REMOTE MONITORING OF ATMOSPHERIC POLLUTION AND EMISSIONS IN URBAN TERRITORIES

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The salient features and difficulties associated with solution of specific problems of monitoring of the pollution of atmospheric air in urban territories are considered under conditions of limited instrumental base for local measurements. Presented here are the examples of measurements of temporal behaviors of atmospheric air pollution levels in cities and in their outskirts as well as emissions of industrial plants into the atmosphere using optical, including laser, measuring facilities and means for remote sampling placed onboard the small–size remotely controlled vehicles.

This paper is applied in nature and is oriented to the search for the concept of the incorporation of the remote measuring facilities into the network of stations of the atmospheric air monitoring for salvaging our environment. Both stationary and mobile groun–based means are considered.

Monitoring of the polluted atmosphere and emissions in the industrial regions is aimed at promoting information support to control salvaging of environment and ecological safety in cities, industrial centers, and their outskirts. To determine the strategy of such a control, the difference between chemical and photochemical activities of the emitted components should be taken into account. For instance, in urban territories carbon monoxide, carbon dioxide, methane, and freons are relatively conservative, while nonmethane hydrocarbons and nitric axides take part in fast photochemical reactions of formation of oxidants (primarily ozone), acetylnitrate peroxides, and other components which have harmful effect on people, animals, and plants. The control of the conservative pollutants implies taking decision on the reduction of total emissions from the polluting sources. The photochemically active pollutants also require the concentration ratio to be regulated since uncontrollable reduction of the concentration of one of the components can cause undesirable side effects (e.g., in Los Angeles the ozone concentration increased due to reduced emissions of nitric oxides[1]).

The decisions on the changes of the amount and composition of emissions must be made only based on the information about average daily or maximum instantaneous concentration with allowance for the meteorological conditions. The information supplied by monitoring facilities must contain the data obtained in real time needlessly averaged over the period of a month, season, or year. In local measurements in urban territories this is connected with a problem of optimal position of network of the stationary monitoring stations with 1–km minimum separation at the centers of megapolices.[2–4] Such a density of location of traditional stations cannot be achieved in Russian cities within the next decades due to the lack of a domestic base for large–scale production of automatic gas analyzers and means for their metrological support.

The base optical measurement instruments with 100 m–10 km path lengths are capable of fast acquisition (with the time constant varying from 1 s to 10 min) of the representative data due to the effect of spatial averaging. The examined volume of air in the base measurements is 10^4–10^8 times larger than that of a sample in the local analysis on the assumption that the time of measurements with a base instrument and that of sampling with local means are equal. Our results of base measurements of diurnal variations of different pollutants in analogy with the results of other investigations are in a good agreement with meteorological parameters which is indicative of the high degree of representativity of the data on the total content of pollutants obtained along the extended paths. The information of this sort obtained at a few stations (e.g., in Moscow it is sufficient to have five stations) can be used for short–range prediction of hazardous meteorological situations, rational planning of ways of salvaging our environment, and effective control of the results of their implementation.

Our measurements in the territory of the museum–estate "Yasnaya Polyana" (see Ref. 5) and memorial complex "Gorki Leninskie" (see Ref. 6) illustrate the usage of the base instruments for obtaining the information needed for making the decision on changing the amount of emissions.

In both cases a base gas analyzer based on a CO_2 laser recorded the ammonia content continuously with 1 s time constant. Near the first object the services on environmental protection constantly observed destruction of forest canopies caused by high ammonia concentration in the atmosphere. A small number of stations for discrete air sampling under conditions of complex orography give no way of determining the relative contribution of possible sources of ammonia emissions (stock–breeding complexes, metallurgical plant, chemical plant, fertilized fields) to the air pollution in the territory of the estate. The measurements along 500–m path located at 2–m altitude at different wind directions and speed higher than 1 m/s made it possible to determine the principal source of pollution – the chemical plant of the Alkali Production Union (APU) "Azot". Depicted in Fig. 1 are the path–averaged ammonia concentrations at different wind directions.

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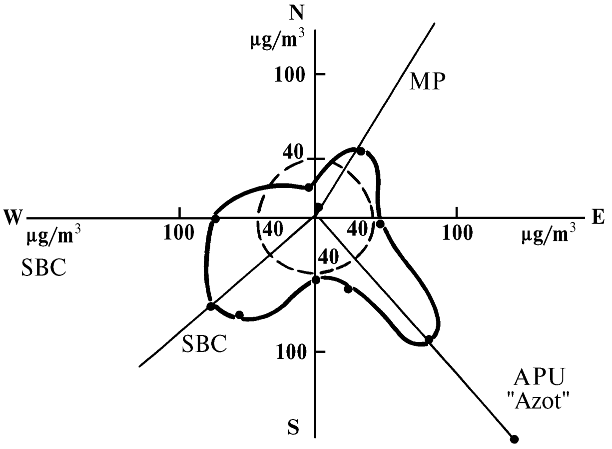

FIG. 1. Distribution of average ammonia concentrations in atmospheric air over the territory of the museum–estate "Yasnaya Polyana" as functions of the wind direction and location of ammonia emission sources. SBC denotes the stock–breeding complex, APU "Azot" denotes the chemical plant, MP denotes the Kosogorsk metallurgical plant, and dotted line indicates the average daily value of the MPC (maximum permissible concentration).

The results were averaged over the samples taken in September–October of 1981 and July–August of 1982. The sources of emissions are easily identified by the wind direction. For South–East winds (from the chemical plant) we observed the atmospheric states with very high ammonia concentrations. One of such situations with concentration significantly exceeding the level of maximum permissible concentration (MPC) (maximum instantaneous concentration was 200 $\mu g/m^3$ = 335 ppb) is shown in Fig. 2. The model calculation was used to estimate 0.5–t amount of emission of ammonia in the atmosphere. The results of our measurements served as the basis for making a decision to change the profile of the APU "Azot". The emission was reduced by the order of magnitude. The stock–breeding complexes were removed and the effect of fertilized fields on canopies of the museum–estate was eliminated.

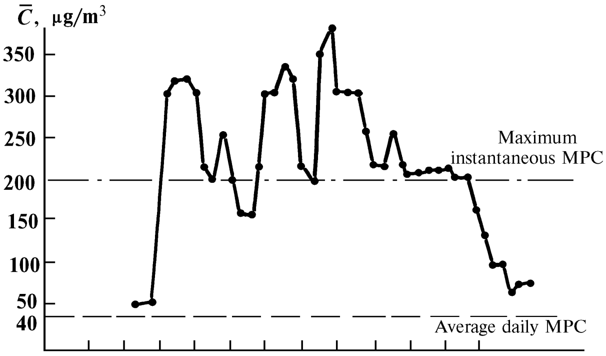

FIG. 2. Temporal dynamics of ammonia concentration in the atmospheric air in the territory of the museum–estate "Yasnaya Polyana".

For another object, the memorial complex "Gorki Leninskie", the oppression of forest canopies was also observed which was caused by the enhanced ammonia concentration in the air. The potential sources were assumed to be the Vidnensk coke–gas plant, the Moscow petroleum–gas plant, and the small dairy farm. The location of the polluting sources is shown in Fig. 3. In the presence of North–West winds the measurements are of a pronounced cyclic character.

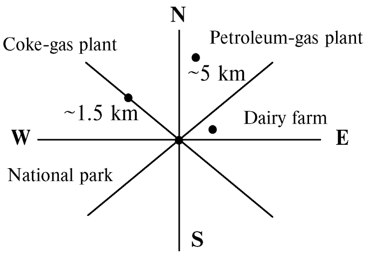

FIG. 3. Location of the sources of emissions in the region of the memorial complex "Gorki Leninskie".

Figure 4 depicts the temporal behavior of ammonia concentration on October 11, 1984 at 5–7 m/s North–West wind. Only the Vidnensk coke–gas plant possessed such cyclicity of technological process in this region. Here the discharge of the prepared coke pig, its quenching, and subsequent charge into the empty cooking cell are performed over 9–12 min period. The same period is pronounced in the concentration behavior shown in Fig. 4. The disturbance of the technological process also affected the concentration behavior depicted in Fig. 4 with allowance for the transit time of a gaseous cloud from the plant to the measurement site (the interval from 15:25 to 15:55). The experimentally estimated individual amounts of emissions were about several kilograms of ammonia per one emission. The services on environmental protection took efforts to modify the technology and to reduce the amount of emissions.

The given examples show that the stationary stations equipped with base instruments can be the efficient means of continuous monitoring of the level of atmospheric air pollution at the most hazardous places near intense sources of emission. The base laser gas analyzers based on dye, CO_2, and semiconductor diode lasers as well as the nonlaser optical base gas analyzers are capable of recording much larger number of ingredients (sulphur dioxide, nitric oxides, carbon monoxide, benzene, toluene, phenol, styrene, xylenes, carbon bisulfide, formaldehyde, hydrogen chloride, hydrogen fluoride, ammonia, and others) than the conventional automatic gas analyzers.

The observations of the general evolution of the processes occurring in the atmosphere of large cities must be carried out in those regions in which the high degree of mixing of the main pollutants, determining chemical and photochemical evolution of the atmosphere, is provided. The observational means must be the systems which can record in real time (with 4–min time constant) the total contents of a large number of ingredients in those air masses which are not affected by the individual weak sources of emissions.

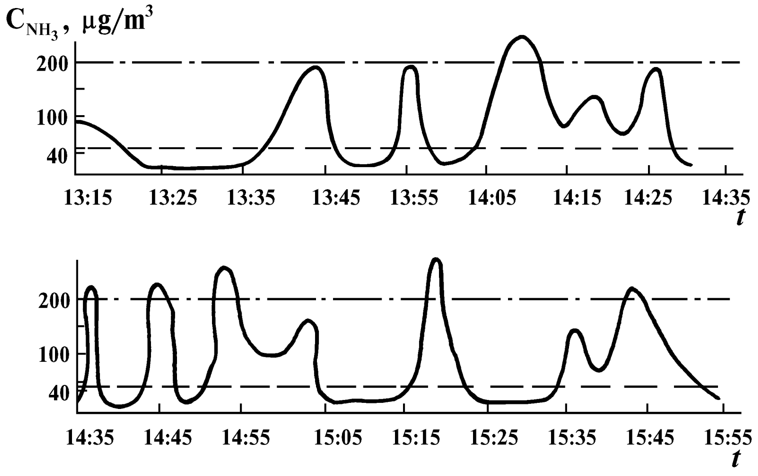

FIG. 4. Temporal dynamics of ammonia concentration (solid line) in the atmospheric air obtained in the region of the memorial complex "Gorki Leninskie". Dot–dash line denotes the instantaneous MPC and dashed line denotes the average daily MPC.

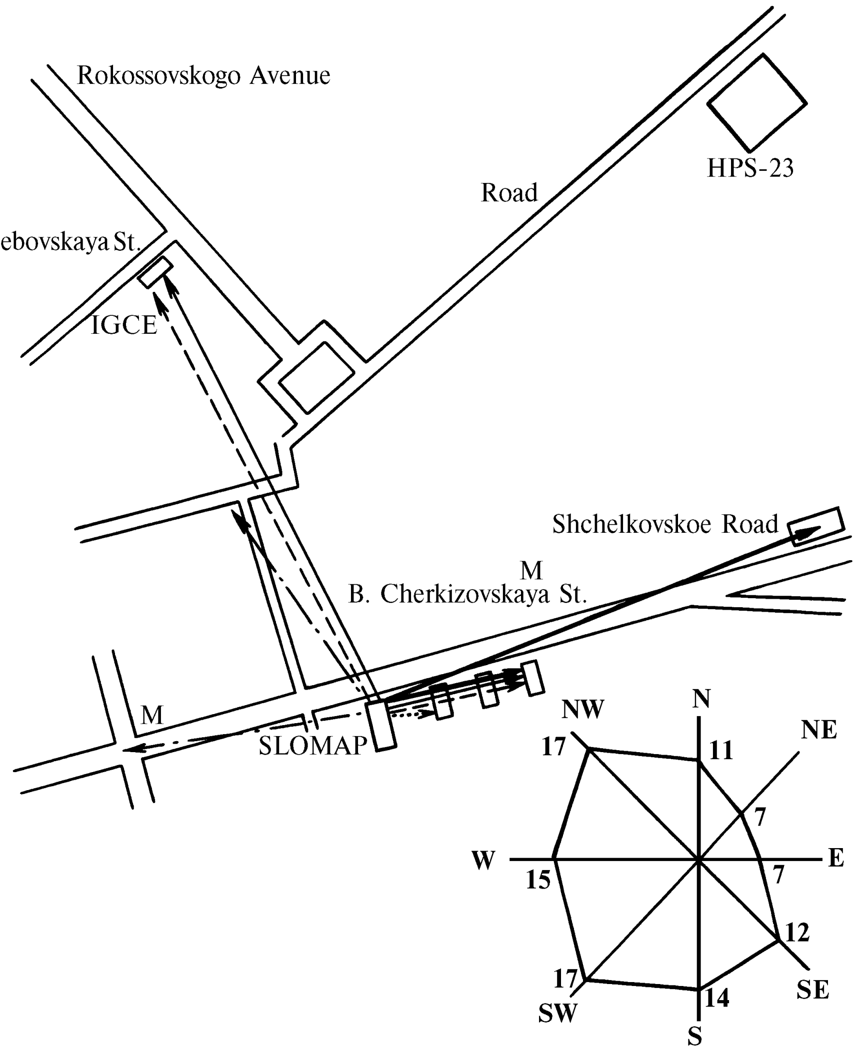

FIG. 5. Scheme of remote laser sounding paths at the station of laser–optical monitoring of air pollution in Moscow and averaging of wind rose for this region. ——→ SO_2, – – → O_3, NH_3, C_2H_4, *and others,* · · · → CO, ——→ NO_2, *and* — · —→ *aerosol.*

A station of laser–optical monitoring of atmospheric pollutions (SLOMAP) in the Kuibyshev district of Moscow was developed at our Institute in 1984. The station was positioned in the attic of 82–m building. It is possible to use the radiation paths in the directions intersecting air flows from differently polluted regions of the city (Fig. 5). The West winds carry out the pollutants from the central regions of the city. The North winds transport relatively clean air masses from Losinyi island. The paths were located at 82–20 m altitudes.

The following facilities were used at the station:

– a lidar base system based on the dye lasers for monitoring of sulphur dioxide,

– a base gas analyzer based on an argon laser for monitoring of nitrogen dioxide,

– several base gas analyzers based on a semiconductor diode laser (SDL) for monitoring of carbon monoxide,

– a base gas analyzer based on the SDL for monitoring of carbon dioxide,

– a single–frequency lidar in combination with an aerosol particle counter,

– base correlation gas analyzers for monitoring of sulphur and nitrogen dioxides,

– a multicomponent base gas analyzer based on a CO_2 laser,

– automatic gas analyzers for monitoring of nitrogen oxides and carbon monoxide, and

– a meteorological station.

The specifications of remote gas analyzers used at the SLOMAP are listed in Table I.

TABLE I. Laser–Optical Station.

Instruments	Ingredient	Wavelength	Path length, m	Sensitivity	Power (pulse energy)
Lidar base system based on a dye laser with reflection from a wall of a building	Sulphuric dioxide	299.40 nm 300.01 nm	600	28 μg/m^3 10–pulse train	10 mJ
Base system based on an argon laser with a corner–cube reflector	Nitrogen dioxide	476.5 nm 496.5 nm 514.5 nm	2000	10 μg/m^3 600–m path	0.4 W 0.2 W 0.9 W
Base system based on a semiconductor diode laser with reflection from a corner–cube reflector	Carbon monoxide	4.7 μm	200	13 μg/m^3	30 μW
Base system based on a CO_2 laser with a diffuse reflector	Ammonia, ozone, ethylene, benzene, freons, perchloroethylen, and others	Emission lines in the 9.2–10.8 μm wavelength range	500	for ammonia 0.76 μg/m^3 for ozone 5 μg/m^3	1 W
Base automated multicomponent system based on a CO_2 laser	10 ingredients simultaneously NH_3, O_3, C_2H_4, C_6H_6, and others.	9.2–10.8 μm	2000	–	0.05 mJ
Aerosol bifrequency lidar: horizontal and vertical paths		0.53 μm 1.06 μm	1500	pulse repetition frequency of 10 Hz resolution of 20 m	20 mJ 50 mJ
Base correlation gas analyzers	Sulphuric dioxide nitrogen dioxide	0.30 μm 0.45 μm	up to 1000	1 ppm·m	–

While working at the station, the diurnal and daytime variations of path–averaged concentrations of sulphuric[7] and nitrogen[8] dioxides, carbon monoxide,[9] carbon dioxide, and aerosol[10] were obtained and test measurements of concentrations of ozone, ammonia, ethylene, benzene, freons, silane, and other components were made.[11]

To detect sulphuric dioxide, we used 600–m path oriented in west–east direction (see Fig. 5) with 82–45 m altitude difference. The reflector was a wall of a building. The maximum in the daytime variation of sulphuric dioxide concentration (Fig. 6) was observed at noon for West wind in sunny weather (with elevated inversion and developed turbulence). This result can be accounted for by the effect of ground–based sources, mainly, motor transport. In the seasonal behavior of daytime averaged measurements of sulphuric dioxide concentrations (Fig. 7) the concentration increased noticeably in October when a heating period started. The maximum permissible concentration was not exceeded with the exception of the case in which 120 ppb concentration was observed when the plume of the heat–and–power station (HPS–23) intersects the measurement path. Also observed was washing out of sulphuric dioxide by rain.

The nitrogen dioxide concentration was measured along 600–m paths (in west–east direction with 82–45–m altitude difference), 1000–m path (in south–north direction with 82–20–m altitude difference), and 2000–m path (in south–north direction with 82–45–m altitude difference). The measurements along 600–m path were carried out over the spring–summer period under similar weather conditions (alternating cloudiness and dry weather) with West, North–West, South, and South–East winds. In the diurnal variations (Fig. 8) regularly self–reproducing maxima are pronounced at 11, 13–14, 15, and 18 h, LT which were also observed in Atlanta, USA[12] under similar weather conditions and in the measurements with the base correlation gas–analyzer in Moscow.[13] These maxima can be attributed to the amount of city motor–transport traffic and evolution of photochemical processes in the atmosphere. Measurements along 1 and 2–km paths were made in different seasons. In winter no regular daytime maxima were observed. It was typical that the nitrogen dioxide concentration increased gradually, reached its maximum at about 16 h, and decreased at nighttime. Frequent and multiple excess of the MPC was observed in any season of the year. In some cases nitrogen dioxide was washed out from the atmospheric air by rain and snow. In the joint measurements with the sodar of the Institute of Atmospheric Physics of the Academy of Sciences of the USSR (IAP sodar) we observed correlation between the nitrogen–dioxide concentration decrease and the mixing–layer height increase at about 12 h.

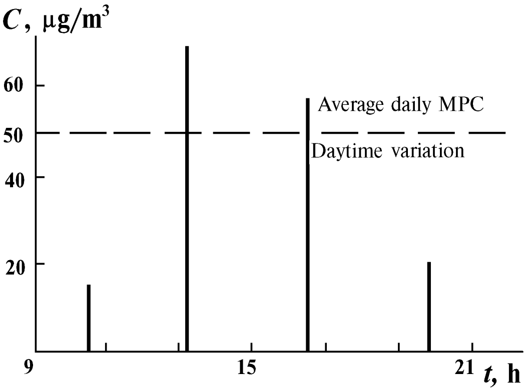

FIG. 6. Daytime variations of sulphur dioxide concentration in atmospheric air along the path shown in Fig. 5.

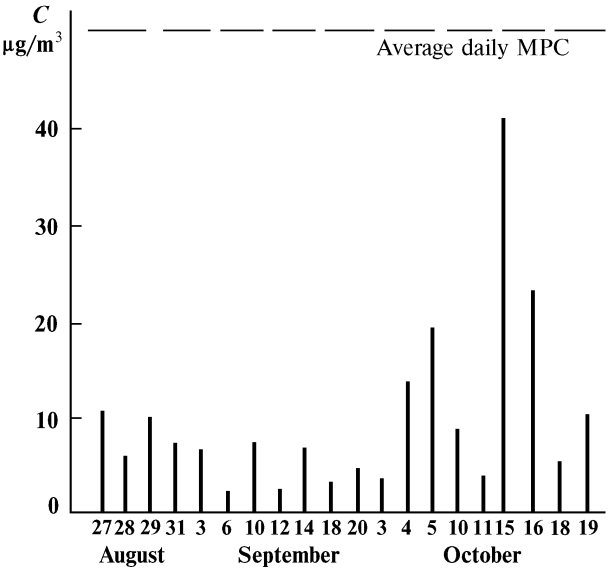

FIG. 7. Seasonal trend of sulphur dioxide concentration in atmospheric air. Vertical lines indicate daytime averaged measurements.

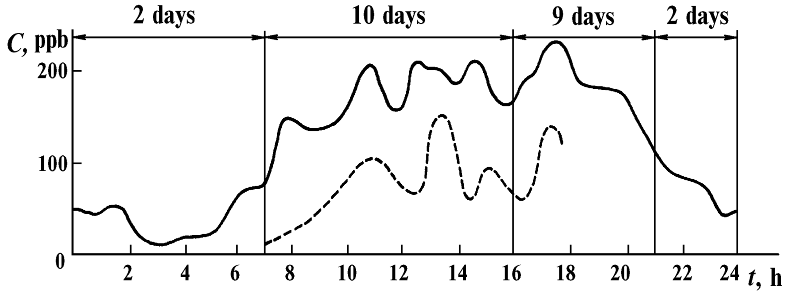

FIG. 8. Average diurnal variation of nitrogen dioxide concentration at West winds over the spring–summer period. Solid line is for Moscow in May–July of 1986, and dashed line is for Atlanta, USA, on October 27, 1972.

In the measurements of carbon monoxide with the help of the AGL–02 gas analyzer the correlation with the amount of the motor–transport traffic, wind direction, and atmospheric stratification was observed. It was also noticed that carbon monoxide was accumulated near the base of the inversion layer (Fig. 9). The MPC was not exceeded. Based on the detected increase of the variance of the results with increase of the concentration, the method was developed of determining the regional background concentration, i.e., such a concentration for which the variance of an individual measurement (a train of 120 pulses) vanishes. To the north–east of Moscow this value for nitrogen monoxide was 0.2 ppm (Fig. 10).

As a whole, the measurements at altitudes of from 10 to 100 m along the paths located above the buildings determine mainly the pollution levels of air masses transported to the examined region from outside as well as the contribution from high local sources and from pollutant accumulated under the inversion layer. The long–term regular measurements make it possible to estimate the ecological reserve of the territory under study which is determined as difference between the MPC values and the regional background pollution of the territory. The latter is estimated based on minimum values of concentrations in long–term runs of observation performed at fixed directions of wind. For example, for carbon oxide the ecological reserve of the SLOMAP–monitored territory has not been exhausted at any wind directions (Fig. 11), while for nitrogen dioxide there is no ecological reserve.

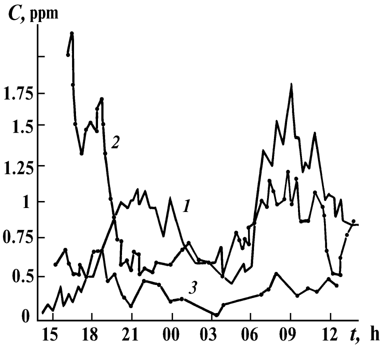

FIG. 9. Diurnal variation of carbon dioxide concentration: 1) elevated inversion for West and South–West wind, 2) elevated inversion in calm weather, and 3) North–West wind.

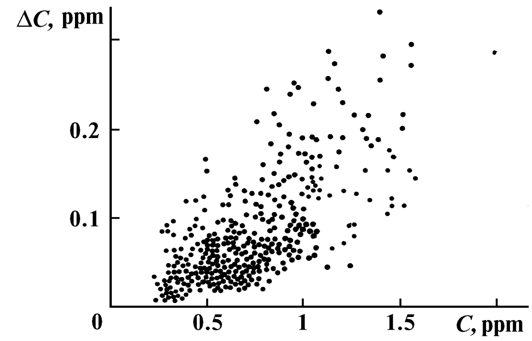

FIG. 10. Variance of the results of measurement of carbon dioxide as a function of the average concentration.

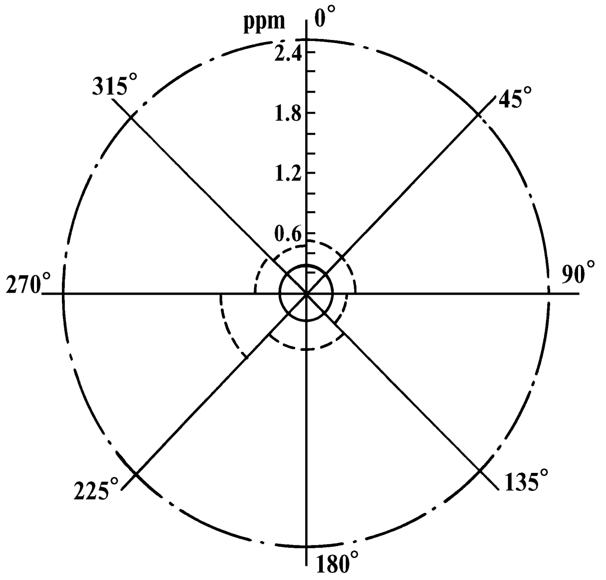

FIG. 11. Ecological reserve of carbon dioxide of the atmospheric basin of the region: solid line indicates the regional background concentration, dashed line indicates the average carbon dioxide concentration in downwind direction in ppm, and dot–dashed line indicates the average daily values of the MPC.

The integral information yielded by the stationary systems can be supplemented by measurements made with mobile stations equipped with passive correlation instruments (based on a monochromator with mask and interference–polarization or gaseous filter), standard automatic gas analyzers, sampling means, and simple means of elevation of the portable samplers at different altitudes [e.g., using a small–sized remotely controlled vehicle (SRCV)] as well as various lidars, in some cases.

Passive correlation instruments recording the absorption of sky scattered solar radiation in the polluted air enable one, under the appropriate meteorological conditions, to carry out the most reliable (compared to any contemporary devices) measurements of fluxes of the pollutants passing through the surfaces formed by scanning line of sight of such a device. The measurement in the fixed direction of the line of sight yields the total content of polluting substances along this line. Integration over the scanning path (due to the rotation of the device or its transportation through the polluted area) yields the total content of the detectable component on the surface formed by moving line of sight. If this surface is a plane perpendicular to the axis of a plume from the emission source, then multiplication by the wind speed averaged over the plume cross section yields the flux of the pollutant from the source or its emission. Vertical sounding when moving round the polluted area allows one to determine the flux out of this territory. The flux can also be determined using a radio–controlled vehicle with onboard samplers flying across the flux section or by scanning with a lidar.

We have our own experience in sounding of the plumes of power stations and industrial plants using an automotive correlation passive gas–filter radiometer for measuring sulphuric dioxide,[14] aerosol lidar,[15] and SRCV (for measuring dioxides of sulphur and nitrogen, sulphides and sulphates, and nitrites and nitrates) by means of air sampling onto paper filters.[16] We also measured vertical profiles of ammonia[17] and methane by taking samples into injectors from aboard of the SRCV and their subsequent analysis by the laser opto–acoustic method.

A gas–filter correlation radiometer was used for measuring the emissions of sulphuric dioxide from the state regional power stations (SRPS's) in European part of the USSR and heat–and–power stations (HPS's) in Moscow and Erevan (Fig. 12). Large difference in the amounts of emissions is caused by different source strengths and types of fuel (coal, fuel oil, and gas). The accuracy of a single measurement was about 30 %. The accuracy can be increased by increasing the number of scannings. The time of single scanning at a distance of 1 km from stacks was about 1 min. The main drawbacks of passive correlation instruments operating by the aforementioned methods are limited time of a day and season suitable for conducting the measurements and a limited number of detectable components. The operation time is limited by the sun height above the horizon at which the sky–scattered radiation intensity is still sufficient. Depicted in Fig. 13 are the operation times of the passive instrument for which the solar zenith angle must be 30°. The time in hours counted off from the local noon was calculated for different latitudes (40, 50, 60, 70, and 80°). For example, for 50° latitude on June 1 we can operate in the ± 4.6 h interval counted off from local noon (the point A), while the operation season for 50° latitude lasts from February 23 to October 19. The dates of beginning and termination of the operation season are indicated near the curve for different latitudes. At the beginning and termination of the season only a short–time operation about the noon is possible. The operation of passive instruments is also impeded by continuous cloudiness or by cumulus clouds passing through the field of view of the instrument.

To date the passive correlation instruments have been used only for detecting dioxide of sulphur and nitrogen. Some other components, e.g., formaldehydes and hydrogen sulphide, whose absorption bands lie in the wavelength range 0.3–1.0 μm, can also be detected. The number of detectable ingredients with simultaneous determination of their fluxes from the sources can be substantially increased due to combined measurements with the correlation instrument and SRCV for taking samples in the plume. In the laboratory analysis of the selected samples it is sufficient to determine the ratio of contents of one of the gases being detected with the correlation instrument in the plume to other ingredients of this plume and then to use the proportionality between the fluxes and contents.

The sampling from onboard the SRCV at different altitudes makes it possible to determine the vertical profile of pollutants transported from the polluting sources. This enables one to determine the removal of pollutants from air masses in the process of their far transport. The maximum altitudes at which it is possible to take samples is 2 km in the daytime and 0.5 km at nighttime. The laser opto–acoustic gas analyzer in the mobile laboratory is capable of analyzing the samples taken into injectors in real time at the altitude of sampling that is very important for unstable substances. The volume of the air in the sample required for the analysis is 8 cm^3. A sensitivity of $(2–5)\cdot 10^{-8}$ $cm^{-1}\cdot W$ was provided by a CO_2 laser delivering 0.5–1 W which was used as a radiation source. The minimum detectable concentrations are 0.5 for ammonia, 1 for ethylene, and 3 ppb for ozone. The time of analysis is 4 min. Not more than 20 min is required for setting the SRCV for flight including installation of the sampler, assembly, and refuelling. The time of sampling into injectors is 1 s, the number of transported injectors is nine. A free space within 100 m and an even 20–25 m runway with any type of covering except water, are needed for take–off and landing.

The mobile system in combination with SRCV and opto–acoustic gas analyzer was used for studying the transport of ammonia from agricultural sources (stock–breeding complexes and fertilized fields) and ammonia absorption by agricultural crops. Vertical profiles of ammonia concentrations were determined at six levels within the 40 m–1 km altitude range in fine weather or at the cloud base (300–400 m) in cloudy weather. It was found that within the mixing layer (at altitudes up to 1–1.5 km) there was 200–m layer at the altitude 1/2 H through which ammonia was transported from the nearest sources. The distribution of ammonia concentration over stalks of different agricultural crops was studied by manual air sampling into injectors and absorption of ammonia contained in soil and air by plants was quantitatively estimated

The salient features of ammonia absorption by plants[18] (linearity of absorption of physiologically active radiation (PAR), the absence of dependence of the absorption rate on concentration, etc.) as well as the results of investigations of the spread of air pollutants in plant canopy[19] allowed us to derive an analytical expression for estimating the rate of ammonia dry sedimentation from air onto the plants[20]

$$V_d = \sqrt{K_H s} \cdot (A + BP) ,$$

where K [$cm^2\cdot s^{-1}$] is the turbulent mixing coefficient at the top of the plant canopy, s [cm^{-1}] is the vegetative cover index (ratio of the area of leaf laminae to the volume occupied by plants), P is the PAR intensity [$J\cdot cm^{-2}\cdot h^{-1}$].

The expression $(A + BP)$ has dimensionality [$cm^{1/2}\cdot s^{-1/2}$], the values of the coefficients vary depending on the type of plants. Using this expression for determining V_d, it is possible to estimate the ammonia nitrogen loading factor for the given vegetation by measuring the ammonia concentration at the top of the plant canopy with the base gas analyzer. The same approach can also be used for other physiologically active gases (NO_2, SO_2, etc).

Joint application of stationary and mobile systems in the city will provide the detection of harmful levels of atmospheric air pollution in real time both over large areas of a regional scale within the 10–100 m altitude range as well as near the sources of pollutants with independent recording of their surface levels and total amounts transported by air mass flows. It will also enable one to determine the vertical profiles of concentrations using both the SRCV and lidars. Recording of enhanced levels of pollution in cities with the use of stationary complexes will allow us to monitor the spread of polluted air masses to rural areas and the evolution of these air masses during transportation with the mobile stations.

Thus a complex of three elements: (1) stationary special–purpose stations near especially harmful sources of emissions, (2) stationary complexes intended for observations of total contents of a wide variety of components, and (3) mobile complexes intended for determining fluxes and spatial distributions, in particular, vertical profiles of concentrations will allow one to solve the following problems of monitoring of atmospheric pollutants in the urban territory:

– detection of concentration levels exceeding the maximum permissible norms for the pollutants and identification of sources of polluting substance emission;

– monitoring of dynamics of composition of the urban atmosphere and that of individual city regions to elucidate physicochemical and photochemical nature of processes which determine the evolution of the atmosphere under different meteorological conditions to choose optimal strategy of salvaging our environments, to control its efficiency, and to provide a short–period prediction of air pollution;

– determination of a polluting substance flux from the emission sources in individual regions of a city and in a city as a whole to find relative contributions of emission sources to the total pollution of the urban atmosphere as well as to find that portion of emissions which is further involved into the far transport;

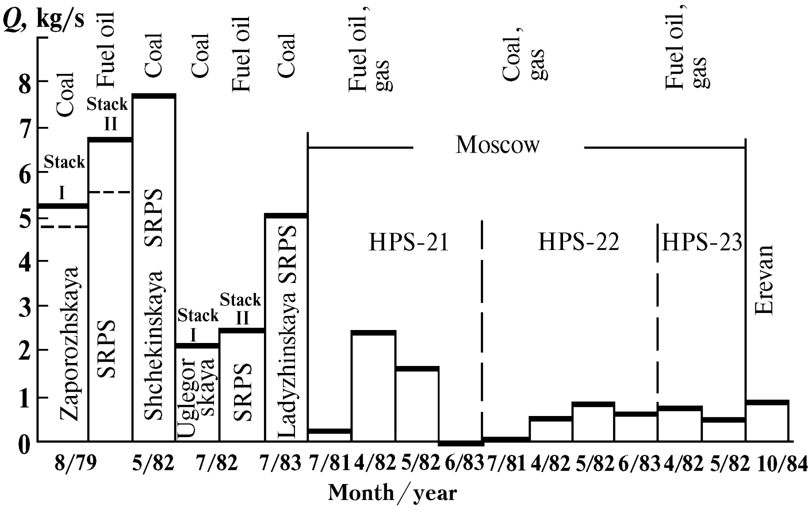

FIG. 12. Results of an inventory of sulphurous gas emissions from different heat–and–power and town district power stations (HPS and SRPS) with correlation radiometer.

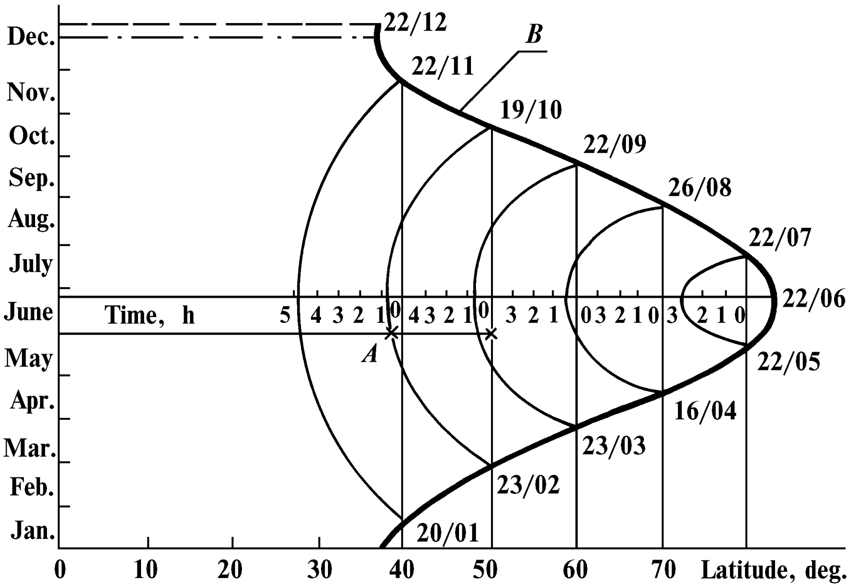

FIG. 13. Operation time of the correlation nondispersion radiometer used for measuring the sulphurous gas concentration.

– measurement of vertical profiles of pollutants in air masses carried out from megapolices to determine the coefficients of removal of the pollutants in the process of their far transportation and loading factors for pollutants and to refine the models of physicochemical evolution of the urban atmosphere.

A set of the data obtained will provide the solution of scientific problems on refined determination of balance of carbon, nitrogen, sulphur, and other elements in global cycles with allowance for their removal from the urban areas and on the estimation of anthropogenic effect on the global content of greenhouse gases in the atmosphere.

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