0.6	0.8	0.4	-0.3
Beki-Bent	8	V	-0.4	0.3	0.8	0.8	0.5	-0.2
	8	VI	-0.4	0.4	0.9	0.9	0.5	-0.1
	8	VII	-0.4	0.4	0.9	1.1	0.6	-0.1
	9	VIII	-0.4	0.3	1.0	1.0	0.7	-0.1
	9	IX	-0.4	0.2	0.7	1.0	0.5	-0.4
	8	X	-0.3	0.0	0.5	0.7	0.3	-0.4
	4	I	-0.3	-0.3	0.0	0.3	0.1	-0.4
Beki-Bent	4	II	-0.2	-0.3	0.1	0.4	0.3	-0.3
	4	III	-0.3	-0.2	0.5	0.8	0.6	-0.2
	4	IV	-0.2	0.1	0.6	0.9	0.6	0.0
	4	V	-0.2	0.2	0.7	1.0	0.7	0.0
	4	VI	-0.4	0.3	0.9	1.2	0.8	0.0
	3	VII	-0.2	0.2	0.6	0.8	0.3	0.1
	3	VIII	-0.3	0.0	0.6	0.6	0.5	0.0
Beki-Bent	3	IX	-0.3	0.0	0.5	0.8	0.4	-0.1
	3	X	-0.3	-0.1	0.4	0.6	0.2	-0.3
	3	XI	-0.4	-0.3	0.1	0.4	0.1	-0.4
	2	XII	-0.2	-0.2	-0.1	0.0	-0.2	-0.2

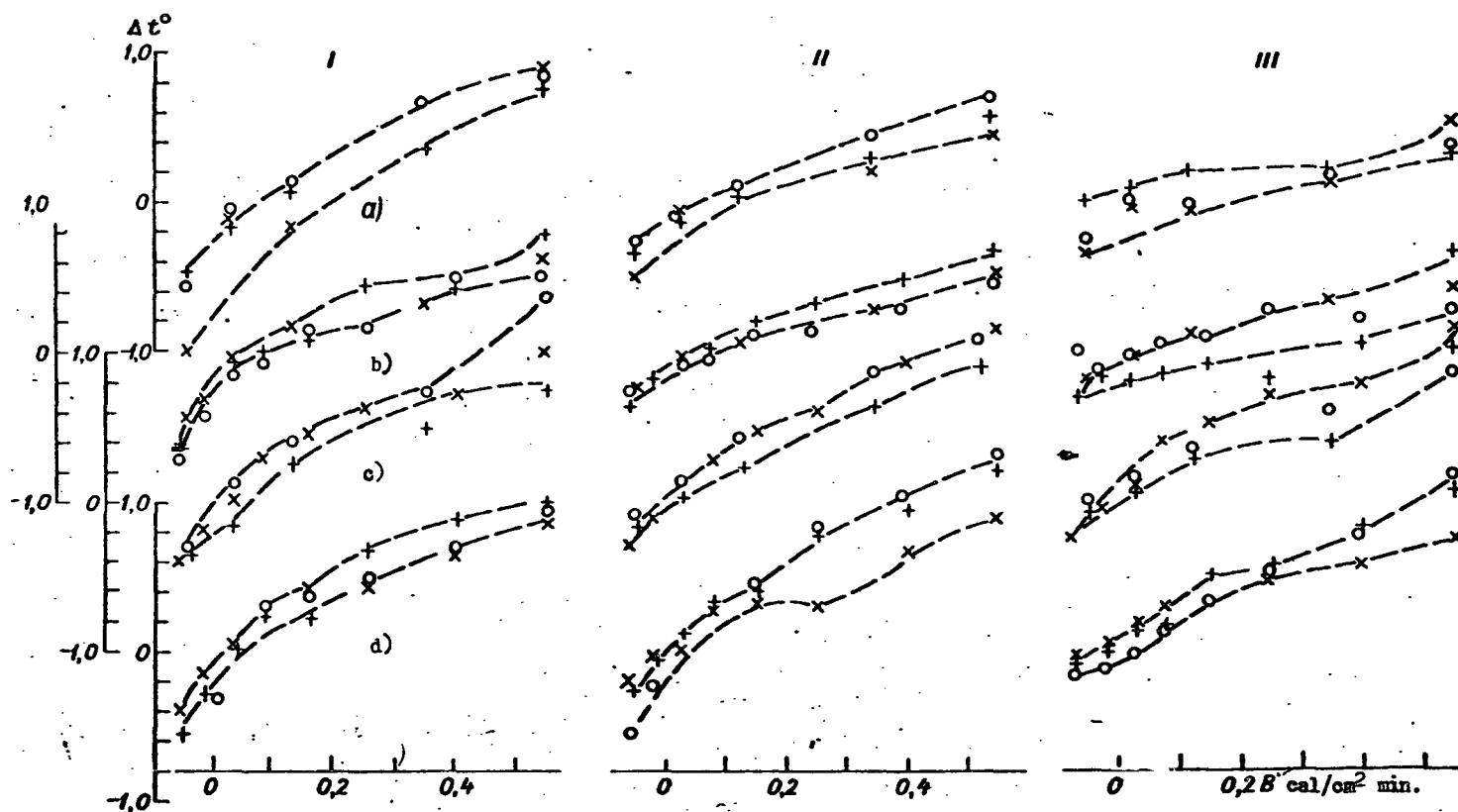


Fig. 2. Relationship of temperature gradients ( $\Delta t$ ) to radiation balance ( $B$ ) and wind velocity ( $u_2$ ) in various landscape zones.

I -  $u_2 < 2 \text{ m/sec}$ ; II -  $u_2 \sim 2.1-4 \text{ m/sec}$ ; III -  $u_2 > 4 \text{ m/sec}$

a) - conifer forest (O Turukhansk, x Ust'-Vym', + Skovorodino) b) - mixed forest (O Pinsk, x Sobakino, + Nikolayevskoye)  
c) - steppe (O Askaniya-Nova, x Chita, + Ogurtsovo); d) - desert (O Ardarly, x Tamdy, + Beki-Bent).

As was mentioned earlier, the graphs of Fig. 2 were plotted from mean values of  $\Delta t$  at a given limit. However, a rather close relationship between the temperature gradient and the radiation balance is also obtained from individual measurements. As an example, Fig. 3 illustrates such a graph for the Solyanka station. It was plotted from mean decade values of  $\Delta t$  for specific periods of various months (denoted by points) and radiation balance values corresponding to them, and also from maximum values of  $\Delta t$  for the same decade (denoted by crosses); observations for May-September 1965 and 1966 at wind velocities from 2 to 4 m/sec were used. The correlation coefficient was found to be 0.81. For other stations (Borispol', Ogurtsovo, Khomutovo) in this velocity range the correlation coefficient is respectively equal to 0.63, 0.68, and 0.71, and for velocities above 4 m/sec, to 0.50, 0.70, and 0.77. The coefficients (a, b) in regression equations of the type  $\Delta t = a + bB$ , obtained from these data for stations of the steppe zone, were also found to be rather close, as shown in Table 7.

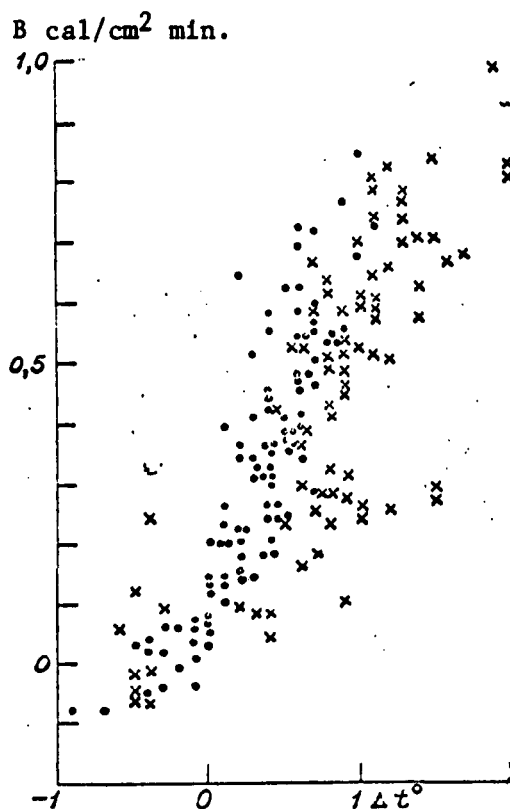


Table 7

Values of Coefficients a and b in Regression Equations for Wind Velocity of 2-4 m/sec for Stations of the Steppe Zone.

Station	a	b
Khomutovo	0,10	1,47
Solyanka	0,17	1,80
Ogurtsovo	0,21	1,40
Borispol'	0,15	1,08
Gigant	0,10	1,34
Tselinograd	0,02	2,25

Fig. 3. Relationship of temperature gradients ( $\Delta t$ ) to radiation balance (B) for Solyanka station at wind velocity of 2-4 m/sec.

In conclusion, we shall give a general characterization of possible conditions of temperature stratification. We shall use for this purpose the known parameter  $\Delta t/u_1^2$  ( $u_1$  being the wind velocity at a level of 1 m above the surface of the soil), calculated from mean monthly values for the individual years. For July at 1 P.M. in the steppe and desert zones, the average  $\Delta t/u_1^2$  is 0.10-0.15 (stations of Beki-Bent, Aydarly, Askaniya-Nova, Gigant, Astrakhan', Khomutovo, Solyanka); these values are determined by the high values of the temperature gradients in these regions. The same values of  $\Delta t/u_1^2$  are observed at the Beregovo and Skovorodino stations located in the forest zone and having lower gradients and low wind velocities. The highest mean value  $\Delta t/u_1^2 = 0.4$  was observed in Dushanbe, this being the result of the relatively high temperature gradients and a wind velocity not exceeding an average of 1.5 m/sec in July. In the forest zone at 13 h in July, the average  $\Delta t/u_1^2$  amounts to 0.03-0.10.

At night, the parameter of the temperature stratification in absolute value exceeds the daytime values, changes considerably with data of the individual stations, and amounts to  $>0.5$  for a majority of the regions; at stations with high diurnal values of  $\Delta t/u_1^2$ , the nighttime values amount to  $<-1.0$ .

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# EXPEDITIONARY STUDY OF THE POLLUTION OF THE AIR RESERVOIR OF INDUSTRIAL CITIES

N. S. Burenin, B. B. Goroshko and B. N. P'yantsev

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p. 100-108, (1968).

## 1. Organization of Observations Based on Collection of Air Samples

In order to conduct a successful drive for purity of atmospheric air in cities and to adopt active measures for decreasing the emission of noxious substances into the atmosphere, it is necessary to know the degree of pollution of the air reservoir of the city and the meteorological conditions leading to the accumulation of impurities in the lower layer of the atmosphere.

To this end, an expedition with separate groups for studying the state of pollution of an air reservoir under urban conditions was created by the A. I. Voeykov Main Geophysical Observatory. The present paper presents the main principles of the work of these groups and some of the results obtained.

In carrying out expeditionary studies it is necessary to consider that the territory of cities may contain not only large enterprises that constitute major sources of noxious discharges, but also many small enterprises with relatively small discharges. The general background of atmospheric pollution of a city is produced by the activity of all these sources. According to the existing arrangement of industrial enterprises on their territory, cities may be divided into two types. The program of investigations can be selected and conducted in accordance with the type of city.

The first type includes cities where the industry and hence the noxious discharges are concentrated in some part of the city on a single industrial site. In such cases, the direction of the plume from the complex of enterprises plays a major part in the pollution of the city districts. The presence of a small number of small sources scattered over the entire city will create an additional, less substantial pollution. For this reason, in studying atmospheric pollution in this case, it is first necessary to concentrate one's attention on determining the surface concentrations in the zone of influence of the plume originating from the main industrial site at various distances from its center. The concentration of ingredients that are discharged by the industrial enterprises was determined.

The second type of cities is characterized by a more uniform arrangement of major sources and a large number of minor ones, which make a great contribution to the pollution of the atmosphere. In these cases, in order

to obtain the main characteristics of pollution of the air reservoir of a city, a network of points is organized for taking air samples in characteristic areas equipped with special booths. This makes it possible to create representative sampling points and to obtain reliable results. For a more detailed evaluation of the field of concentrations, observations are made at points located along a preselected route. An automobile specially equipped for taking samples moves along this route. This makes it possible to increase substantially the number of sampling points on the territory of the city, move rapidly the sampling points to various areas of the city by changing the route of the automobile, obtain additional information on the state of atmospheric pollution over the entire area of the city, and to determine the degree of pollution of various neighborhoods, and so forth. The selection of the route is determined by the location of approach roads, residential sections, and pollution sources, by the arrangement of stationary sampling points, and by the particular objective to be reached as the result of the observations.

In studying air pollution, the problems of propagation of noxious impurities from various industrial sources are of major interest. To solve these problems, the main sources of atmospheric pollution on the territory of the city studied are determined. Such sources include individual stacks or groups of stacks of industrial or power plants, as well as random discharges of noxious substances into the atmosphere reaching the latter from sources other than stacks. In order to evaluate the contribution of each of the main sources to the pollution of the city's air reservoir and determine the range of propagation of noxious impurities, the sampling is organized under the plumes to determine the ingredients which are characteristic of these sources. The samples are taken at different distances from the source (0.5, 1, 2, 3, 5, 8, 10, 15 and 20 km). The number of distances and magnitude of the distance of the sampling points from the source are determined up to the distances where the plume is still detected. This in turn will be determined by the height and capacity of the discharge and the meteorological conditions causing the propagation and dispersal of the impurities. Inasmuch as large-size plumes frequently spread beyond the city limits, the data taken under the plume will permit an evaluation of the magnitude of expected concentrations in areas of prospective construction of residential sections.

In order to cover the various meteorological conditions that are known to have a strong influence on the propagation of impurities and to determine the average daily variations of the concentrations, the sampling is carried out along a grazing graph. [Ed. note: Kymograph]. According to the latter, a daily alternation of morning and evening changes, which also include the daytime, is obtained.

The sampling and their chemical analysis in laboratories were carried out according to the recommendations of the A. I. Voyeykov Main Geophysical Observatory, whose principles are matched with the techniques of the Health Ministry of the USSR [7]. Samples to be analyzed for gaseous ingredients are usually taken by means of U-shaped absorbers filled with an absorbing solution through which the air is drawn at a certain rate by various types of stimulators. The dust concentration is determined by a gravimetric method, the sample being separated on filters made of FPP-15 cloth.

In addition to the collection of samples of atmospheric air, many additional observations are made at meteorological and aerological stations, including gradient observations, and an additional release of a surface radiosonde. Data of gradient observations are used to calculate an important characteristic such as the coefficient of turbulent exchange, and the radio sounding makes it possible to obtain the distribution of the meteorological elements in height. The air temperature and humidity and the wind velocity and direction at a single level are measured at the points of sampling. These data are necessary for the study of the microclimatic characteristics of the city, whose influence on the field of concentrations should be considerable.

Of great importance for the analysis of the field of concentration is the knowledge of the characteristics of discharges of industrial enterprises and allowance for the operation of purifying installations. For this reason, data on discharges at the main industrial facilities of the city are constantly being corrected.

## 2. Preliminary Analysis of Observational Results.

The establishment of relationships between the pollution of a city's atmosphere and the change of the meteorological elements, the arrangement of pollution sources and other factors determining the magnitude of surface concentrations is relatively difficult, since many of these factors are variable. However, in the presence of a large number of observations, the statistical method makes it possible to obtain a series of relationships. We shall consider the data for mean monthly concentrations of phenol for four months representing different seasons. These data were obtained from regular observations at points located in characteristic areas of one of the cities. Fig. 1 shows a diagram of this city, indicating the boundaries of the residential area and the location of the points of sampling. The same figure shows lines of equal concentrations based on data of average monthly values of phenol concentrations for February (a), April (b), June (c), and November (d). The figure clearly shows the area of maximum air pollution, located in the northern part of the city, with its center at the main source of discharge of phenols. In all seasons of the year, a slower decrease of the concentrations in the direction southeast of the center of the city than in the other directions is observed. In this part of the city, there are no additional major sources of discharges, but a heavy flow of automobile traffic is observed, and the narrow streets of the old part of the city promote the accumulation of noxious substances. Fig. 1 also indicates a change of average monthly concentrations from one season to the next. The lowest phenol concentrations were observed in autumn, in November, when the concentrations on the whole were considerably lower over the entire territory of the city than in the remaining seasons. The average maximum monthly concentrations are observed in spring. The data show that the air reservoir over the entire territory of the city is heavily contaminated with phenols, particularly its northern part, i.e., the area of new construction.



In the study of pollution of the environment, considerable attention is given by Soviet [7, etc.] and foreign authors to the analysis of the daily and annual distribution of pollutants in the atmosphere. It is well known that these distributions depend on the synoptic and meteorological conditions and also on the conditions of operation of pollution sources such as industrial enterprises, automobile transportation, the heating system, etc. When a plot is made for the curves of the annual variation of mean monthly phenol concentrations in April and September, based on observational data at two points of the city, a distinct daily variation is observed with a concentration maximum in the morning and evening and a minimum at 1-2 P.M. We plotted the distribution curves of sulfur dioxide for September and October based on the data of two cities. The shape of the curves is similar. The marked decrease of surface concentrations of phenol and sulfur dioxide during the day is probably caused by meteorological conditions, since discharges into the atmosphere should not increase during this period.

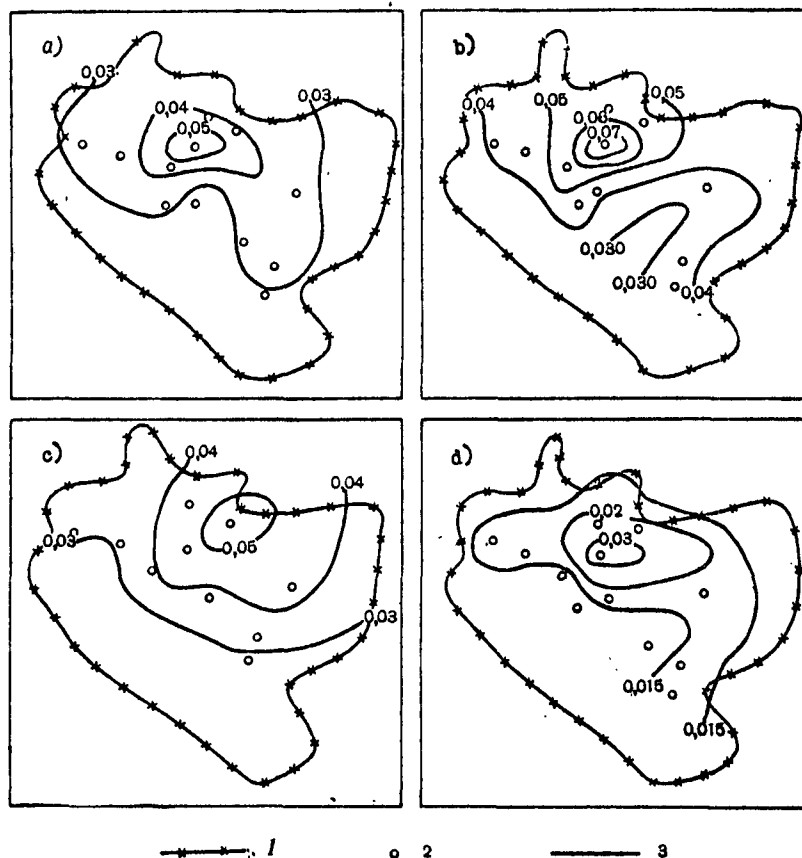


Fig. 1. Distribution of phenol concentrations over the city in various months of the year: February (a), April (b), June (c), November (d).

1 - boundary of residential section of city, 2 - points of collection of air samples, 3 - lines of equal phenol concentrations ( $\text{mg}/\text{m}^3$ ).

The daily variation of mean monthly concentrations of sulfur dioxide for a summer month (July) in the interval from 6 A.M. to 9 P.M. shows slight fluctuations with a maximum, on the contrary, during daytime hours. The increase of the mean concentration (by 0.01-0.04 mg/m<sup>3</sup>) in the daytime is explained by the fact that, as was shown by several authors [1, 2, 6, 9], under convective conditions the concentrations in the zone of influence of high sources increase, i. e., the daytime maximum in summer is explained by the influence of pollution from high sources.

In the daily variation of mean monthly concentrations of phenol and sulfur dioxide for the winter period (February), a decrease of the concentrations is observed from morning hours to daytime, and these concentrations reach their maximum values at 4-5 P.M., then decrease again by 9 P.M. The increase of concentrations before sunset is probably due to a decrease of turbulent mixing, and a further decrease occurs as a result of reduction in the discharge of pollution from minor sources into the atmosphere.

In summary, it may be stated that the daily variation of concentrations in the city substantially depends on the change of meteorological elements, which in turn depend on the time of day and the synoptic conditions. A drawback of the results obtained is the fact that no observational data are available for nighttime concentrations.

Fig. 2 shows the annual variation of mean monthly concentrations for different ingredients calculated from observational data at 12 points of sampling, during the period from September 1966 to September 1967 for two cities (a and b). The curves have a complex shape. However, there is a distinct minimum of all the ingredients during the autumn period and in December 1966. This is probably due to the self-purification of the atmosphere during the autumn period as a result of frequent precipitation, which washes the pollutants out of the atmosphere. Fig. 2 also gives a plot of the mean monthly wind velocity. The annual variation of the mean monthly wind velocity agrees relatively well with the concentration change. As the average wind velocity increases, the degree of air pollution decreases.

### 3. Single Episodic Survey of the Air Pollution of a City Reservoir.

In addition to stationary-type experimental studies, it is of interest to make an episodic survey of individual cities. The chief objective of such work was to determine the degree of pollution of the city's atmosphere for a relatively short period and to solve a number of problems pertaining to methods. One of such surveys was made in the summer of 1967 in a large industrial city in the south of the Ukraine. The start of the work was preceded by a study of the location of the sources of pollution relative to the residential areas of the city, which stretches out over a distance of tens of kilometers from the northeast to the southwest. A large number of major and minor sources were found to be located within the city outline and on its borders.

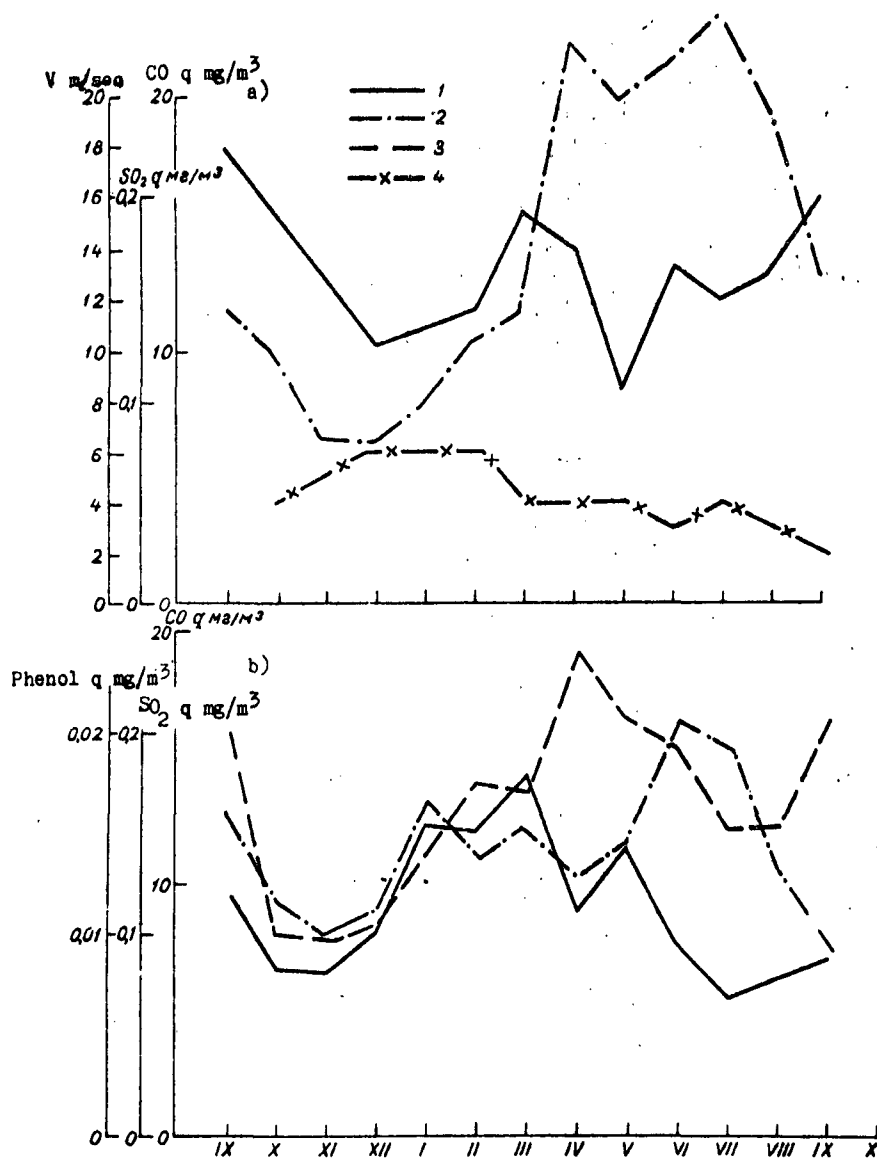


Fig. 2. Annual variation of impurity concentrations in two cities (a and b).  
1 - sulfur dioxide, 2 - carbon monoxide, 3 - phenol, 4 - mean monthly wind velocity.

A detailed familiarization with the parameters of discharges of the city's industrial enterprises showed that the greatest discharges occur at one of the industrial sites, containing a metallurgical plant with blast-furnace, open-hearth furnace and rolling sections, and coking, sintering, and cement plants. Southwest of this industrial site at distances of 4.5 and 8 km are located two ore-dressing complexes which also contribute substantially to the atmospheric pollution.

After the analysis of the arrangement of the sources and quantities of discharged pollutants, a program of experimental observations was worked out to determine the degree of pollution of the city's atmosphere. The program included a study of the background pollution of the atmosphere caused by the large number of sources scattered over the entire city, and also a study of the spreading of pollutants from the main sources located on the site, and measurement of meteorological quantities: air temperature and humidity at heights of 0.5 and 1.5 m, wind velocity at heights of 0.25, 0.5, 1.0, 2.0 and 4 m. Release of a surface A-58 radiosonde made it possible to obtain the distribution of the values of meteorological elements in height in the boundary layer.

A combined analysis of the annual wind rose for the given area and of the arrangement of the main sources shows that the latter are stretched out along the line of the direction of the prevailing wind, i.e., the individual plumes of the main sources are frequently superimposed. Maximum surface concentrations are observed in this case, since to the general pollution background of the city are added the discharges from main sources, whose common plume is directed toward the southeastern part of the city. As a result of a study of the distribution of the ground concentrations in the zone of influence of the plume, carried out in accordance with a technique described in [5], a concentration field was obtained at different distances from the source under certain meteorological conditions, and the zone of maximum and unsafe contaminations was determined.

During the period of the survey, samples for analysis of sulfur dioxide, carbon monoxide, nitrogen oxides and dust were taken at different distances from the source at a height of 1.5 m from the earth's surface. The number of samples taken according to the ingredients is shown in Table 1.

Table 1

Name of ingredient	Number of collected samples	
	Under plumes of sources	In city
Sulfur dioxide	301	330
Nitrogen oxides	307	343
Carbon monoxide	123	146
Dust	240	163

The distribution of maximum concentrations of sulfur dioxide, carbon monoxide, nitrogen oxides, and dust shows that the concentrations of the main ingredients in the zone of influence of the plume exceed severalfold maximum permissible norms within a radius of 15 km from the source. As is evident from Fig. 3, the curves have several maxima, i.e., the magnitude of impurities near the ground was determined by discharges of pollutants from a complex of enterprises located on a single industrial site (metallurgical

and coking plants, sintering plant), which gave the first maximum at a distance of 2-3 km. The second and third maxima were located at distances of 5-6 and 11-12 km, respectively, and are determined by discharges of ore-dressing complexes whose industrial sites are located at distances of 4.5 and 8 km from the metallurgical complex. The air pollution increases with the distance from the source, as it passes over the other sources. Even at a distance of 15 km from the first source, the concentrations are higher than the background in the city.

Fig. 3 shows the distribution of sulfur dioxide concentrations with the distance from the source for two gradations of wind velocity from 0 to 4 and over 4 m/sec. The wind was measured at a weather station at a height of 2 m from the earth's surface. Earlier it was shown [6] that the minimum sulfur dioxide concentrations in the zone of the plume of a single high source located outside the city limits were observed at wind velocities up to 2 m/sec, and the maximum ones, at velocities above 3 m/sec. As is evident from Fig. 3, the concentration field formed under the plumes of the sources in the city is inversely proportional to the wind velocity. The maximum sulfur dioxide concentrations near the ground are observed at wind velocities up to 4 m/sec. As the wind velocity increases, the surface concentrations decrease appreciably. Thus, in the distribution of impurity concentrations arriving from high sources, reinforcement of turbulent mixing takes place with increasing wind velocity in the city, resulting, on the one hand, in the elimination of stagnant zones, and on the other hand, in a more vigorous transport of impurities from the plume to the lower layer of the atmosphere. In this case, the first factor predominates over the second, and the atmosphere of the city becomes cleaner as a result of a more extensive aeration of the residential sections.

Having obtained the distribution of maximum concentrations from the main sources and for the mutual superposition of their plumes, one can evaluate the degree of the air pollution of other areas of the city when the plume is directed toward these areas. Of interest in this connection is the pollution background, produced by the action of minor pollution sources and automobile traffic.

In order to obtain the magnitude of background air pollution in the city, four main residential areas were selected, and in each area, four characteristic points for the given area. The sampling was made for sulfur dioxide, nitrogen oxides, carbon monoxide, and dust during morning, daytime, and evening hours. Results of treatment of the data are presented in Table 2.

The data of Table 2 show that the background pollution is relatively high. The city's atmosphere is heavily polluted with carbon monoxide and dust, and to a lesser degree with sulfur dioxide and nitrogen oxides. In the fourth area, concentrations above MPC were observed in all of the samples taken for analysis of carbon monoxide. This may be explained by the fact that, first, a heavy automobile traffic exists in this area, and second, that during the period of sampling, an air current was directed toward this district from the territory where a large number of mine pits are located.

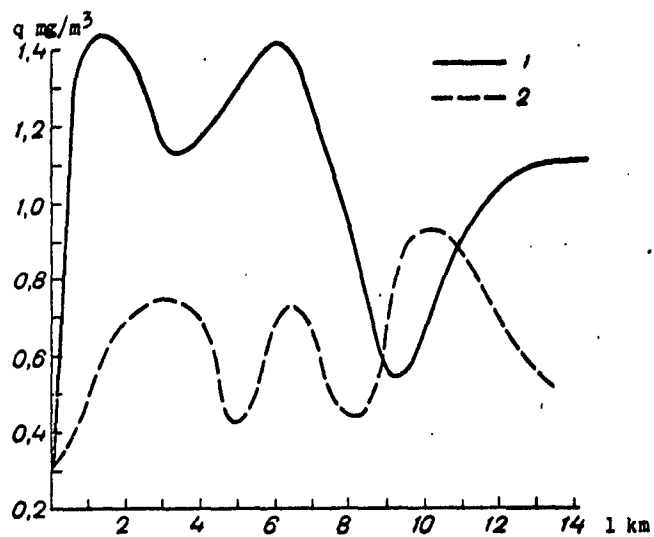


Fig. 3. Distribution of sulfur dioxide concentrations as a function of distances from sources at different wind velocities.  
1 - from 0 to 4 m/sec, 2 - above 4 m/sec.

Table 2

Concentrations, mg/m <sup>3</sup>									
No. of Area	No. of Point	Carbon Monoxide		Dust		Sulfur Dioxide		Nitrogen Oxides	
		Maximum	Mean	Maximum	Mean	Maximum	Mean	Maximum	Mean
1	1	66	27	2,38	0,64	1,1	0,3	1,1	0,27
	2	101	24	2,06	0,90	1,3	0,27	0,37	0,16
	3	72	19	1,17	0,61	0,75	0,25	0,4	0,14
	4	54	26	2,25	1,23	0,51	0,25	0,33	0,13
2	5	62	27	1,53	0,61	0,92	0,36	0,33	0,11
	6	89	26	1,06	0,63	0,95	0,32	0,34	0,09
	7	73	18	2,21	0,65	0,92	0,35	0,40	0,13
	8	73	23	1,31	0,64	0,85	0,26	0,45	0,10
3	9	54	23	1,71	0,68	0,69	0,22	0,09	0,05
	10	51	25	1,43	0,77	0,41	0,22	0,17	0,07
	11	25	26	1,7	0,51	0,39	0,25	0,27	0,1
	12	43	21	1,42	0,52	0,64	0,26	0,15	0,07
4	13	46	24	1,84	0,92	0,85	0,42	0,25	0,26
	14	41	22	1,14	0,69	0,84	0,54	0,71	0,16
	15	49	20	1,15	0,52	0,83	0,75	0,28	0,1
	16	36	18	1,76	0,96	0,85	0,35	0,30	0,1

From the standpoint of atmospheric pollution with dust, one can separate the first area, where maximum concentrations were observed (up to  $2.4 \text{ mg/m}^3$ ), and the frequency of concentrations above MPC was higher than in other areas. This is due to the fact that open-cut mines are located around the first area, and large areas are occupied by ash heaps.

During the survey, the program of sampling near the ground was supplemented with sampling from a helicopter at certain heights above the city and in the plumes of the main sources. Preliminary results of the treatment of these data are given in [4].

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ORGANIZATION OF EXPERIMENTS FOR STUDYING  
THE PROPAGATION OF NOXIOUS IMPURITIES FROM LARGE SOURCES

B. B. Goroshko

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p. 109-115, (1968).

1. Introduction

At the present time, considerable experience has been accumulated in organizing experimental studies of propagation of noxious impurities from large single sources as a function of the meteorological conditions. The data obtained are of major importance for checking the methods of calculation of dispersal of impurities in the atmosphere, for establishing the patterns of their propagation as a function of the meteorological conditions, for the further development of studies of propagation of impurities from platform sources, and for studying the problems of pollution of air reservoirs of heavily industrialized cities.

The object of the present paper is to generalize the experience gained from a broad range of expeditionary work in a region of large sources of atmospheric pollution, carried out in the past few years by the A. I. Voyeykov Main Geophysical Observatory with the participation of the F. F. Erisman Moscow Scientific Research Institute of Hygiene, the Southern Trust for the Organization and Efficiency of Electric Power Plants, and others. The general program of work includes meteorological ground observations, determination of parameters of the plume, selection of air samples under the plume of the source by analysis of ingredients that are discharged into the atmosphere, and determination of the characteristics of the discharge.

2. Meteorological Observations

In the atmosphere, an impurity is subjected to the influence of meteorological factors. A theoretical study [1-3, 13] and processing of the experimental material [10] have shown that the magnitude of surface concentrations strongly depends on the distribution of the meteorological elements in the boundary layer of the atmosphere. For this reason, during expeditionary work aimed at determining the concentration field near the ground in the zone of influence of the plume of a large source, it is necessary to organize a broad range of meteorological observations. The latter may be divided into two types: ground observations - up to a height of 15-17 m, and aerological observations - up to heights of 500-1000 m. The program of measurement of meteorological characteristics near the ground should include the determination of the wind velocity, temperature and air humidity up to a height of 17 m, and wind direction and balance observations [9]. The necessity of detailed measurement of the meteorological



elements in the ground layer is because of the fact that they are subjected to the greatest changes in both height and time. These data make it possible to obtain the values of the exchange coefficient near the ground and then, by using a series of models, to extend it to greater heights.

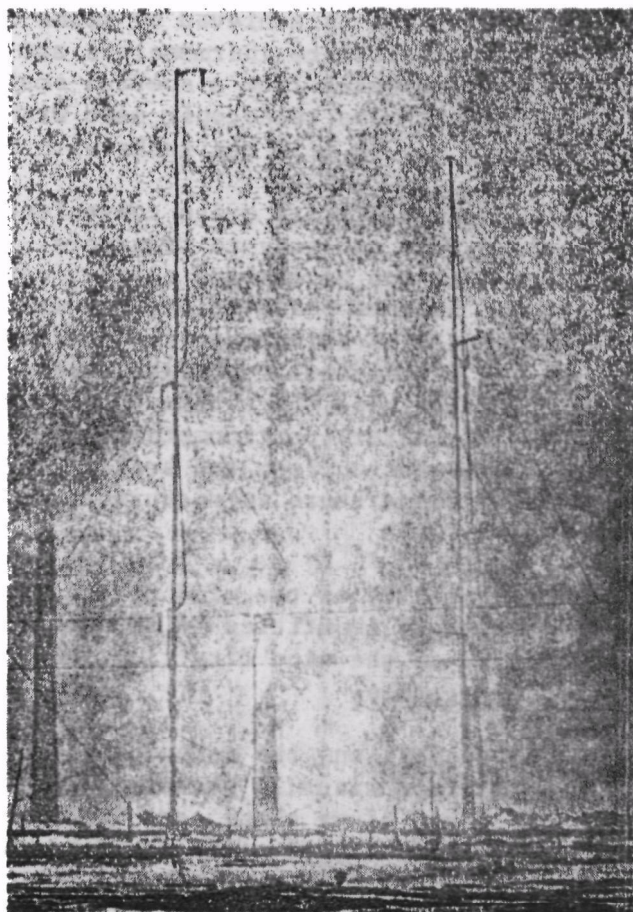


Fig. 1. Measurement of the profile of wind velocity and air temperature and humidity up to a height of 17 m on telescopic masts.

In order to obtain the wind profile in heights, the velocity is measured with contact anemometers, with recording by means of electromagnetic counters. The anemometers are placed at the following heights from the surface of the ground: 0.25, 0.5, 1.0, 2.0 and approximately 5, 10, 12 and 17 m. Starting at the 5 m level, the anemometers are mounted on brackets (80 cm long) of a telescopic mast (Fig. 1). The whole circuit is supplied with 6-8 volt dry batteries. This method of recording the wind velocity is one of the simplest and most reliable. It permits an automatic averaging of the wind velocity over the required time and the taking of remote measurements. Usually, the wind velocity is averaged over a 20-minute interval and this is followed by averaging over 1 hour.

The air temperature and humidity are determined at heights of 0.2, 0.5 and 2 m by means of Assman psychrometers, which are mounted in a horizontal position. At higher levels (5, 10 and 17 m), the temperature and humidity of the air are measured by means of resistance thermometers. The design of the thermometers used is such that a 0.1°C. temperature change in the ambient medium leads to a 0.1 ohm change in the resistance of the gauge, which is measured with a Wheatstone bridge. Radiation shielding of the thermometer is provided by placing the gauge in the housing of the Assman psychrometer, and ventilation is achieved by means of an electric motor. On a single level, two resistance thermometers are installed, one of which is wetted with distilled water from a special small tank welded to the frame of the psychrometer. The telescopic mast with the resistance thermometers is lowered once a day to fill the tanks with water.

In such studies, telescopic masts are quite convenient, since they are compact in the telescoped form and easy to transport. When the gauge goes out of adjustment, the mast can be lowered easily, permitting a rapid elimination of malfunctions. Balance observations are made according to a standard procedure: the radiation balance of the underlying surface, direct solar radiation on the perpendicular surface, reflected short wave, scattered and total radiation and also the albedo and effective ground radiation are measured. Balansometers, actinometers and albedometers are used in the measurements. Evaporation from the soil is measured by means of small evaporators, and the temperature is measured at depths of 2, 5, 15, and 20 cm.

According to [3], the stability of the wind direction has a considerable influence on the scattering of impurities in the atmosphere. For this reason, a continuous recording of the wind direction is made by means of M-12, M-45 or M-64 anemorhumbographs.

During the work of the expedition, the meteorological elements, including those which are determined visually such as cloud cover, visibility, atmospheric phenomena and plume shape, are under observation day and night: during the day for 1 hour, at night for 3 hours, and during the sampling, every hour.

As a rule, the discharge of noxious substances from large sources takes place at a great height (100 m and higher). Moreover, the considerable rate of ejection of gases from the stacks and (for hot sources) the large difference between the temperature of the gas and that of the surrounding air cause, under certain conditions, an effective ascent of the plume, whose height may reach 1000 m, and in some cases even higher, and it is only after this ascent that the noxious substances begin to descend and reach the ground. In such cases, a substantial layer of the atmosphere participates in the dispersal of the impurity, and the corresponding meteorological conditions in this layer substantially affect the magnitude of the ground concentrations. It is important, therefore, to know the magnitudes of the aerological characteristics. Their determination involves the use of methods of aerostatic and airplane-helicopter sounding of the atmosphere in the region of the source and reference pilot balloon observations of the wind velocity and direction. As a result, the distribution of tempera-

ture, humidity, wind velocity, structural characteristics of the wind, and a number of other characteristics from which the turbulence coefficient is calculated are determined. For atmospheric sounding up to a height of 500 m, a 125 m<sup>3</sup> MAZ-1 captive balloon is employed. To the cable of the aerostat is attached a mechanical aerostatic meteorograph that ascends every hour during the sampling period, measuring the pressure with an average error of  $\pm 5$  mb, the temperature with an average error of  $\pm 0.2^\circ$ , the humidity with an average error of  $\pm 1.4\%$ , the wind velocity with an error of  $\pm 0.5$  m/sec, and gustiness in the horizontal plane at levels of 25, 50, 100, 150, 200, 300, 400 and 500 m. Between the ascents of the main meteorograph, an instrument for measuring the average velocity and for determining the structural characteristics of the wind ascends with the platform to levels of 100, 200 and 300 m. For sounding the atmosphere to a height of 1000 m, use is made of a helicopter (Mi-1) or airplane (YaK-12, AN-2), which is equipped with an electrometeorograph with gauges for pressure, humidity, temperature, fluctuations of temperature and overloads, and also, for duplication purposes, a mechanical meteorograph with pressure, temperature, and humidity gauges. A detailed description of the instruments and technique of aerological observations is given in [5-7].

In order to consider the dispersal of impurities, it is very important to know the shape of the plume and its dimensions, which are determined by means of a helicopter or airplane flying through the plume at a known velocity, at various distances from the source, the flight time being recorded with a stopwatch. The distance of the visible zone of propagation of the plume from the source is determined in the same manner. Photographing of the plume from a point located at a distance of 2-3 km from the plume along a straight line perpendicular to the latter yields additional data on the parameters of the plume, whose dimensions and shape depend on the distribution of the values of the meteorological elements in height. Photographs are taken every 15 min and also in cases where the shape of the plume changes sharply. The plume parameters thus obtained make it possible to calculate, by means of a procedure worked out by M. Ye. Berlyand [4], the coefficient of turbulent exchange [11] and the effective height of the plume [8], making use of the fact that the scale of the stacks, that are always found on the photographs, is known.

### 3. Collection of Samples in the Zone of Propagation of the Plume of a Large Source

The determination of ground concentrations at man's breathing level (1.5 m) in the zone of influence of the plume of the enterprise being studied is made in two steps: collection of samples and their analysis in the chemical laboratory by existing techniques. In cases where small volumes can be used, the sampling is carried out in small containers (flasks, gas pipets, etc.). As a rule, substances in the gaseous and vapor phase are collected in liquid media, where they are dissolved or combined chemically, i.e., suction methods of collection are chiefly used, based on the drawing of a known volume of gas through an absorbing medium.

When thermal power stations operate in the zone of the plume, where the main noxious ingredients are sulfur dioxide and dust, the sampling is carried out by means of v-shaped absorbers with porous plate No. 1 (for taking samples to be analyzed for sulfur dioxide) and filters of FPP-15 fabric (for taking samples to be analyzed for dust). For drawing air through absorbers or filters, depending on the volume of collected air required, use is made of suction tanks, OK-1 aspirators, automobile aspirators, etc. according to the techniques of [14].

The sampling is carried out under a plume in accordance with a predetermined program and simultaneously with an extensive group of meteorological measurements and with the determination of the characteristics of the discharges. The samples are taken simultaneously under the plume at approximately 20 points, at three to five and sometimes more distances (Fig. 2). At each distance, from two to five collecting points are established perpendicular to the plume at a distance of 50-400 m from each other depending on the width of the plume. Gradually the distances change, beginning with 500 m from the source, and on up to 15-20 km. The sampling lasts 20 min. The spacing of the points is carried out under the plume, and is arranged according to its physical position. The distance from the source is determined by comparing the location of the sampling point with a map of the area, or by means of telemeters (clinometer, binoculars) or from readings of an automobile speedometer when there is a straight road along the direction of the plume. When there is no visible plume, the spacing of the sampling points is made by considering the odor of a specific ingredient discharged by the source and by using the direction of visible plumes of the nearest sources, with ejection at the same height.

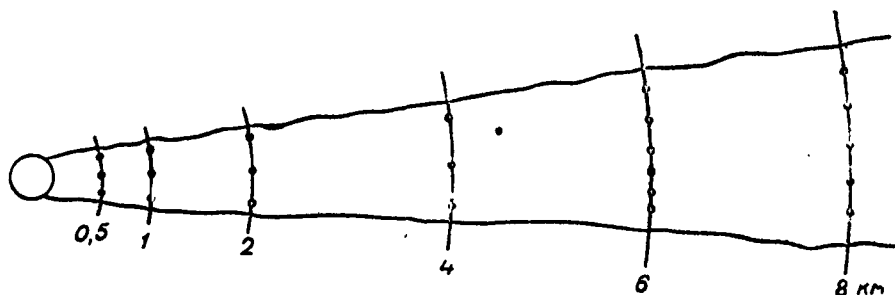


Fig. 2. Schematic diagram of the spacing of air sampling points under the plume of a source.

In order to elucidate the influence of the daily variation of meteorological elements on the dispersal of impurities, the sampling is set up at different times of day: in the morning, in the presence of slight convective exchange; during the day, during the period of maximum turbulent exchange; and in the evening, when the turbulent exchange has been attenuated. The influence of seasonal changes of meteorological elements is brought out by organizing the studies in various seasons of the year, and the climatic changes are brought out in various climatic zones.

#### 4. Determination of the Characteristics of a Discharge

In [3] and [14], the magnitude of ground concentrations is expressed by the following formula:

$$c_m = \frac{AMFm}{H^2 \sqrt[3]{V\Delta T}},$$

where A is a coefficient dependent on the temperature stratification of the atmosphere, which determines the conditions of the vertical and horizontal dispersal of the impurity in air, M is the amount of noxious substances discharged into the atmosphere, H is the stack height, V is the volume of the gas-air mixture discharged,  $\Delta T$  is the difference between the temperature of the escaping gases and the air temperature, F is a dimensionless coefficient allowing for the rate of deposition of the noxious substances, and m is a dimensionless coefficient allowing for the conditions of ejection of the gas-air mixture from the mouth of the discharging source.

It is apparent from the formula that the magnitude of the surface concentration observed in the zone of the plume of the source is affected by several variable quantities that change with the conditions of operation of the plant. They are determined directly at the plant under study.

To determine the total volume of escaping gases, measurements are made in gas conduits behind the filters, the gas conduit having first been calibrated. A Prandtl tube is used to determine the dynamic head, and the mean velocity is calculated, by means of the formula

$$W_{av} = 4.37 k_c \frac{\sqrt{h_g}}{\gamma_g} \text{ m/sec.}$$

where  $k_c$  is the calibrated coefficient,  $h_g$  is the dynamic head (mm H<sub>2</sub>O) and  $\gamma_g$  is the specific weight of the flue gases under the operating conditions (kg/m<sup>3</sup>).

Knowing  $W_{av}$ , one can calculate the total volume of escaping gases

$$Q = 3600FW_{av} \text{ m}^3/\text{hr}$$

where F is the cross-sectional area of the gas conduit (m<sup>2</sup>).

The determination of the amount of noxious substances discharged into the atmosphere is made behind the filters. For example, dust samples are taken by means of a special dust-collecting probe, and the total volume of the dust discharged is calculated from the formula

$$G = \frac{q_0 60 \cdot AF}{1000z \cdot \pi D^2} \text{ kg/hr}$$

where  $q_0$  is the weight of ash trapped by the dust-collecting probe (g), F is the cross-sectional area of the gas conduit (m<sup>2</sup>), Z is the duration of the sampling in minutes, and D is the mouth diameter of the dust-collecting probe (m).

The collected dust samples are analyzed for their fractional composition by the air classification method.

The temperature of the escaping gases is measured in the gas conduits behind the filters.

In order to determine how closely the actual situation is reflected by the samples of gases collected behind the filters in the gas conduits, the data obtained are checked by collecting samples of fuel and analyzing it chemically for the content of ash and sulfur, considering the amount of fuel burned; then taking the efficiency of the purifying equipment into account, the amount of noxious substances discharged into the atmosphere per unit time is calculated. Thus, for example, the amount of sulfur dioxide discharged is calculated from the formula

$$M_{\text{SO}_2} = \frac{B \cdot 10^6}{3600} \frac{SP \cdot \mu_{\text{SO}_2}}{100 \cdot \mu_S},$$

where  $M_{\text{SO}_2}$  is the amount of sulfur dioxide discharged from the stack (g/sec),  $B$  is the consumption of fuel (t/hr),  $SP$  is the sulfur content of the fuel (%),  $\mu_{\text{SO}_2}$  is the molecular weight of sulfur dioxide, and  $\mu_S$  is the molecular weight of sulfur.

Carrying out the appropriate calculations, one can reduce the formula to the form

$$M_{\text{SO}_2} = 5,56 BS^p.$$

The amount of ash discharged is determined from the formula

$$M_{\text{ash}} = \frac{B \cdot 10^6}{3600} \left(1 - \frac{\eta_{\text{ash}}}{100}\right) \left[\left(1 - \frac{q_n}{100}\right) \frac{AP}{100} a_{\text{esc}} + \frac{q_n}{100}\right],$$

where  $M_{\text{ash}}$  is the amount of ash discharged (g/sec.),  $B$  is the fuel consumption (t/hr),  $\eta_{\text{ash}}$  is the efficiency of the purifying equipment,  $q_n$  is the heat loss due to the mechanical incompleteness of combustion,  $AP$  is the ash content of the fuel (%), and  $a_{\text{esc}}$  is the fraction of fuel ash which escapes with the gases into the gas conduits.

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## DETERMINATION OF THE AIR POLLUTION POTENTIAL

E. Yu. Bezuglaya

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### 1. State of the Problem

The level of air pollution is determined by discharges from various sources and by the nature of their dispersal in the atmosphere. To evaluate possible air pollution for given discharge parameters, the concept of "air pollution potential", referring to the ability of the atmosphere to scatter impurities, is used in many countries. It is a function of the meteorological and topographic parameters and thus, for a given locality or district, may be evaluated from data on the relief and on the expected meteorological conditions.

The air pollution potential from a meteorological standpoint may be defined as a combination of weather factors determining the level of possible air pollution. Its changes take place under the influence of turbulent motions, particularly changes in the vertical thermal stability, velocity and direction of the wind, and a whole series of other meteorological parameters.

A frequent combination of meteorological conditions unfavorable to the dispersal of impurities indicates a high air pollution potential in the given region. A low air pollution potential takes place in regions where meteorological conditions favorable to the dispersal of discharges are frequent.

Thus, the degree of air pollution for the same amount of discharges may differ appreciably depending on the climatic characteristics of the various geographical regions. An evaluation of the air pollution potential based on climatic and aeroclimatic data is an important characteristic that must be considered in planning urban industrial construction and development.

Systematic studies of the climatology of the air pollution potential and its prediction are being carried out abroad. The first part of the present paper will consider certain conclusions drawn from this work, and then results of an analysis of the air pollution potential on the territory of the USSR will be given.

American researchers assume that weak winds and a stable state of the atmosphere may serve as the basis for predicting unsafe pollution conditions. They have worked out an experimental program for determining the air pollution potential for forecasting purposes. Prediction based on this program was conducted during the period 1957-1959. By a high



air pollution potential was meant a combination of the following conditions:

- 1) velocity of ground winds, less than 4 m/sec;
- 2) velocity of winds at the level of the 500 mb surface, less than 12 m/sec;
- 3) descending motions of air, below the 600 mb surface;
- 4) duration of such conditions, about 36 hours.

Six periods of high potential during the indicated two years were accompanied by a high level of air pollution in many U.S. cities simultaneously [8].

A recent study of meteorological conditions promoting a high air pollution has established that they are observed in quasi-stationary anticyclones [10].

Since 1960, predictions of the air pollution potential over the entire territory of the U.S.A. have been made in Cincinnati. In the last few years, several different forecasting methods have been developed. In some cases, the air pollution potential is determined from data on the height of the base of revised temperature inversions and the average wind speed in the layer from the ground to the base of the inversions [15]. Data on temperature inversions may serve as a good indicator of the limitation of dispersal of impurities in height, but, in the absence of these data, another criterion is necessary, which could be used to evaluate the vertical thickness of the layer where mixing of the discharges takes place. Holzworth [11] has introduced the concept of "maximum mixing height", which enables one to evaluate the thickness of the layer with the greatest vertical dispersal in the course of 24 hours. The maximum mixing height is assumed to depend solely on the surface temperature maximum for 24 hours and on the vertical thermal stratification of the layer. Its value characterizes the thickness of the convection layer in which vertical mixing of the impurity takes place. It is determined from data of radio sounding of the atmosphere as the level of intersection of a dry adiabat drawn from the temperature maximum on the ground, with a vertical temperature profile.

In the western part of the U.S.A., air stagnation has been forecast as follows. Analysis of synoptic conditions showed regions where a high pressure area was located near the ground, and a warm nucleus or crest was located at the heights, indicating that the surface pressure system would move relatively slowly. Further, for the next 24 hours, the conditions of vertical thermal stability of the atmosphere and horizontal transport in two layers of the troposphere were calculated. The first layer is located below 1000 m, and the second, between 1000 and 6000 m. The product of afternoon mixing height times the mean velocity of the wind from the surface of the ground to a height of 1000 m is found for the first layer. For the second layer, radio sounding results were used

to find the thickness of the layer (located above 1000 m) in which the potential temperature changed by  $5^{\circ}$ , then the product of this thickness times the mean velocity in the layer from 1 to 6 km was obtained. The sum of the products of the first and second layer was taken as the air pollution potential. The experiment performed established that if this sum was equal to 20 or less, a high level of air pollution could be expected. Thus, by using data of aerological measurements, the air pollution potential was calculated, and regions with its unsafe values were identified [17].

Forecasts of high pollution potential are also prepared from data on the maximum mixing height and wind velocity [18]. Recently, instead of the maximum mixing height, which is difficult to predict, use has been made of parameters that are more convenient for forecasting and are related to the thickness of the mixing layer, for example, the difference of the ground maximum of potential temperature and the potential temperature at the isobaric surface of 850 mb. To refine these values, the temperature at the 500 mb. isobaric surface is employed. The forecast of the maximum mixing height is made on the basis of data on numerical forecasting of the temperature maximum on the ground and on isobaric surfaces of 850 and 500 mb. Attempts have been made to calculate the impurity concentration in the city from the total amount of discharges and from data on the maximum mixing depth and wind velocity [19].

For a climatic evaluation of the air pollution potential, Hosler [13] studied the propagation of temperature inversions in the lower 500 meter layer of the atmosphere above the U.S.A. He found that a limitation of vertical mixing is observed in almost 25% of the observations in all seasons. In winter, ground and raised inversions in the east and in the continental part of the U.S.A. are observed in 50%, and in the west above mountains, in 90% of all cases. He showed that at night, as a rule, a stable vertical stratification temperature and a minimum vertical mixing above the entire territory of the U.S.A. are observed.

To evaluate the unsafe periods of air pollution during the daytime, Holzworth [11] calculated the average values of the maximum mixing height according to seasons, using for this purpose the data of 45 radio sonde stations of the U.S.A. for 10 years and maximum temperature data for 30 years.

Maps of distribution of this quantity over North America were plotted. The maximum mixing height increases in summer and decreases in winter to 200-800 m. In the coastal areas of the Pacific and Atlantic Oceans, it changes only slightly in the course of the year.

An important element for the climatic evaluation of the air pollution potential is the wind velocity at the surface of the ground. In [12], the frequency of weak winds was studied in an annual cycle. In that paper, weak winds include those having velocities up to 2 m/sec. Three groups were distinguished according to the character of the annual variation of the wind velocity at 48 stations: 1) weak winds are absent in all seasons; 2) there

is a large number of weak winds in winter and a small number in summer;  
3) the largest quantity of weak winds occurs in autumn.

A map of the geographical distribution of different types of annual variation of the velocity was plotted, and regions with periods having weak winds lasting from 10 to 20 days were identified.

Synoptic conditions associated with periods of long-time high air pollution potential were studied, and it was found that stationary anticyclones are located on the surface map and a warm crest is located at the heights.

M. E. Miller and L. E. Niemeyer calculated the number of cases per year in which a high air pollution potential can be expected, and plotted the corresponding map. In most cases, there was an actual rise of the air pollution level [16].

Korshover [14] studied the frequency of stationary anticyclonic centers above the eastern part of the U.S.A.

In the last few years, more extensive climatic generalizations have been made for determining the air pollution potential of separate regions in which the construction of major air pollution sources is contemplated. For example, an atlas of a series of climatological characteristics such as the wind direction, wind velocity, frequency of inversions, precipitation, precipitation with winds, and the like was compiled for the region of Southern California, where the construction of an atomic power station was proposed [9].

## 2. Possibilities of Evaluating the Air Pollution Potential above the Territory of the Soviet Union

In order to identify the regions where conditions favoring the accumulation of impurities in air arise most frequently, it is of interest to make a climatological evaluation of the air pollution potential above the territory of the Soviet Union.

To select the meteorological parameters that must be considered in such an evaluation, use is made of conclusions obtained in a series of papers, and additional treatment is presented involving the study of the influence of temperature inversions of the lower layer of the atmosphere and various wind velocities on the conditions of increase of the impurity concentrations in cities.

Theoretical and experimental studies show that raised inversions stop impurities coming from pollution sources and create conditions favorable to the accumulation of high impurity concentrations in air [1].

The influence of ground inversions on the air pollution of cities has been studied less extensively. Theoretical estimates indicate that highly

developed ground inversions present a danger in cities containing many low sources of air pollution.

When high sources of air pollution are present in a city, considerable impurity concentrations near the ground are observed in the presence of the so-called "unsafe velocity" of the wind  $u_m$ , which for high-capacity thermal power stations is about 5 m/sec [2]. The heaviest air pollution from low sources of discharges in a city is observed at a wind velocity of 0-1 m/sec [5].

As the wind is weakened in the ground layer, unsafe air pollution conditions may also arise if an inversion layer is located above the stack of a high pollution source [1].

In order to study the influence of ground inversions at different wind velocities in the boundary layer of the atmosphere on the air pollution level of a city, use was made of data on the concentrations of dust, soot, nitrogen oxides, carbon monoxide and sulfur dioxide in several cities indicated in Table 1.

City	Period of Observations	Number of Observations
Alma-Ata	1966	888
Dushanbe	1966	1014
Tallinn	VI 1965 - XII 1966	638
Tbilisi	1962 - 1963	2385
Moscow	1966	1000

Unfortunately, the quantity of observations of the air pollution level in cities is still insufficient to permit a reliable statistical treatment in order to obtain quantitative relationships between the impurity concentrations and the meteorological characteristics. It becomes necessary to restrict oneself mainly to the identification of qualitative relationships for the purpose of using them later in the forecasting and climatology of air pollution.

Mean impurity concentrations were calculated for different conditions of thermal stratification of the atmosphere (Table 2).

As is evident from Table 2, the concentrations of all the impurities in the presence of ground inversions are higher than in their absence.

In the cities of Irkutsk and Tbilisi, where the quantity of observations of air pollution in the presence of ground inversions was found to be sufficient, mean values of the concentrations of soot and dust were calculated for each month.

Table 2

Mean Impurity Concentrations ( $\text{mg}/\text{m}^3$ ) in Various Cities.

City	Type of Atmospheric Stratification		
	Ground Inversion	Inversions With Base Above 0.01 km	Absence of Inversions
Soot			
Tbilisi . . .	0,17	0,16	0,09
Dust			
Tallinn . . .	0,37	0,34	0,27
Moscow . . .	0,30	—	0,21
Tbilisi . . .	0,54	0,57	0,41
Sulfur dioxide			
Dushanbe . . .	0,12	0,10	0,11
Moscow . . .	0,33	0,40	0,32
Alma-Ata . . .	0,17	0,16	0,12
Nitrogen oxides			
Dushanbe . . .	0,38	0,36	0,26
Alma-Ata . . .	0,61	0,50	0,47
Carbon monoxide			
Tallinn . . .	3,0	2,9	1,9

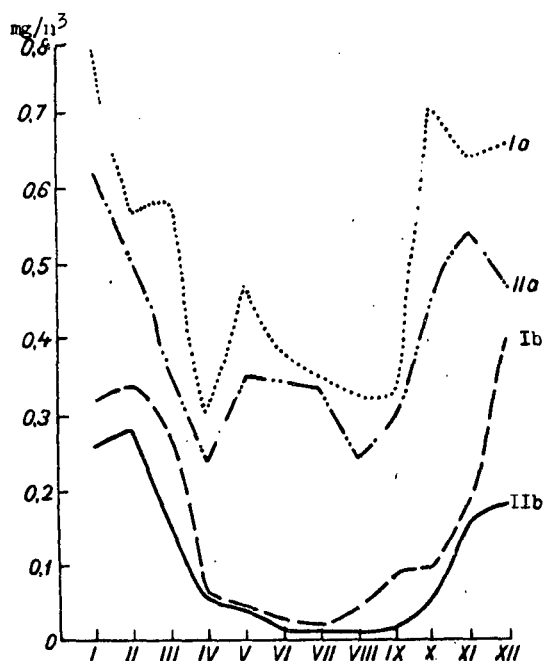


Fig. 1. Mean concentrations of dust (a) and soot (b) in Tbilisi in the presence (I) and absence (II) of ground inversion.

Fig. 1 shows the annual variation of dust and soot concentrations in the city of Tbilisi in the presence (I) and absence (II) of ground inversions. The highest concentrations of the impurity are observed in the presence of stable stratification of the ground layer in all months. Similar results were obtained in Irkutsk.

A comparison of the data on the concentrations of sulfur dioxide and nitrogen oxides obtained by the Alma-Ata and Moscow stations under different stratification conditions also shows a definite increase of pollution during the warm and cold halves of the year in the presence of ground inversions.

In addition, the influence of inversions formed above the earth's surface on the pollution level in a city was also analyzed. Mean impurity concentrations in the cities of Tbilisi, Moscow and Alma-Ata were calculated separately for inversions with a base below and above 500 m. The

results obtained show that there are no clearly defined differences in the pollution level of the two groups of inversions. This is obviously determined by the variety of the sources of discharges in the city. High sources in the presence of low-raised inversions turn out to be higher than the inversion, and their discharges do not reach the ground.

As is evident from Table 2, the concentrations of various impurities under conditions of raised inversions are always higher than in their absence, but usually slightly lower than under conditions of ground inversions.

Thus, it may be definitely concluded that the presence of an intercepting layer in large cities containing many low sources of discharges promotes the accumulation of noxious impurities in the air.

For the cities of Moscow and Alma-Ata, mean concentrations of sulfur dioxide and nitrogen oxides were calculated for the cold half of the year at various wind velocities (Fig. 2) without considering the stratification. As is evident from Fig. 2, in winter the highest concentrations of these impurities are observed in the presence of calm. As the wind grows stronger, the impurity concentrations decrease rather sharply. According to observations in Moscow, there is also observed a new increase of the concentration at a wind velocity of 6 m/sec, probably due to the presence of high sources of discharges.

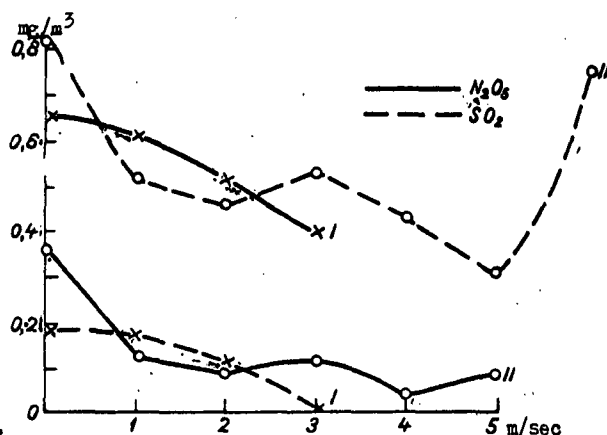


Fig. 2. Mean concentrations of sulfur dioxide and nitrogen oxides at different wind velocities based on Alma-Ata (I) and Moscow (II).

The most extensive observations of dust and carbon monoxide concentrations were obtained for the city of Tallinn. They permitted a calculation of mean values of these impurities for various gradations of the wind velocity near the ground, for three types of thermal stratification of the atmosphere (Tables 3 and 4).

The data of Table 3 show that the greatest air pollution with carbon monoxide was observed in ground inversions in the presence of a wind of 1 m/sec. High concentrations of this impurity are also noted at wind velocities of 5 m/sec. For inversions with a base below 500 m, maximum

pollutions are observed, as in the case of ground inversions, at wind velocities of 5 m/sec. This may be explained by the fact that the CO discharges originate from low sources, chiefly from automobile transportation and stacks 10-30 m high, for which the "unsafe" wind velocity is about 1 m/sec. The second maximum is due to high sources, for which the unsafe wind velocity is close to 5 m/sec.

Table 3

Mean Concentrations of Carbon Monoxide in Inversions  
at Different Wind Velocities in Tallinn

Type of atmospheric stratification	Wind velocity, m/sec										
	0	1	2	3	4	5	6	7	8	9	≥10
Ground Inversions	1,8	5,9	1,9	2,4	2,3	3,6	3,1	2,8		—	2,3
Inversions with base at 0.01- 0.5 km	1,8	2,9	2,2	2,3	2,4	4,3	2,1	2,7	2,9	—	1,7
Without inver- sions	—	1,7	2,8	1,5	2,8	3,1	1,1	1,8	1,8	—	3,3

Table 4

Mean Concentrations of Dust in Summer in Tallinn

Type of Stratification	Wind velocity, m/sec			
	0-1	2-4	5-7	≥ 8
Ground Inversions	0,51	0,28	0,46	—
Raised Inversions	0,29	0,33	0,32	0,50
Without Inversions	0,32	0,30	0,23	0,28

The data listed in Table 4 enable one to reach a number of conclusions concerning the influence of inversions in combination with various wind velocities on the concentration of dust in city air.

The greatest air pollution with dust is observed in ground inversions at wind velocities no higher than 1 m/sec. A rise of the air pollution level in ground inversions is caused by a weak vertical Austausch, and the simultaneous presence of calm causes the absence of horizontal transport. Heavy air pollution with dust was observed in the presence of ground inversions at wind velocities of 5-7 m/sec, and in the case of raised inversions, at wind velocities above 8 m/sec, which is obviously due to the "smoke pollution" effect, i.e., the descent of the plume from smokestacks toward the earth's surface. Finally, in the absence of inversions, the greatest air pollution with dust occurs at wind velocities of 0-1 m/sec and decreases with increasing wind velocity.

The conclusions reached show that for large cities, the conditions of formation of thick ground inversions at wind velocities of no more than 1 m/sec are very unsafe. These conclusions agree with those obtained earlier on the accumulation of impurities in cities in the cold half of the year in the presence of stationary anticyclones [6], which are characterized by a high frequency of ground inversions [4] and weak winds. To confirm the above, the three highest values of the concentrations of each impurity for each point in a month were selected from observational data in Leningrad on the concentrations of sulfur dioxide, carbon monoxide and nitrogen oxides during May-September 1967 (a total of 3315 observations). In all, 223 observations were selected. Analysis of these data for all the impurities shows that 74% of the latter were detected when in the 12 hours preceding the collection of air samples the wind velocity did not exceed 1 m/sec.

The considerable influence of a long duration of a wind velocity of 0-1 m/sec and thick ground inversions (air stagnation) on the increase of the air pollution level to unsafe values indicates the necessity of considering the frequency of such conditions in cities. To this end, we determined the frequency of days when a wind velocity no higher than 1 m/sec was observed in the course of 24 hours.

Using data of daily meteorological observations of the wind velocity based on a weather vane, days were selected at 220 stations when wind velocities of 0-1 m/sec were detected at a given point in the course of all the observation periods. Using the results of selections for January, April, July and October 1962-1966, the author in cooperation with L. I. Yelikoyeva plotted maps of frequency of air stagnation conditions (wind velocities of 0-1 m/sec in the course of 24 hours). It was found that in each case, the air stagnation was observed at several stations simultaneously. Analysis of synoptic maps showed that the observed cases of weak winds were related to large-scale synoptic processes. In winter, they were chiefly observed in the central portion of slowly moving anticyclones or in gradientless fields of diffuse anticyclone or pressure col type.

The maximum number of cases of air stagnation over the territory of Eastern Siberia, the Far East, Trans-Caucasia and mountainous regions of Central Asia in the annual cycle is observed in winter. In the eastern regions of Western Siberia and in the Ural region, in addition to a winter maximum, an increase in the number of cases of air stagnation is also observed in July. Above the European territory of the USSR, the frequency of the number of air stagnations changes insignificantly in different seasons.

Data on the average number of days with wind velocities of 0-1 m/sec in the course of 24 hours (air stagnation) for January, April, July, and October were plotted on maps. They must of course be considered as preliminary and in need of further refinement. This is because only a five-year period of observations and a somewhat limited number of stations were used.



To determine the degree of openness of the location of a station, use was made of an evaluation based on V. Yu. Milevskiy's classification. Stations of the 5th. category of openness and below were excluded from consideration. Analysis of the maps showed that it was possible to distinguish zones where the number of days per month with wind velocities of 0-1 m/sec in the course of 24 hours (air stagnation) was: 1) less than 1; 2) from 1 to 5; 3) above 5.

In all the selected months in the west of the European territory of the Soviet Union including the western part of the Ukraine, the inner areas of the Baltic region, and Belorussia, air stagnation was observed for 1 to 5 days a month. The same frequency of the number of days with air stagnation conditions was noted at a number of points on the western and eastern slopes of the Urals. In the forest zone of Western Siberia, air stagnation above 1 day per month are observed in January, and in other months, once every few years.

On the shores of the border seas of the Soviet Union, in the steppe and forest-steppe zone of the European territory, and in northern Kazakhstan, cases of air stagnation are very rare. Their frequency in all the selected months is less than 1 day, i.e., air stagnation may be observed once every 3-4 years or less often.

The mountainous regions of Trans-Caucasia and Central Asia are marked by an extremely complex map of frequency of air stagnation cases. Here we should note only a high frequency of such cases in certain cities (Yerevan - up to 18, Fergana - 17, Alma-Ata - 13, Khorog - 18 days per month).

In winter, Eastern Siberia and regions of the Far East, excluding the sea shores, are in the area of a heavy anticyclone causing weak winds and cases of air stagnation. In January, the number of days of air stagnation in this part of the territory of the Soviet Union exceeds 5, and in some areas (Bodaybo, Chul'man, Blagoveshchensk, Nizhneudinsk, Oymyakon, Olenek, Verkhoyansk, Ekimchan) reaches 20-25 a month. In other middle months of the seasons, the frequency of air stagnation conditions is also considerably higher than in other regions, but less than in January (reaching 10-13 days a month).

Considering the frequencies of calms, and also data on the frequency of winds of 0-4 m/sec at a height of 500 m, we attempted to make a rough estimate of the possibility of appearance of unsafe air pollution levels in various areas of the Soviet Union. The distribution of the frequency and the thickness of ground inversions were taken into consideration [3]. As a result, four areas were distinguished (Fig. 3):

- 1) A large area of Eastern Siberia, where the greatest frequency of stagnation conditions is observed. In winter, up to 25 days per month with air stagnation conditions are observed in some parts of this region;

- 2) Western regions of the European territory of the Soviet Union, western and eastern foothills of the Urals, where a moderate frequency of

air stagnation is observed;

3) The northwestern part of the European territory of the Soviet Union and forested regions of Western Siberia, where air stagnation conditions are observed only in spring and winter;

4) Kazakhstan, land along the Volga, northern part of Central Asia, a large part of Western Siberia and the shores of border seas, where air stagnation conditions virtually are not observed (once or twice every five years).

The Ural, southern regions of Central Asia, Caucasus, and mountainous regions of Eastern Siberia are not included in the zoning.

The data cited give a certain measure of the distribution of the air pollution potential. A more complete evaluation of the latter requires extensive studies of the frequency of the vertical stability of the lower part of the troposphere and distribution of weak winds in height in the boundary layer of the atmosphere. In order to allow for the capacity of the atmosphere to accumulate impurities, data on fogs should obviously be employed. Also useful will be data on the duration and magnitude of atmospheric precipitation, probability of clear and cloudy sky, coefficient of turbulent exchange, amount of incoming radiation, frequency of stationary anticyclones, and the like.

In addition, since sources of different heights are affected by different meteorological parameters, the air pollution potential should be evaluated separately for high and low sources of discharges.

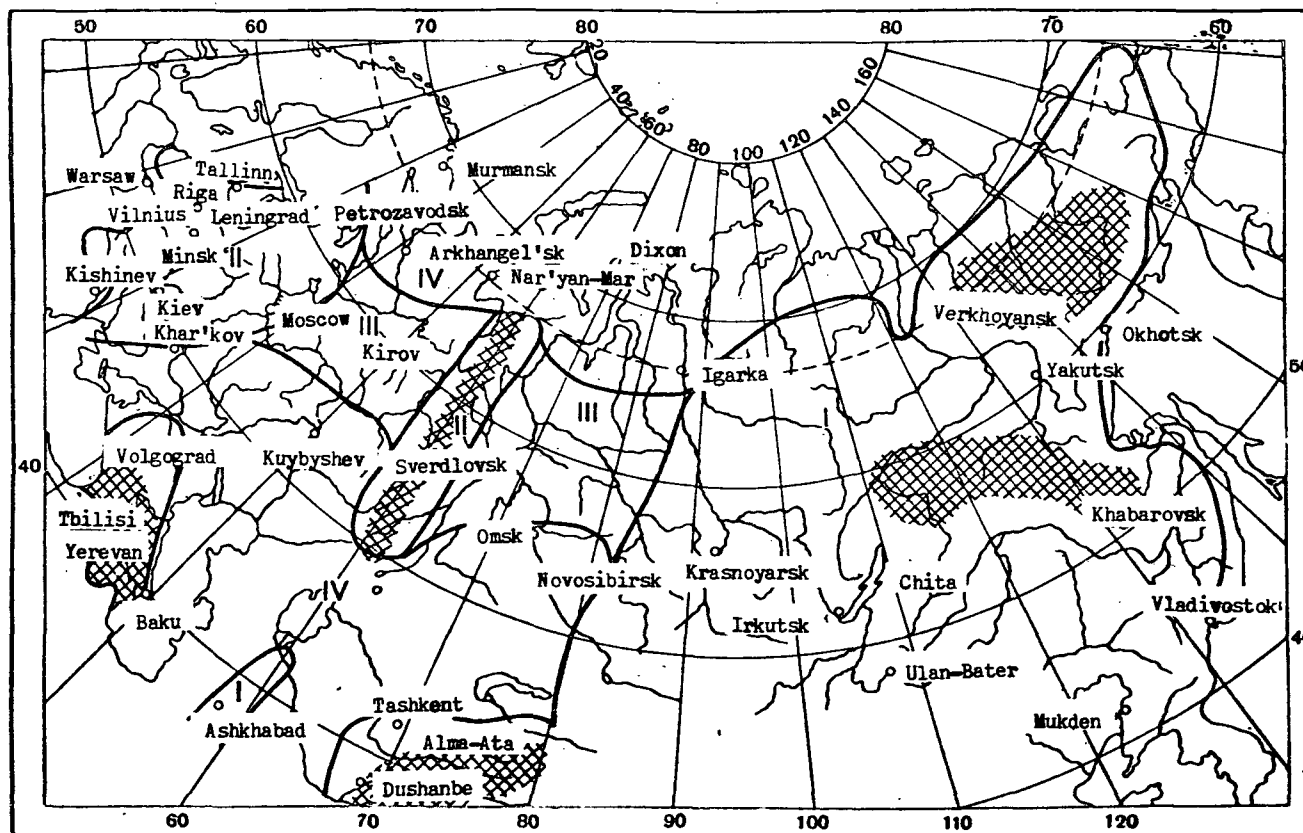


Fig. 3. Map-diagram of the territory of the USSR. Areas of different numbers of days with wind velocities of 0-1 m/sec in the course of 24 hours (shaded areas are those not covered by the zoning).

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# SOME GENERALIZED CONCLUSIONS CONCERNING THE EXPERIENCE OF OBSERVATION POSTS IN REFERENCE TO THE CHEMICAL COMPOSITION OF THE ATMOSPHERE OF CITIES

I. A. Yankovskiy

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## 1. Introduction

The system of the hydrometeorological service (HMS) has now initiated work on a systematic control of the chemical composition of atmospheric air in heavily industrialized cities by organizing stationary observations and expeditionary surveys with simultaneous observations of meteorological conditions.

The organization of stationary posts and arrangement of systematic observations were preceded by major preparatory work involving the collection of information on the main sources of pollution and characteristics of parameters of industrial discharges, selection of areas for taking the air samples, their coordination with sanitary epidemiological stations and urban architects, the outfitting of posts with special booths (kiosk type), compilation of sanitary topographical descriptions of the location of the posts, etc. As early as 1965, observations of the content of sulfur dioxide and nitrogen oxides in the atmosphere were organized at meteorological stations in many cities.

For purposes of systematic control of air pollution, an additional network of stationary observations of the atmospheric content of sulfur dioxide, dust, carbon monoxide, nitrogen dioxide and hydrogen sulfide was created in 1966-1967 in 60 major industrial cities. In each city, from three to six posts are set up with double or triple observations of concentrations of ingredients most characteristic for a given city, and associated meteorological conditions.

In the present paper, on the basis of inspection trips and studies of observational material received at the A. I. Voyeykov Main Geophysical Observatory, an attempt was made to draw some generalized conclusion concerning the experience of local subdivisions of the HMS in studying the pollution of air reservoirs of cities, primary attention being given to problems of methods and organization of the work of the posts.

## 2. Choice of Areas for Stationary Posts

In organizing work aimed at studying atmospheric pollution, primary attention should be focused on the problem of selection of locations for stationary observation posts, since a suitable selection of such posts permits an evaluation of the degree of pollution of city air even

on the basis of a few points. For this reason, the selection of such locations is based on a detailed familiarization with the layout of the city, the location on its territory of industrial facilities, residential areas, recreation zones, transportation routes carrying the heaviest traffic flow, and on a detailed study of the wind conditions.

Stationary posts are set up in areas permitting observations of the general diffusive atmospheric pollution in open spaces representative of a given city district (at the intersection of streets, squares, in aerated yards, etc.). Furthermore, they are placed in such a way that one can characterize the state of city air both in the center and at the outskirts, covering all types of industrial enterprises, electric power stations, petroleum product storage and distribution centers, heating centers, automobile transportation, and other sources of pollution.

In general, a certain number of posts should be set up in every city, in the proportion of one post for an area of 5-10 km<sup>3</sup> in large industrial cities and one post for an area of 2.5-5 km<sup>3</sup> in relatively small cities. However, for a number of reasons, some deviations from these requirements are tolerated in the solution of this problem. At the present time, 5-6 posts have been created in most industrial cities, and only in a few cities are there 8, 10 or even 12 posts. In keeping with the recommendations given in [1], they are placed on the leeward side (along the direction of the prevailing wind) relative to major industrial facilities or to the center of the area where the stacks of main pollution sources are located, at a distance over 20 times the average stack height.

In the Northwestern, Ural, Zabaykale and other subdivisions of the HMS, in order to find the most representative locations for the posts, a preliminary study of the wind conditions was made in different districts of each city; data for many years of meteorological observations of a series of stations located within the city outline were used for this purpose.

Comparison of data of measurements of air pollution at points selected with and without allowing for the wind conditions and the height of the sources shows that in the second case the concentration of pollutants in the atmosphere was usually higher than in the first case.

It is quite obvious that the most effective method of selecting locations for stationary sampling of air is to make a comprehensive preliminary survey of the city air by means of specially equipped motor vehicles. Such a selection method was used by A. A. Gorchiyev in the Azerbaijan subdivision of the HMS. During a certain time period, regular itinerary observations were made at a number of points selected in various districts of the city of Baku, and in addition to the collection of air samples, the meteorological conditions were recorded. From the results of the survey and data on wind conditions taken for many years, locations were chosen for stationary observations of air pollution.

### 3. Outfitting of Stationary Posts

Booths are set up for taking air samples and carrying out the meteorological observations at stationary posts. Inside the booths are placed the necessary equipment and apparatus. For direct sampling in the booth, special holes (two in each of the four walls of the booth) located at a height of 1.5 m from the earth's surface are made, with one hole to the outside through the ceiling and roof of the booth. The latter hole is used only for taking samples for analysis of gaseous ingredients, especially in cases where for whatever reasons (substantial height of snow cover, proximity of underbrush, etc.) it is inadvisable to take samples through the lateral holes. Let us note that the difference in determinations of a single concentration of atmospheric pollutants for air sampling at a height of 1.5 m (through holes in the booth walls) and 2.2 m (through the hole in the roof) is within the precision limits of the corresponding methods of measurement.

At the present time, the construction and outfitting of posts in many territorial subdivisions of the HMS is being carried out with relative success. In the Ukrainian subdivision of the HMS, the preparation of booths was organized at the Kiev Plant of Experimental Commercial Equipment. The walls of the booth were covered with glass-reinforced plastic on the outside, and were lined on the inside with structural cardboard and insulated with glass wool. Boards of pressed shavings were used for the floor and ceiling. Inside the booth is an electric instrument board, a daylight quality lamp, an electric oven and other equipment. A general view of the booth is shown in Fig. 1. Such booths have found relatively broad applications not only in the Ukraine but also in other cities of the Soviet Union.

In the Uzbek and Tajik subdivisions of the HMS at stationary posts, hydrometric GR-70 booths produced by the Tashkent Plant of Hydrometeorological Instruments were set up. In the Ural subdivision of the HMS, a project for a stationary post was worked out, and the construction of brick booths was organized. These booths have the advantage of sturdiness over other booths. However, when the location of the air sampling changes, a booth of this type can no longer be used at the new location.

To insure a normal operation of the posts from the time they are set up, in a number of territorial subdivisions of the HMS the construction of the booths was organized by the service's own assembly-and-repair teams.

In the Kamchatka subdivision of the HMS, in order to avoid changing manually the position of the intake tube projecting above the roof of the booth, a special device was constructed (Fig. 2) causing it to turn automatically under the influence of the wind's force. This device consists of a fixed metal tube 1 projecting to the outside through the ceiling and roof of the booth, and moving tube 2 with weather vane 3. Below and above are mounted ebonite sleeves 4 into which the moving tube is inserted. The latter rotates relative to the fixed tube on rolls 5 mounted in the upper sleeve. In the hole of the lower sleeve is inserted a rubber stopper 6 with two glass tubes 7, which are connected to absorbing instruments by

means of rubber hoses when the samples are taken. The device described has been tested and adopted at all stationary posts of the Kamchatka subdivision of the HMS.

While noting the desirable qualities of the system, it is often necessary to indicate its insufficient sensitivity in the presence of a weak wind, and an unjustified use of a metal tube through which the air reaches the absorbers, instead of a glass tube. For this reason, when this system is used in the HMS on a large scale, it is necessary to introduce certain changes that will eliminate these defects.

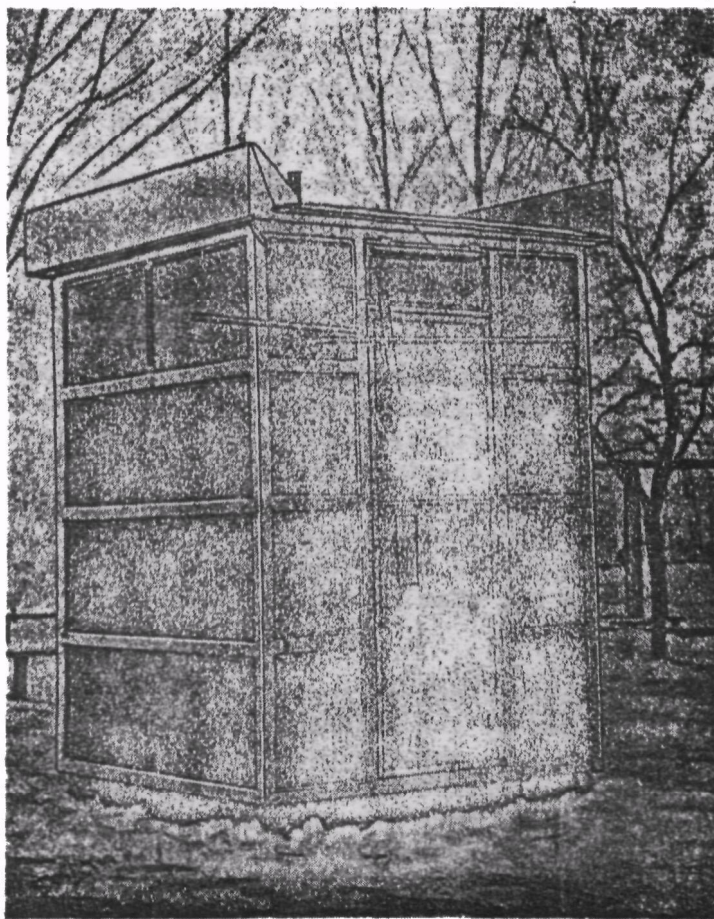


Fig. 1. General view of a booth made by the Kiev Plant of Experimental Equipment.

In cities where for one reason or another no booths have been installed for setting up and protecting the instrumentation, utility buildings of various municipal organizations are used. In this case, the collection of air samples in winter is achieved through glass tubes communicating with the outside through holes in the window frames or walls of buildings. During the warm period of the year, the samples are collected on an open platform, where the necessary equipment is placed after being brought out of the building.



In some subdivisions of the HMS (Azerbaijan, Lithuania and Kazakhstan) in collecting samples at stationary posts, use is made of motor cars that stop off at posts at certain hours every day. The cars carry the same type of apparatus as is contained in the booths. The sources of electric power used are the municipal power lines or independent sources of electric power carried by the automobile (battery, portable electric generator).

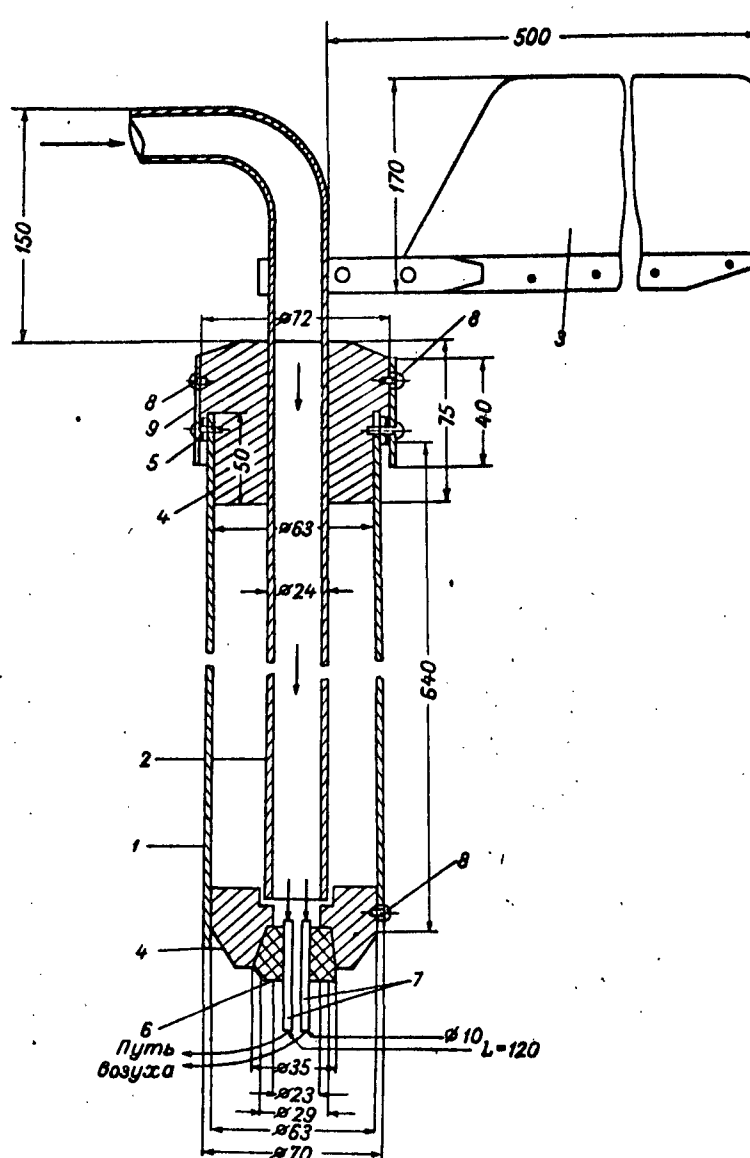


Fig. 2. Attachment for automatic rotation of intake tube under the influence of wind.

1 - stationary metal tube, 2 - moving metal tube, 3 - weather vane, 4 - ebonite sleeve, 5 - gas rollers, 6 - rubber stopper, 7 - glass tubes, 8 - bracing bolts, 9 - iron ring.

In the western Siberian subdivision of the HMS, at fixed points of the itinerary and under the plumes, instead of an electric respirator and dust collector, the UAZ-452 automobile engine is used as the air blower for collecting samples. The vacuum manifold of the engine is connected by a metal tube to the air distribution system of an LK-1 electric aspirator of L. F. Kachor's system and to an auxiliary unit replacing the automobile aspirator used in taking samples for dust analysis.

During the operation of the engine, external air is sucked in at the required rate, varied by means of performance regulators; the air enters through the absorbers and filters and reaches the engine. In the view of its originator, A. N. Seletskiy, this sampling method does not appreciably affect the wear of the automobile engine. In addition, the use of this method considerably shortens the time required for the preparation of the apparatus for sampling at each point.

#### 4. Collection of Air Samples

One of the most important steps in the operation of the posts is a suitable collection of air samples for subsequent chemical analysis. When this requirement is not met, substantial errors result in the determination of the concentration of pollutants in the atmosphere.

At the present time, at all posts, including those having no booths, air samples are taken every day except Sunday in double or triple collections in accordance with technical recommendations confirmed by the GUGMS (Main Administration of the Hydrometeorological Service of the USSR). For the purpose of comparative evaluation of the results of measurement of polluting ingredients and determination of the degree of influence of atmospheric conditions on their concentration, a single period of sample collection and meteorological observations was established for all the posts. The duration of sampling for all the ingredients was taken to be the same and equal to 20 minutes. However, these requirements are not always fulfilled by the sample takers, in many cases deviations are not considered when the results of measurement are processed, and the actual concentration of a given ingredient is thus distorted.

Electric aspirators are used, as a rule, for direct collection of air samples to be analyzed for sulfur dioxide, nitrogen dioxide, hydrogen sulfide, and soot, while gas pipets or other chambers are used for carbon monoxide. In the Krasnoyarsk subdivision of the HMS, the collection of samples for carbon monoxide analysis is carried out by means of 500 ml bottle aspirators in all types of observations (stationary posts, itinerary points and under plumes). In the collection of dust samples, the air blower used is always a "Raketa" type vacuum cleaner combined with an automobile aspirator produced by the Moscow Scientific Research Institute of Hygiene.

An important condition for the collection of samples is the consideration of the wind direction, and in a collection of samples for dust analysis, of the wind velocity as well. To this end, the intake tubes and

filters are mounted against the air flow.

In the collection of air samples for gaseous ingredients during the cold period of the year, special heaters made on the spot are used for heating up v-shaped absorption instruments.

In the section of observations of the Minsk Hydrometeorological Observatory (HMO), as suggested by S. A. Dragun, a special "radiator" was built for heating the air entering the absorption instruments. This radiator (Fig. 3) is in the form of a wooden box with a lid, measuring 20 x 20 x 27 cm. Inside the box in the bottom part is mounted a 75-100 W electric bulb that serves as the heat source, and a spiral with three turns made of tubing 6-8 mm in diameter surrounding the bulb. The ends of the spiral come out of the radiator through holes in its lateral walls.

Rubber tubes connect one end of the spiral to the outside, and the other to the absorption instrument. If samples to be analyzed for 2-3 ingredients are collected simultaneously, the second end of the spiral is provided with a T joint in order to supply the necessary quantity of absorbers. In this method of heating, the air moving toward the absorbers passes through the spiral tube, is heated up and acquires a positive temperature. The use of such a radiator in the Belorussian subdivision of the HMS made it possible to obtain the necessary number of observations in winter when the collection of air samples was made at air temperatures down to  $-30^{\circ}\text{C}$ .

Although the above-described method of sample collection at negative air temperatures is simple, it has not yet been sufficiently studied, and therefore requires careful checking in the course of practical operation of the posts under different climatic conditions with the use of spiral tubes of various materials.

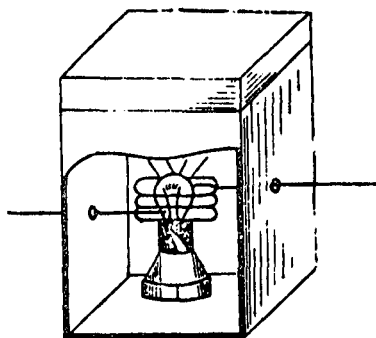


Fig. 3. Radiator for heating air during sampling in winter.

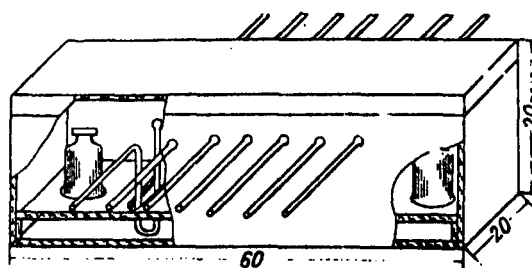


Fig. 4. Thermostat

This problem is handled in a somewhat different manner by the laboratory of water and atmosphere chemistry of the Krasnoyarsk HMO. In order to keep the solutions of reagents from freezing during the collection of samples and their transportation to the laboratory, the absorption instruments are placed in a special thermostat designed by P. D. Solonitskiy and N. S. Sigovaya.

The thermostat (Fig. 4) is in the form of a wooden box with a lid measuring 60 x 20 x 20 cm. The inner surface of the box is lined with felt. To maintain a positive temperature, two polyethylene bottles containing hot water are placed in the thermostat together with the absorption instruments. The absorbers and bottles are braced by means of a support set up inside the box. The support has 8 sockets, 6 of which are designed for the v-shaped absorbers and two for the flasks with hot water.

The longitudinal side walls of the thermostat have six holes each, through which rubber tubes connected to the ends of the v-shaped absorbers reach the outside. During the collection of samples, one end of the rubber tube is brought to the outside of the booth or body of the automobile, and the other is connected to the electric aspirator.

The use of such a thermostat enabled the water chemistry laboratory of the Krasnoyarsk HMO to carry out a systematic collection of samples at negative temperatures down to  $-38^{\circ}\text{C}$ .

In the Northwest subdivision of the HMS, the absorption instruments are heated with electric heaters of the EK-4 system. However, the practical use of these heaters is somewhat restricted by the fact that they require a current of 750 watts.

It should be noted that in practice, cases are sometimes encountered where in observations at stationary posts, pollutants usually observed in the atmosphere of any city (carbon monoxide, sulfur dioxide, etc.) are not systematically detected. This is due either to an improper choice of locations for stationary collection of samples, or an incorrect collection of air samples and errors in their chemical analysis. In order to explain the actual causes, the following is done. Using equipped automobiles, the air pollution is inspected at various points and districts of the city, and also under plumes, at different distances from the sources of pollution. In addition, in cooperation with a sanitary epidemiological station, samples are simultaneously collected at some point of the city, then subjected to separate chemical analyses in the water chemistry laboratories of the SES and UGMS. If some polluting ingredients are detected as a result of such an experiment, the errors made earlier are eliminated, and observations of the atmospheric pollution at certain points of the city are continued.

## 5. Meteorological Observations at Stationary Posts

As already noted, the evaluation of the degree of influence of meteorological factors on the concentration of pollutants in the atmosphere ordinarily utilizes observational data of a reference meteorological station located on the territory of the city or on its outskirts. However, when such a station is far removed from the post, the data of its observations cannot always be used to reduce the volume of air studied to standard conditions (760 mm Hg and  $0^{\circ}\text{C}$ .) and to determine from the wind velocity the diameter of the attachment for the automobile aspirator used for collecting samples for dust analysis. For this reason, at most locations of sample collection, observations are made to determine the air temperature, velocity

and direction of the wind and special weather phenomena (precipitation, fog, mist, etc.). Measurement of the wind is made at all points of sample collection, with the exception of those located on instrument platforms or in their vicinity.

In addition, data of meteorological observations at stationary posts are used for evaluating the influence of the microclimate on the concentration of pollutants in any given district of the city, and can also be used in solving certain special problems of major practical importance.

At the present time, meteorological observations at stationary posts have already been organized in most subdivisions of the HMS simultaneously with the sampling.

In order to carry out observations of the air temperature and wind velocity, in the HMS subdivisions of the Ukrainian SSR and Central Black Chernozem regions, a ventilation psychrometer and an induction anemometer are mounted on a special stand (Fig. 5) which is set up for the period of observations on the windward side of the booth at a distance of 3-4 m. After the observations, the instruments and the stand are placed in the booth. The wind direction is determined from a weather vane or streamer.

The stand is mounted in a vertical position by means of a metal tube 6-8 cm in diameter buried in the ground on each side of the booth. The design of the stand and its bracing tube were worked out by A. M. Lavchenko.

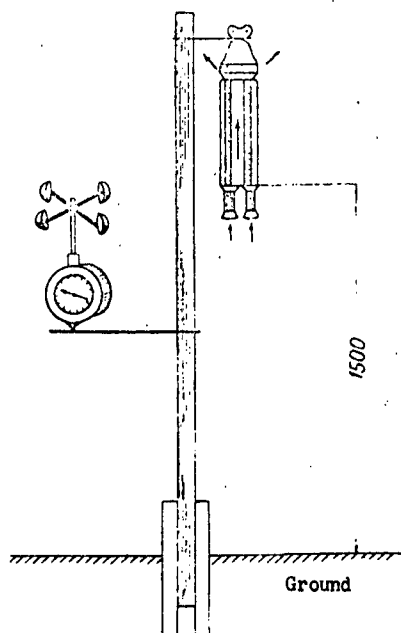


Fig. 5. Stand for suspending a ventilation psychrometer and for mounting the anemometer.

In the Transbaykal'e subdivision of the HMS, all the stationary posts are equipped with weather vanes with rhumb indicators prepared by the staff of the control bureau. The weather vanes are mounted on the roofs of booths at a height of 2.5 m from the ground. These weather vanes permit continuous observations of the direction of the wind and their correct consideration in selecting samples for analysis of polluting ingredients.

The problem of wind measurement is being solved much better in the Krasnoyarsk subdivision of the HMS, where the posts are equipped with anemorhumbometers permitting the determination of the wind velocity and direction with the required accuracy.

In conclusion, it should be noted that even though the territorial subdivisions of the HMS play a certain part in the organization of the operation of stationary posts observing atmospheric pollution, this operation is

still characterized by serious drawbacks whose elimination will be their next objective.

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DISTRIBUTION AND CHEMICAL COMPOSITION OF NATURAL AEROSOLS  
OVER VARIOUS REGIONS OF THE EUROPEAN TERRITORY OF THE USSR

Ye. S. Selezneva, O. P. Petrenchuk and P. F. Svistov

From Trudy, Glavnaya Geofiz. Observat. im. A. I. Voeykova, No. 234, p. 125-129, (1968).

Atmospheric aerosols are studied from various points of view in different areas of atmospheric physics and other sciences. The literature on the problem is very extensive and varied. Surveys on the mechanics of aerosols [9, 16] and microphysics of clouds and fogs [4, 10] have been made; a large number of studies have been devoted to the distribution of aerosols in the atmosphere [8, 4, 21], and, in particular, to the study of coarse salt particles [11, 23, 24]; new data are being reported on the content of aerosol particles in the stratosphere [20, 22], the optical and electrical properties of aerosols are being studied [17, 19], and new methods of their investigation are being developed [7, 18]. Much attention has recently been concentrated on the study of industrial aerosols and their diffusion from sources [1, 2, 3].

However, measurements of the characteristics of aerosols in the free atmosphere remain sporadic, so that information on the distribution and chemical composition of aerosols at different heights is very scarce: in the published studies, some data are given only on certain components [5, 11, 23]. No studies on a geographical scale have yet been published, although it may be assumed that the physiochemical properties of aerosols and their content in the atmosphere are directly related to the physico-geographical conditions.

In order to characterize the world background of aerosols and to study their nature, data on vast geographical territories on the scale of the continents are required. However, the use and correlation for this purpose of studies made in different countries is complicated by the variety of the methods of collection of samples and their chemical analysis, and the lack of estimates of the errors of the methods. Because of the latter factor, data of different authors, pertaining to surface conditions and to the free atmosphere, cannot be compared. Obviously, spatial correlations require specially organized investigations.

In the last few years, the Main Geophysical Observatory has been studying the distribution and chemical composition of aerosols over different regions of the Soviet Union according to a single established method. It is now possible to correlate the results obtained for the European territory (ET) of the USSR.

The chemical composition of the aerosols was studied by directly analyzing air samples collected near the earth's surface and in the boundary layer

of the atmosphere (250-1000 m) in an airplane, and also indirectly on the basis of analyses of samples of cloud and rain water. Special devices were developed for taking the samples.

The aerosol sample collector is based on the principle of inertial deposition of particles present in the air studied and their trapping when the air bubbles through distilled water. The design of the instrument is such that it may be used on land and in airplanes [15].

For collecting water samples from clouds, two types of samplers were constructed. One is used at negative air temperatures and is based on the freezing of supercooled droplets on an exposed plate.

The other type of sampler is used at positive air temperatures and is based on the principle of deposition of droplets during suction of cloud air through the sampler; the cloud droplets strike its inner surface and flow down this surface and the receiving plate into a collector [12].

All the samples were analyzed by chemical and spectral methods; their content of  $\text{Cl}^-$ ,  $\text{SO}_4^{--}$ ,  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Mg}^{++}$  and  $\text{Ca}^{++}$  ions was determined; the analytical procedure is described in a monograph [6].

In order to collect the samples and to measure the concentration of aerosol particles, a large number of flights over different regions were made. In correlating the results, the ET of the USSR was divided into 4 regions:

- I - north of ET (regions to the north of  $62^\circ\text{N}$ ),
- II - northwest of ET (Leningrad region and Baltic region),
- III - southwest of ET (White Russia and Ukraine),
- IV - east and southwest of ET (mainly land along the Volga from Kazan' to the Caspian Sea).

The characteristics of the aerosols in these regions differ considerably.

In the north of ET, the concentration of aerosol particles is 3-4 times lower than in the southwestern regions. A difference in the vertical distribution of the aerosols is observed: in the north, the decrease of the particle concentration with the height is slower than in more southern regions of the Baltic and east of the ET, where the ground layer is contaminated; beyond the limits of the boundary layer the concentration of aerosol particles is slight. The southwest of the ET stands out in aerosol distribution, where not only the ground layer of air but also the superjacent layers are heavily contaminated because of an intense vertical exchange.

The chemical composition of the aerosols in the regions under consideration also differs appreciably, as shown in Table 1. Thus, in the north, chlorides predominate in the aerosols ( $\text{Cl}^-$  ions make up 35% of the total ions), whereas in other regions sulfates predominate heavily ( $\text{SO}_4^{--}$ , 40%).



Here the main source of chlorides dissolved in water is the surface of the sea (the nonfreezing Barents Sea).

High atmospheric concentrations of aerosols and their components such as  $\text{SO}_4^{--}$ ,  $\text{Ca}^{++}$ ,  $\text{NH}_4^+$  in the south of the country may be explained by the high density of the population and of industrial enterprises, and also by a considerable wind erosion of the soil, here observed during almost the entire year.

In addition to a high content of water-soluble aerosols, a large amount of water-insoluble particles are present here; in some samples, it reached  $300\text{--}600 \mu\text{g}/\text{m}^3$ . These were cases of winter flights in subinversion layers; the trapped aerosols consisted of coarse particles of industrial origin. If these cases are excluded from the analysis and only the soluble part is considered, the aerosol concentration in the  $250\text{--}1000 \text{ m}$  layer above the southern regions amounts to an average of  $\approx 18 \mu\text{g}/\text{m}^3$ , and over northern ones, to  $\approx 2 \mu\text{g}/\text{m}^3$ .

The countable concentration of aerosol particles was determined in the same layer with the aid of a nucleus counter. On the average per year, this layer in the south contains  $2000\text{--}3000$  particles per  $\text{cm}^3$ , i.e.,  $2 \times 10^9 - 3 \times 10^9 \text{ m}^{-3}$ , and in the north  $600\text{--}700$  ( $6 \times 10^8 - 7 \times 10^8 \text{ m}^{-3}$ ). If the average size of the particles counted in the counter (condensation nuclei) is taken as  $r \sim 10^{-5} \text{ cm}$ , their mass per unit volume (for a density of 2) amounts to about  $20 \mu\text{g}/\text{m}^3$  in the south and to about  $6 \mu\text{g}/\text{m}^3$  in the north. The comparatively close coincidence of these values with the concentration of trapped water-soluble particles (Table 1) is obviously accidental, since this calculation must be regarded only as a rough estimate, but the order of magnitude of the values obtained by the different methods is the same. For this reason, more detailed comparisons may be made if the experiment is suitably arranged.

Table 1

Chemical Composition and Concentration of Atmospheric Aerosols in the  $250\text{--}1000 \text{ m}$  layer  
(Average for Year)

Region of sampling	Number of samples	Concentration, $\text{g}/\text{m}^3$									Number of particles per $\text{cm}^3$ at height of		
		$\text{SO}_4^{--}$	$\text{Cl}^-$	$\text{NO}_3^-$	$\text{NH}_4^+$	$\text{Na}^+$	$\text{K}^+$	$\text{Mg}^{++}$	$\text{Ca}^{++}$	$\Sigma \text{ion}$	$250 \text{ m}$	$500 \text{ m}$	$1000 \text{ m}$
I	13	0,29	0,58	0,05	0,07	0,29	0,10	0,03	0,21	1,62	1500	600	400
II	53	3,76	2,33	0,18	0,65	1,46	1,28	0,55	1,74	11,95	2500	1500	1000
III	95	6,96	2,26	1,44	1,92	1,88	1,11	0,56	1,50	17,63	3500	2500	1500
IV	16	3,44	1,89	0,68	0,84	1,39	0,96	0,41	1,79	11,40	3000	2000	1250

The indicated characteristics of the change of the concentration of aerosols in space and of their composition are supported by data on the chemical composition of cloud water and atmospheric precipitation [6, 13]. The largest amount of impurities in samples of cloud water and precipitation was obtained in regions of highest aerosol pollution of air. Table 2 lists data on the composition of atmospheric waters, averaged over the same four regions.

In southern regions of the ET, particularly in the southwest, sulfates predominate in the water of clouds and precipitation, whereas chlorides predominate in the north.

It is of interest to compare the relationships between the components in the aerosols and in the water samples (Table 3). They differ considerably with the different types of samples and vary with the geographical regions. Thus, the ratio  $\text{Cl}^-/\text{Na}^+$ , in the north, in aerosols and clouds is close to the value of this ratio for sea water, and even slightly higher; whereas it becomes less in the precipitation. In two other regions, the value of  $\text{Cl}^-/\text{Na}^+$  is above 1.88 in cloud water, and less in aerosols and precipitation, which may be explained by the presence of HCl in the atmosphere. This gas is more intensively absorbed by cloud elements than by aerosol particles, causing low pH values in cloud water ( $\leq 5.0$ , and in some measurements  $< 4.0$ ). In the precipitation, the pH value increases, while the ratio  $\text{Cl}^-/\text{Na}^+$  decreases as compared with cloud water, this being obviously because of the trapping of aerosol particles, for which the ratio  $\text{Cl}^-/\text{Na}^+$  is substantially less than in cloud water.

Table 2

Chemical Composition of Samples of Cloud Water and Precipitation											
Type of water sample	Content, mg/l										pH
	SO <sub>4</sub> <sup>-</sup>	Cl <sup>-</sup>	HCO <sub>3</sub> <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>	NH <sub>4</sub> <sup>+</sup>	Na <sup>+</sup>	K <sup>+</sup>	Mg <sup>++</sup>	Ca <sup>++</sup>	Σion	
North of ET											
Cloud	0,6	6,3	—	0,1	0,6	2,4	1,7	1,0	0,4	13,1	5,3
Precipitation	4,8	2,8	3,7	0,5	0,8	1,6	0,8	0,5	0,8	16,3	5,4
Northwest of ET											
Cloud	7,0	2,4	—	0,8	1,5	1,2	0,7	0,3	0,5	14,4	4,8
Precipitation	7,4	1,4	1,5	0,7	0,9	1,2	0,7	1,4	1,2	16,4	5,2
Southwest of ET											
Cloud	8,9	2,1	—	0,8	1,8	0,7	0,6	0,5	1,0	16,4	4,5
Precipitation	10,5	2,5	7,7	1,8	1,3	1,7	0,8	1,8	2,2	30,3	5,7
Southeast of ET											
Cloud	5,6	1,2	—	0,3	0,9	0,5	0,4	0,9	1,4	11,2	5,6
Precipitation	9,2	2,1	5,6	1,3	0,9	1,5	0,7	1,5	2,0	24,8	6,0

Table 3

Regions of Sampling	Mean Values of Ionic Ratios*					
	Type of Sample					
	Aerosols		Clouds		Precipitation	
	$\text{Cl}^-/\text{Na}^+$	$\text{SO}_4^{--}/\text{Cl}^-$	$\text{Cl}^-/\text{Na}^+$	$\text{SO}_4^{--}/\text{Cl}^-$	$\text{Cl}^-/\text{Na}^+$	$\text{SO}_4^{--}/\text{Cl}^-$
I	2,0	0,7	2,6	0,1	1,8	1,7
II	1,6	1,6	1,9	3,0	1,2	5,3
III	1,2	3,1	3,0	4,2	1,5	4,2
IV	1,4	1,8	2,6	1,6	1,4	4,4

\* For sea water  $\text{Cl}^-/\text{Na}^+ = 1.88$ ,  $\text{SO}_4^{--}/\text{Cl}^- = 0.14$ .

The ratio  $\text{SO}_4^{--}/\text{Cl}^-$  everywhere considerably exceeds the value of the ratio characteristic of sea water and it is lowest in aerosols. An increase of the ratio  $\text{SO}_4^{--}/\text{Cl}^-$  shows that the content of sulfur compounds in the cloud water and precipitation increases, apparently as a result of absorption of sulfur dioxide and sulfate aerosol particles.

The relative increase of the amount of  $\text{Na}^+$  (the ratio  $\text{Cl}^-/\text{Na}^+$  decreases) in aerosols and precipitation simultaneously indicates that particles of sodium compounds are present in the subcloud layer. This may be sodium bicarbonate, since a considerable amount of  $\text{HCO}_3^-$  ions was observed in precipitation of these regions. This type of particles appear to be of terrigenous origin.

The above characteristics in the proportions of the ions may be regarded as indicators of transformation of the composition of cloud water into precipitation under the influence of aerosols and gaseous impurities of the subcloud layer of air.

The total amount of impurities in the precipitation is greater than in cloud water, this being due to the above-mentioned washing of impurities out of air by the precipitation. As a result, the highest limit in the mineralization of cloud water and precipitation is observed in regions of heaviest aerosol pollution of air (regions III and IV).

A detailed quantitative analysis of all these questions also necessitate data on the structure of the clouds, rate of precipitation, spectral characteristics of aerosols before and after precipitation, and other data. The mean values listed give only an outline of the spatial changes in the composition of atmospheric precipitation in aerosols, but they serve as indicators of the most essential characteristics of these changes on the European territory of the USSR under consideration.

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## CHEMICAL COMPOSITION OF CLOUD WATER

### IN REGIONS OF WESTERN SIBERIA

O. P. Petrenchuk

From Trudy, Glavnaya Geofiz. Observat. im. A. I. Voeykova, No. 234, p. 130-136, (1968).

An important factor in atmospheric self-purification involving various impurities, both those of natural origin and those caused by man's industrial activity, is atmospheric precipitation. The composition of the latter depends on a number of parameters, for example, the chemical nature of the condensation nuclei, the physico-geographical location of the points of sampling, and the content of aerosols and gaseous impurities because of local pollution sources (industrial discharges, underlying surface, etc.) in the subcloud layer of the atmosphere, this content being considerably dependent on the stratification of the atmosphere. The process of formation of the chemical composition of the precipitation takes place in the cloud itself, and it is, therefore, important to study the chemical composition of cloud water and to explain its causes, particularly the influence of meteorological conditions.

The present paper examines the chemical composition of cloud water on the basis of samples collected in the vicinity of Krasnoyarsk, Yeniseysk, Podkamennaya Tunguska and other cities and towns of Western Siberia. The organization of systematic work involving the collection of samples of cloud water at a point of airplane sounding in Krasnoyarsk made it possible to obtain for the first time data on the chemical composition of cloud water in regions of Western Siberia under various meteorological conditions. The cloud water samples were collected by flight aerologists V. G. Dedkov, A. A. Ignat'yeva, and N. A. Volkova, and analyzed in the chemical laboratory headed by V. M. Drozdova.

An inspection of the data obtained is of special interest, since, according to studies of the distribution of condensation nuclei [3] and chemical composition of the precipitation [4], the atmosphere is basically pure in the north of the European territory of the USSR and over a vast area of the Asian taiga. On the average, the total impurities dissolved in the precipitation do not exceed 10-15 mg/l. However, under unfavorable meteorological conditions, in the vicinity of large industrialized cities, a heavy pollution of the atmosphere is observed, which causes an increase in the concentration of condensation nuclei and in the mineralization of the precipitation.

It is of interest to determine the mineralization of cloud water in regions of western Siberia and the extent of its variation with the meteorological conditions.

The analysis of the influence of meteorological conditions made use of data on the chemical composition of cloud water collected in various synoptic situations (about 60 cases in all). Samples of water were obtained from clouds formed under anticyclonic and cyclonic conditions, in cases of slight advection and marked transport of air masses, and a considerable part of the samples were collected from precipitating frontal clouds.

The stratification of the atmosphere under anticyclonic conditions and slight advection was characterized by the presence of a temperature inversion. The clouds were located under the inversion, and their vertical thickness was 150-500 m.

Let us consider a few characteristic examples, for which data of chemical analysis are given in Table 1.

Under anticyclonic conditions with a slight advection of the air masses, two samples of cloud water were obtained (No. 1, 2). Sample No. 2 was collected on 16 May 1966, 50 km to the north of Krasnoyarsk. An extensive anticyclone with its center at Abakan was located in the south of Siberia at that time. Krasnoyarsk was in the central part of the anticyclone. At the sampling point, a wind from the direction of Krasnoyarsk was observed. The sample was taken from subinversion fine-droplet Sc clouds with a water content of  $0.13 \text{ g/m}^3$  and with a vertical thickness of 350 m; its total mineralization was  $30.8 \text{ mg/l}$ . The results of temperature sounding in the atmosphere on 16 May 1966 during the collection of the sample are shown in Fig. 1 b.

Sample No. 1 was collected from Ac clouds at a height of 2230 m on 19 October 1965 in the vicinity of Krasnoyarsk, in the northwestern part of a vast anticyclone with its center on the territory of Mongolia. The clouds were located under a layer of a marked temperature inversion (fig. 1 a) and were characterized by a slight vertical thickness (190 m) and low water content ( $0.15 \text{ g/m}^3$ ). The collected sample of cloud water had a dark color, and its mineralization was  $32 \text{ mg/l}$ .

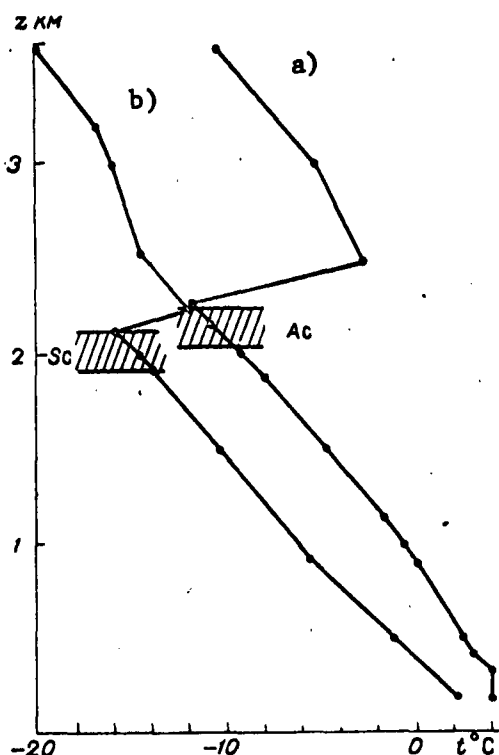


Fig. 1. Results of temperature sounding of the atmosphere in Krasnoyarsk.

a - 19 October 1965; b - 16 May 1966.

Chemical Composition of Samples of Water from Clouds in the Vicinity of Krasnoyarsk in Various Synoptic Situations.

Sample Number	Date	Cloud form	Height of sampling in meters	Concentration, mg/l									pH	$\times 10^6$ $\text{ohm}^{-1} \text{cm}^{-1}$	$\Sigma \text{ion}$	Synoptic situation
				$\text{SO}_4^{=}$	$\text{Cl}^-$	$\text{NO}_3^-$	$\text{HCO}_3^-$	$\text{NH}_4^+$	$\text{Na}^+$	$\text{K}^+$	$\text{Mg}^{++}$	$\text{Ca}^{++}$				
1	19 X 1965 r.	Ac	2230	25,0	1,0	0,9	0,0	2,5	0,3	0,3	1,7	0,3	3,8	—	32,0	Slight advection of air masses in central part of the anticyclone or in intermediate pressure field
2	16 V 1966 r.	Sc	2130	18,3	1,7	0,9	0,9	5,2	0,6	0,6	0,4	2,2	4,9	65	30,8	
3	16 X 1965 r.	Ac	2990	1,4	0,3	0,3	0,3	0,2	0,0	0,0	0,1	0,4	5,2	8	3,0	Development of cyclonic activity; frontal cloudiness associated with precipitation
4	16 X 1965 r.	Ac	3630	1,1	0,3	0,6	0,4	0,1	0,0	0,0	0,1	0,4	5,2	9	3,0	
5	16 X 1965 r.	Ac	3780	0,9	0,8	0,8	0,0	0,2	0,3	0,4	0,1	0,3	5,0	10	3,8	
6	23 V 1966 r.	Sc	2240	3,4	0,4	0,2	0,0	0,7	0,1	0,1	0,5	0,2	5,0	16	5,6	
7	23 V 1966 r.	Sc	2080	3,3	0,4	0,1	0,0	0,5	0,2	0,1	0,3	0,3	5,0	13	5,2	Marked advection of air masses from the north in forward part of anticyclone or rear of cyclone.
8	4 V 1967 r.	Sc	1120	4,6	0,6	0,0	0,0	0,5	0,8	0,1	0,2	0,3	5,0	16	7,1	
9	4 V 1967 r.	Sc	1310	3,7	1,0	0,0	1,7	0,6	1,0	0,4	0,5	0,4	5,5	18	9,3	
10	4 V 1967 r.	Sc	1800	5,6	2,0	0,0	0,0	0,6	1,7	0,7	0,6	0,3	4,8	26	11,5	
11	29 IX 1965 r.	Sc	1580	4,2	0,5	0,6	1,7	0,4	0,2	0,2	0,6	1,1	5,4	17	9,5	
12	10 V 1966 r.	Sc	1780	6,6	0,6	0,8	0,6	1,1	0,4	0,4	0,2	1,6	4,8	27	12,3	

The presence of pronounced temperature inversions in combination with low wind velocities under anticyclonic conditions in the region of Krasnoyarsk promotes the accumulation of impurities in the subcloud layer of the atmosphere, and this, as shown by the data of Table 1, is reflected in the composition of the cloud water, despite the considerable height of the sampling.

The  $\text{SO}_4^{=}$  ions, chiefly due to industrial discharges, predominate in water samples from subinversion clouds (No. 1 and 2). They amount to 80% of the total of all the ions. Accordingly, the ratio  $\text{SO}_4^{=}/\text{Cl}^-$  is also large. There is also observed a somewhat increased concentration of  $\text{NH}_4^+$  ions as compared with the concentration of the remaining ions.

The chemical composition of samples taken under cyclonic conditions from precipitating frontal clouds (samples No. 3-7) is substantially different.

On 16 October 1965, the development of cyclonic activity was observed over a considerable area of the south of western Siberia. A marked pressure drop was observed in the region of Krasnoyarsk and to the west of the latter. A wave front passed from the southwest to the northeast and across Krasnoyarsk. A vast area of precipitation was observed along this front. In the vicinity of Krasnoyarsk, at three heights beginning with approximately 3000 m and higher, three samples (No. 3, 4, 5) were taken from Ac clouds.

It is evident from the table that the mineralization of samples in this case is one order of magnitude lower than that of samples from subinversion clouds and does not exceed 4.0 mg/l. The value of the ratio  $\text{SO}_4^{=}/\text{Cl}$ , which indirectly characterizes the influence of industrial pollution, decreased substantially.

The Ac clouds consisted of fine droplets and crystals, and their water content increased from 0.20 g/m<sup>3</sup> at heights of 2990 and 3600 m to 0.43 g/m<sup>3</sup> at 3800 m. Petrenchuk et al. [2] noted the existence of a dependence of the composition of cloud water on the microphysical characteristics of the cloud. In particular, it was observed that the more mineralized samples of cloud water corresponded to lower values of the effective droplet radius and of the water content of the cloud. Such low mineralization values of cloud water for comparatively moderate values of the water content and the predominance of fine droplets indicate in this case an extreme purity of the water on precipitating frontal clouds.

On 23 May 1966, an occlusion front associated with a vast precipitation zone moved across Krasnoyarsk. In the area where the hydroelectric power station is located, to the southwest of Krasnoyarsk, two samples (No. 6 and 7) were taken from fine-droplet clouds with a water content of 0.71 g/m<sup>3</sup> at two heights. They were characterized by a relatively low mineralization and were similar in chemical composition to the samples taken on 16 October 1965.

Clouds forming in an air mass of Arctic origin in the forward part of large anticyclones or in the rear of cyclones may be classified in a separate



Table 2

Chemical Composition of Water From Clouds on the Territory of Western Siberia in Various Synoptic Situations

Sample number	Date	Cloud form	Height of sampling	Concentration, mg/l										pH	$\times 10^6 \text{ ohm}^{-1} \text{ cm}^{-1}$	$\Sigma \text{ion}$	Point of sampling	Synoptic Situation
				$\text{SO}_4^{--}$	$\text{Cl}^-$	$\text{NO}_3^-$	$\text{HCO}_3^-$	$\text{NH}_4^+$	$\text{Na}^+$	$\text{K}^+$	$\text{Mg}^{++}$	$\text{Ca}^{++}$						
1	19 X 1966	Sc	1650	6,6	1,0	0,7	4,6	3,1	0,2	0,1	0,3	0,1	6,2	33	16,7	Yeniseysk region	Slight advection of air masses	
2	3 VI 1966	Ac	3420	3,8	7,7	0,3	0,0	0,9	2,6	3,4	0,2	0,5	5,0	46	19,4	Northwest (50km) from Podkanennaya Tunguska		
3	6 X 1966	Sc	2320	1,6	0,8	0,1	0,0	0,3	0,2	0,3	0,2	0,3	5,0	8	3,8	To the north (100 km) of Yeniseysk	Development of cyclonic activity	
4	13 IX 1966	Sc	1580	0,8	0,2	0,3	0,6	0,3	0,0	0,0	0,2	0,2	5,4	6	2,6	Region of Boguchany		
5	9 IX 1965	Cb	1380	0,8	0,5	0,1	2,0	0,2	0,2	0,1	0,2	0,3	5,3	11	4,4	Region of Podkanennaya Tunguska	Marked advection of air masses from the north	
6	9 IX 1965	Cb	1620	0,9	0,2	0,1	0,0	0,1	0,1	0,0	0,0	0,3	5,2	5	1,7			
7	9 IX 1965	Cb	1840	1,1	0,4	0,1	1,0	0,2	0,1	0,1	0,2	0,2	5,4	8	3,4			

intermediate group. Samples from such clouds also are characterized by low mineralization values. However, their impurity concentration is somewhat higher than that of precipitating frontal clouds.

The chemical composition of samples of cloud water (No. 8-12) collected under conditions of advection of air masses from the north is given as an example. Samples No. 8-10 were taken on 4 May 1967, 50 km north of Krasnoyarsk from Sc clouds of mixed structure with a water content of 0.35-0.50 g/m<sup>3</sup>. The area of collection was located in the frontal part of a large anticyclone in the area of a pronounced northern transport. Results of atmospheric sounding show that the clouds were located under a layer of temperature inversion, which is frequently observed in the forward part of the anticyclone (Fig. 2 a). However, the presence of an inversion in this layer does not cause the accumulation of impurities in the subinversion layer, as was the case in a low-gradient pressure field. Indeed, in a rapid displacement of the air masses arriving from the sparsely populated areas of northern Siberia, their transformation is slowed down, and they are less exposed to the influence of ground sources of pollution.

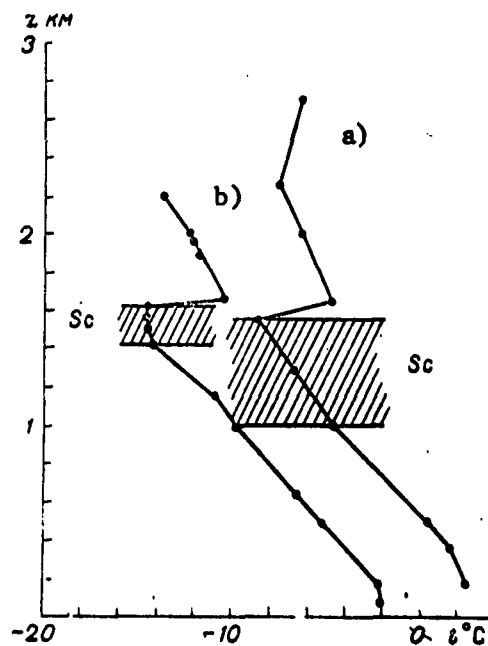


Fig. 2. Results of temperature sounding of the atmosphere.

a - 4 May 1967 in Krasnoyarsk;  
b - 19 October 1966 in Yeniseysk.

Table 1 shows the results of chemical analysis of samples taken on 10 May 1966 in the area of Krasnoyarsk, located in the rear of the cyclone, from clouds formed during passage of secondary cold fronts (No. 12), and on 29 September 1965, when the point of sampling was located in the northern part of the anticyclone (No. 11). The chemical composition of these samples differs only slightly, and in mineralization they resemble samples No. 9 and 10.

In regions of western Siberia farther to the north (environs of Turukhansk, Podkamennaya Tunguska, Yeniseysk, Achinsk, Boguchany), the cloud water is still less mineralized. However, in subinversion clouds (Fig. 2 b), formed in a low-gradient pressure field, an increased impurity concentration reaching 20 mg/l is observed. However, in clouds formed in other synoptic situations, particularly upon intrusion of Arctic air masses into the forward part of an anticyclone or the rear of a cyclone or during the development of cyclonic activity, the total mineralization amounts to an average of 3-6 mg/l. Some typical cases with results of chemical

## COULOMETRIC METHOD OF DETERMINATION OF SULFUR-CONTAINING COMPOUNDS IN AIR

N. Sh. Vol'berg

From Trudy, Glavnaya Geofiz. Observat. im. A. I. Voeykova, No. 238, p. 107-114, (1969)

The widespread use of automatic instruments for the determination of toxic impurities in air is hindered by an inadequate sensitivity and by the high cost of gas analyzers. The creation of a bank of automatic instruments in every laboratory which would make it possible to determine even the most common air pollutants is a very difficult problem. Therefore, the development of universal, continuous-action gas analyzers suitable for determining microimpurities in air is quite urgent. Of major interest in the design of such instruments is the coulometric method.

The coulometric method has long been extensively used in analytical chemistry for the analysis of solutions, but its chief advantages in gas analysis have been discovered only recently thanks to the works of Hersch [7], Novak [8] and a number of other scientists. The coulometric analysis of gases is based on the measurement of the current of the electrode reaction involving the substance being determined, which acts as a depolarizer and is continuously supplied to the electrolytic cell with the stream of analyzed gas.

Depending on the nature of the reaction taking place at the electrode, the coulometric method may be used for the determination of reductants or oxidants.

The advantage of this method is the theoretical possibility that the electrode reaction can take place with a 100 percent current efficiency. This makes it possible to calculate the value of the measured concentration from Faraday's law without the need for calibration. The participation in the reaction of the entire substance supplied ensures a high sensitivity of the determinations, making the method particularly suitable for determining microconcentrations of various impurities in air and other gases. A high sensitivity can also be attained even without the use of current amplifiers by using ordinary microammeters. In contrast to the other electrometric (polarographic, conductometric, potentiometric, etc.) methods, in the coulometric method the magnitude of the current is determined only by the amount of the electrochemically active substance supplied to the cell, and is almost independent of factors usually affecting the results of measurements in other methods such as temperature, state of the electrode surface, stirring rate, etc.

Of particular importance is the absence of a temperature dependence of the readings, so that thermostating of the cells is unnecessary. These

analysis of cloud water in various regions of western Siberia are given in Table 2.

Table 3 lists mean values of the impurity concentration in samples of water from two groups of clouds: frontal and subinversion clouds for the entire investigated territory of western Siberia. The first group also includes samples collected in clouds formed during a marked advection of air masses of Arctic origin, since their chemical composition differs little from that of frontal clouds. An inspection of the data of this table shows that the mineralization of the water samples from subinversion clouds is over 3 times that of the water from precipitating frontal clouds.

Table 3

Chemical Composition of Cloud Water in Regions of Western Siberia.													
Cloud Type	Concentration, mg/liter									pH	$\Sigma \times 10^6$ ohm <sup>-1</sup> cm <sup>-1</sup>	$\Sigma$ ion	Number of Samples
	$\text{SO}_4^{--}$	$\text{Cl}^-$	$\text{NO}_3^-$	$\text{HCO}_3^-$	$\text{NH}_4^+$	$\text{Na}^+$	$\text{K}^+$	$\text{Mg}^{++}$	$\text{Ca}^{++}$				
Frontal	2,8	0,6	0,3	0,6	0,4	0,3	0,2	0,3	0,3	5,11	13,6	5,8	44
Subin- version.	11,5	1,9	0,6	1,1	2,2	0,9	0,8	0,7	0,8	4,84	48,2	20,5	13

Since the chemical composition of the water of frontal clouds is relatively unaffected by local pollution sources, the results of its chemical analysis can obviously be used to evaluate the role of clouds in the formation of the chemical composition of the precipitation.

Data on the chemical composition of subinversion clouds may be used for characterizing the degree of atmospheric pollution by local discharges. The mineralization values cited lead to the conclusion that industrial discharges substantially affect the chemical composition of cloud water, but that the atmosphere of the investigated regions of western Siberia is much less polluted than some regions of the southwest of the European territory of the USSR, where the mineralization of samples of water from subinversion clouds occasionally reaches about 300 mg/l [1].

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advantages of the coulometric method favor the creation of portable gas analyzers of high sensitivity.

The coulometric method has been used as the basis for the design of instruments for determining a number of toxic substances in air: sulfur dioxide, halogens, ozone, nitrogen dioxide, carbon monoxide, and some others.

Since the group of substances which can undergo a direct coulometric determination is limited to electrochemically active, chiefly inorganic gases, efforts to expand it through chemical conversions of inactive compounds are justified.

The object of the present study was to determine the conditions for the coulometric determination of sulfur-containing compounds. It was assumed that the substances containing sulfur could be burned to sulfur dioxide, which is readily determinable coulometrically. As the representative of such compounds we selected carbon disulfide, a substance widely employed in industry and possessing a high toxicity.

#### Determination of Carbon Disulfide

A large number of studies have been devoted to the problems of combustion of sulfur-containing substances. Combustion is one of the most common methods of elemental analysis of organic substances and the most widespread method in the determination of sulfur in metal ores.

The combustion of substances containing sulfur forms a mixture of its oxides. As we know, the ratio of  $\text{SO}_2$  to  $\text{SO}_3$  is determined by the temperature of the process. At comparatively moderate temperatures of the order of 400-500°C, the equilibrium is markedly shifted toward the formation of  $\text{SO}_3$  and only at temperatures above 1000°C does the bulk of the  $\text{SO}_3$  dissociate into  $\text{SO}_2$ .

Under analytical conditions, the ratio of  $\text{SO}_2$  to  $\text{SO}_3$  is also affected by the presence of catalysts, the oxygen concentration in the gaseous mixture, and the composition of the latter.

For this reason, in the absolute majority of cases, the final determination is made on the  $\text{SO}_4^{2-}$  ion, eliminating the sulfur oxides by various absorbers and oxidizing  $\text{SO}_3$  to  $\text{SO}_4^{2-}$ .

The determination of the sulfur content in terms of sulfur dioxide is used chiefly in different variants of the lamp method, in which the oxidation to  $\text{SO}_3$  is comparatively slight. For a gas analyzer, it is more convenient to carry out the combustion in a tube heater.

The determination of sulfur in organic compounds in terms of  $\text{SO}_2$  after combustion in an empty quartz tube has been closely studied by Dokladalova [6]. It was found that the systematic negative error caused by the conversion of sulfur dioxide to the trioxide is determined by the temperature in the combustion zone and is independent of the structure of the substances undergoing combustion.

The authors contend that the combustion in an empty quartz tube in a

stream of air at 800°C can be used for determining the content of sulfur in organic substances from the sulfur dioxide formed.

To determine sulfur dioxide, we used a coulometric cell designed by V. Z. Al'perin et al. for the analysis of atmospheric air [1, 4].

It was of interest first to explain the degree of oxidation of  $\text{SO}_2$  on passing air through an empty quartz tube at a given rate and different temperatures.

The gas-air mixture, containing a constant amount of sulfur dioxide, was passed at a rate of 150 ml/min through a quartz tube 6 mm in diameter into a coulometric cell connected with an N 373-1 automatic recorder. The tube was inserted into an MA 2-20 tube heater whose temperature was measured and maintained at a certain level within  $\pm 10^\circ\text{C}$  by means of an EPB-2 controller. The length of the incandescent zone of the heater was 190 mm. The gas was supplied at a constant rate from a diffusion dispenser [2] filled with a solution of sulfur dioxide and water.

A check showed that in the temperature range from 20° to 1100°C in the quartz tube, the current of the coulometric cell, proportional to the amount of  $\text{SO}_2$ , underwent no change, indicating the absence of oxidation of sulfur dioxide by atmospheric oxygen in the empty quartz tube.

Further, the combustion of carbon disulfide was checked in the same tube at a constant feed rate of 1.2  $\mu\text{g}/\text{min}$ , an air drawing rate of 50-250 ml/min, and at different temperatures, 700°-1000°C.

The constant amount of carbon disulfide was steadily supplied from a diffusion dispenser described below [3].

Results of measurements of the coulometric cell current are shown in Fig. 1. It is evident from the latter that at 850°C the yield of sulfur dioxide reaches a maximum and does not increase with a further elevation of temperature. This attests to the completion of the combustion of carbon disulfide.

Considering the possibility of voltage fluctuations in the line, the working temperature chosen was 900°-950°C.

The influence of the flow rate of the analyzed air on the yield of sulfur dioxide at a given temperature was checked. It was found that in the range of 50-250 ml/min, the consumption of the analyzed air did not affect the results obtained.

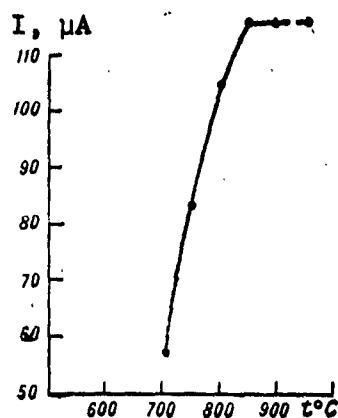


Fig. 1. Cell current vs. combustion temperature of carbon disulfide.

In order to create a technical basis for the construction of a coulometric gas analyzer, it was desirable to use elements as small and as light as possible. An attempt was therefore made to use a heater with a shorter combustion zone, 55 mm. In this case, quantitative combustion began only at 1050°C.

The use of a catalyst - platinum wire with an area of 5 cm<sup>2</sup> rolled into a cylinder - did not yield any positive results. On the contrary, the catalyst decreased the yield of sulfur dioxide by 7% as compared with the empty tube, even at 1150°C.

In addition to the empty quartz tube heated from outside, the following were tested: a fine quartz tube 3 mm in diameter heated from outside to 900°C, placed in a similar tube 6 mm in diameter, and a spiral of platinum wire heated by a current to 1200°C and placed inside a wide quartz tube. In both cases the yield was lower than in the empty quartz tube.

Thus, the following conditions were found to be optimal for the combustion of carbon disulfide: quartz tube diameter 6 mm, length of heating zone 190 mm, temperature 850°-950°C, air rate up to 250 ml/min. Under these conditions, certain carbon disulfide concentrations (from 0.3 to 10 mg/m<sup>3</sup>) obtained by means of the above-described diffusion dispenser were analyzed repeatedly.

The carbon disulfide concentration (mg/m<sup>3</sup>) was calculated from the formula

$$c = 0,0118 \cdot \frac{I - I_0}{v},$$

where 0.0118 is the electrochemical equivalent of CS<sub>2</sub>, µg/min · µA; I is the cell current, µA; I<sub>0</sub> is the background current, µA; v is the air flow rate, l/min.

In a series of 35 determinations, the concentration values found amounted to an average of 97% of the assumed values for a probable square deviation of the arithmetic mean of ±3% (for an accuracy of 0.95).

On the basis of the data obtained, a correction factor of 0.97 was introduced into the formula for calculating the carbon disulfide concentrations. The refined formula is

$$c = \frac{0,0118}{0,97} \cdot \frac{I - I_0}{v} = 0,0122 \cdot \frac{I - I_0}{v}.$$

Removal of interfering impurities. Since in many technological processes carbon disulfide is associated with sulfur dioxide and hydrogen sulfide, particular attention was given to the problem of purification of the analyzed air by removal of the indicated impurities. A series of adsorbents were tested for this purpose: sodium acetate, sodium and potassium

carbonates (for trapping sulfur dioxide) and lead acetate, copper sulfate, and cadmium oxalate (for trapping hydrogen sulfide). These substances were deposited on the inert diatomaceous support TND (spherochrome-1).

The effectiveness of the absorption of  $\text{SO}_2$  and  $\text{H}_2\text{S}$  and the influence of the adsorbents on the accuracy of the determination of carbon disulfide were checked. The experiments showed that the carrier impregnated with lead acetate solution effectively holds hydrogen sulfide without adsorbing  $\text{CS}_2$ . Potassium carbonate deposited on TND in the form of a saturated solution containing phenolphthalein indicator may be recommended for trapping sulfur dioxide.

As little as 0.3 ml of adsorbent is enough to hold either  $\text{SO}_2$  or  $\text{H}_2\text{S}$ .

The adsorbent for sulfur dioxide is prepared in the following manner. Carrier TND-TS-M (spherochrome-1) (the 0.25-1 mm fraction) is boiled with 1:2 hydrochloric acid for two hours, washed until the reaction is neutral, and heated for two hours at 800-900°C. The carrier is wetted with a saturated solution of potassium carbonate in the proportion of 1 ml to 3 ml of carrier, stirred, and dried at 100-150°C. The powder is then colored with a few drops of an alcohol solution of phenolphthalein and dried at 50-60°C. With use, the adsorbent loses its red color. After one-half of its layer has been used up, the adsorbent is replaced.

To prepare the adsorbent for hydrogen sulfide, the inert carrier, purified as described above, is drenched with a lead acetate solution saturated at room temperature in the proportion of 1 ml to 3 ml of carrier, and dried at 100-120°C. As it becomes saturated with hydrogen sulfide, the adsorbent turns black. It is replaced after one-half of its layer has been used up.

#### Determination of Hydrogen Sulfide

As we know, hydrogen sulfide, like sulfur dioxide can reduce elemental iodine to hydrogen iodide, and is determined from the current of oxidation of the latter on a noble metal electrode [9]. However, as was shown experimentally, relative to  $\text{SO}_2$ , the allowed drawing rate of the gas being analyzed through the coulometric cell is 5-7 times lower in this case, owing to the lower solubility of hydrogen sulfide in the aqueous medium and the slower rate of reaction with iodine. It was useful therefore to carry out a preliminary combustion of hydrogen sulfide to sulfur dioxide. It was found possible to use the same conditions for this purpose as for the combustion of carbon disulfide.

Fig. 2 shows comparative current efficiency data for the direct reaction of hydrogen sulfide with the electrolyte and after its combustion to  $\text{SO}_2$  for different drawing rates of air. The amount of  $\text{H}_2\text{S}$  supplied from the



diffusion dispenser remained constant and equal to 1  $\mu\text{g}/\text{min}$ . Although after the combustion the magnitude of the signal obtained decreases by approximately one-half, the sensitivity of the determinations can be increased severalfold because the drawing rate of the analyzed air can be considerably increased.

To determine the yield of  $\text{SO}_2$  from the combustion of  $\text{H}_2\text{S}$ , the results of coulometric determination of the amount of hydrogen sulfide discharged from the dispenser per minute were compared with data of chemical analysis performed by using a standard method [5]. The data listed in Table 1 show a relatively high yield of  $\text{SO}_2$ .

The preliminary combustion of  $\text{H}_2\text{S}$  not only affords the possibility of increasing the sensitivity of the determinations, but considerably increases the selectivity of the analysis. This is because under the conditions adopted for the combustion of hydrogen sulfide, all the interfering organic impurities capable of reducing iodine like  $\text{SO}_2$  (for example, aldehydes, ketones, etc.) oxidize to  $\text{CO}_2$ . Under the selected conditions, only organic sulfur compounds and  $\text{SO}_2$  interfere with the determination of  $\text{H}_2\text{S}$ . To eliminate  $\text{SO}_2$  from the analyzed air, a series of adsorbents were tested. We based their selection on the fact that sulfur dioxide has much stronger acidic properties than  $\text{H}_2\text{S}$ . Therefore, salts of acids weaker than  $\text{SO}_2$  were tested as absorbers. The salts were deposited on the inert carrier spherochrome-1 in the form of saturated aqueous solutions in the proportion of 1 ml to 3 ml of powder and were dried at 120-150°C.

All the indicated adsorbents completely trapped sulfur dioxide (concentration 30  $\text{mg}/\text{m}^3$ ) for a layer volume of 0.3 ml and an air flow rate of 100  $\text{ml}/\text{min}$  (Table 2). The trapping of hydrogen sulfide by these salts was checked on a small concentration of  $\text{H}_2\text{S}$ , about 1  $\text{mg}/\text{m}^3$ .

All the  $\text{SO}_2$  absorbers tested trap to a slight degree also  $\text{H}_2\text{S}$ . The best results were obtained with sodium carbonate, which decreased the hydrogen sulfide concentration by only 3%. An attempt to use the oxidants potassium chlorate and hydroperit for trapping  $\text{SO}_2$  were unsuccessful.

The effect of the presence of organic substances in the analyzed gas on the yield of sulfur dioxide from the combustion of  $\text{H}_2\text{S}$  was checked in special experiments. This check was necessitated by differences in the behavior of sulfur dioxide in the ignited tube depending on its origin. Whereas  $\text{SO}_2$  formed from organosulfur compounds oxidizes partially to  $\text{SO}_3$  during the combustion, ready  $\text{SO}_2$  was shown by our observations to pass with the stream of air through the ignited quartz tube without oxidizing. This suggested the

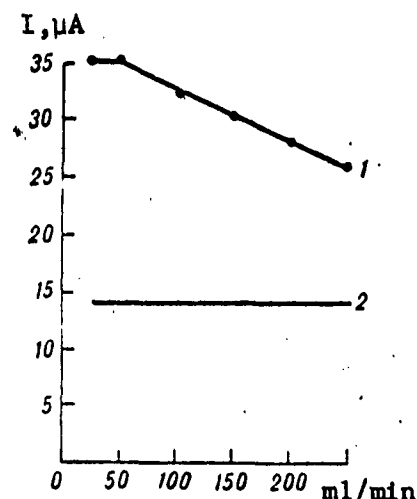


Fig. 2. Current vs. rate of passage of analyzed air through the coulometric cell in the determination of hydrogen sulfide.

1 - without combustion, 2 - with preliminary combustion.

Table 1

Comparative Results of Determination of Hydrogen Sulfide ( $\mu\text{g}/\text{min}$ ) by the Standard Method and the Coulometric Method after combustion to  $\text{SO}_2$ .

Method					
Standard	Coulometric	Standard	Coulometric	Standard	Coulometric
1.40	1.38	2.00	1.9	0.39	0.40
1.27		2.03		0.43	
1.39		1.97		0.35	
1.33		1.93		0.42	
1.43				0.40	

Table 2

Adsorbents for Sulfur Dioxide.

Substance	Carrier	% Retained	
		$\text{SO}_2$	$\text{H}_2\text{S}$
$\text{K}_2\text{CO}_3$	Spherochrome-1	100	5
$\text{Na}_2\text{CO}_3$	"	100	3
$\text{NaC}_2\text{H}_3\text{O}_2$	"	100	5
$\text{K}_2\text{HPO}_4$	"	100	7
$\text{Na}_2\text{CO}_3$	—	100	3

Table 3

Effect of the Presence of an Organic Substance on the Yield of  $\text{SO}_2$  from the Combustion of  $\text{H}_2\text{S}$ .

Concentration of Acetone, $\text{mg}/\text{m}^3$	Cell Current	
	$\mu\text{A}$	%
0	78	100
20	75	96
34	73.5	94

possibility of an induced oxidation of sulfur dioxide. To elucidate this question, different amounts of vapor of an organic substance (acetone) were introduced into the stream of air containing a constant  $\text{H}_2\text{S}$  concentration ( $8 \text{ mg}/\text{m}^3$ ) (Table 3). The value of the coulometric cell current, measured for the case of combustion of  $\text{H}_2\text{S}$  in air without other admixtures, was taken as 100%.

It is evident from the data of Table 3 that the presence of other organic substances affects the yield of  $\text{SO}_2$ , but to a small extent. Therefore, the applicability of this method to the determination of  $\text{H}_2\text{S}$  in gases containing substantial concentrations of organic substances requires further checking.

#### Determination of Sulfur Dioxide in the Presence of Other Reductants

The coulometric method is at the present time the most promising for the determination of microconcentrations of sulfur dioxide in air. However, the scope of this method is considerably limited by the nonspecificity of

the analysis in the presence of other reductants such as acetone. This makes the analysis unreliable in the presence of products of incomplete combustion of organic compounds.

Attempts to select an absorber which removes the interfering impurities and does not hold  $\text{SO}_2$  have been unsuccessful. The selectivity of the determinations was successfully achieved by using the above-mentioned stability of sulfur dioxide to oxidation in an empty heated quartz tube, since all the other substances lose their reducing properties under these conditions.

Despite the inconvenience of using a heater in the apparatus, only this method is applicable in the presence of other reductants (except  $\text{H}_2\text{S}$  and other sulfur-containing compounds). A series of reagents were tested for their capacity to hold hydrogen sulfide: lead acetate and copper sulfate on spherochrome-1, cadmium oxalate on flourolone-4, metallic silver on silochrome-3. The most convenient reagent was found to be copper sulfate, deposited on the inert carrier spherochrome-1 washed with hydrochloric acid and water and ignited at 900-1000°C.

When the method described is used for determining  $\text{SO}_2$  in air containing high concentrations of oxidizing substances, the possibility of induced oxidation must be borne in mind just as in the determination of  $\text{H}_2\text{S}$ .

#### Summary

1. The determination of air of microimpurities of vapors and gases of sulfur-containing compounds is best carried out by using the coulometric method for sulfur dioxide, after passing the gas through an empty quartz tube ignited to 850-1000°C.
2. The size of the combustion zone depends on the required flow rate of the analyzed air and for 0.25 l/min are  $l = 200$  mm,  $d = 6$  mm.
3. When sulfur dioxide and hydrogen sulfide are determined, a preliminary combustion substantially increases the specificity of the determinations in the presence of other reductants. It is shown that in contrast to other reductants,  $\text{SO}_2$  does not oxidize when passing with a stream of air through a heated empty quartz tube. In addition, in the presence of a large amount of combustible substances, there takes place a slight induced oxidation of sulfur dioxide which requires a special study.
4. Selective solid chemical adsorbents that gradually change color as they are used up were chosen for the separate determination of sulfur dioxide, hydrogen sulfide and carbon disulfide when these compounds are present together.

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