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Integrated Studies of Tropospheric Aerosol at the Institute of Atmospheric Optics (Development Stages)

M. V. Panchenko[a, *], M. V. Kabanov[b], Yu. A. Pkhalagov[a, †], B. D. Belan[a], V. S. Kozlov[a], S. M. Sakerin[a], D. M. Kabanov[a], V. N. Uzhegov[a], N. N. Shchelkanov[a], V. V. Polkin[a], S. A. Terpugova[a], G. N. Tolmachev[a], E. P. Yausheva[a], M. Yu. Arshinov[a], D. V. Simonenkov[a], V. P. Shmargunov[a], D. G. Chernov[a], Yu. S. Turchinovich[a], Vas. V. Pol'kin[a], T. B. Zhuravleva[a], I. M. Nasrtdinov[a], and P. N. Zenkova[a]

[a] *V.E. Zuev Institute of Atmospheric Optics, Siberian Branch, Russian Academy of Sciences, Tomsk, 634055 Russia*

[b] *Institute of Monitoring Climatic and Ecological Systems, Siberian Branch, Russian Academy of Sciences, Tomsk, 634055 Russia*

**e-mail: pmv@iao.ru*

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Abstract—In the work dedicated to the 50th anniversary of the V.E. Zuev Institute of Atmospheric Optics, Siberian Branch, Russian Academy of Sciences (IAO SB RAS), experimental studies are reviewed in a certain chronology that have been milestones in the development of an integrated study of aerosol life in the atmosphere. We discuss briefly the main results from the series of expedition studies of the optical and microphysical properties of aerosol of marine coastal hazes, arid zones, and different geographic regions of the World Ocean. The modern set of methods and instrumentation that we use to measure the aerosol characteristics in the monitoring mode at the network of IAO SB RAS stations is described. The results of multiyear studies of tropospheric aerosol using aircraft laboratories are presented.

Keywords: aerosol, optical and microphysical characteristics, scattering, extinction, and absorption coefficients, spectral transparency, aerosol optical depth

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INTRODUCTION

Even at the creation of the Institute of Atmospheric Optics, V.E. Zuev laid down as a basic principle the integrated approach to solving atmospheric optics problems, consisting of optimal combination of comprehensive experimental study, the development of theoretical fundamentals, and multifactor simulation [1]. It is very difficult to select a subject of study, analogous to the atmospheric aerosol, whose leading parameter (size) varies by a few orders of magnitude (from tens of angstroms to hundreds of microns) and whose physicochemical properties are formed under the influence of many processes from different sources. To simulate and predict the optical state of aerosol under specific conditions, it is necessary to account for the spatial coordinates that vary from centimeters to kilometers in the vertical, and from meters to thousands of kilometers in the horizontal. From the evidence above it is clear that only an integrated approach makes it possible to ensure the required development of the instrumental basis and to obtain new scientifically justified results.

We note that national scientists made an enormous contribution to world aerosol science in the second half of the twentieth century owing to large-scale studies in the Institute of Atmospheric Physics (IAP), the Institute of Atmospheric Optics (IAO), the Institute of Experimental Medicine (IEM), the Central Aerological Observatory (CAO), the Institute of Applied Geophysics (IAG), Leningrad State University (LGU), the Main Geophysical Observatory (MGO), the State Optical Institute (SOI), the State Institute of Applied Optics (SIAO), the Institute of Physics of the Academy of Sciences (IP AS) of Belarus, the Astrophysical Institute of the National Academy of Sciences (API NAS) of Kazakhstan and many other organizations. Even a comprehensive review fails to list all the experimental and theoretical work performed in this period (see, e.g., [1–18]).

However, even today, the International Panel on Climate Change (IPCC) experts note a low level of knowledge on aerosol contribution to the Earth's radiation budget [19], underlining acute need for detailed instrumented studies in all Earth's natural zones on different (global–regional–local) scales.

† Deceased.

Guided by the specific features of the journal's number devoted to the 50-year anniversary of the Zuev IAO SB RAS, the paper overviews in a certain chronology the research that, in our opinion, determined milestones in the development of the integrated study of "aerosol life" in the atmosphere.

INTEGRATED AEROSOL STUDIES IN DIFFERENT GEOGRAPHIC REGIONS IN 1970–1995

Coastal hazes. Choice of the research subject was dictated by the following considerations: by the beginning of our measurements, the optical characteristics of near-ground aerosol under continental conditions had already been quite deeply studied by the group headed by G.V. Rosenberg (see, e.g., [5]); while the marine and coastal regions had been much less comprehensively studied in this regard.

The first cycle of works that involved almost the entire range of instruments developed in IAO was performed in the coastal zone of the Black Sea from 1973 to 1981.

For the first time in the practice of studying the optical properties of coastal haze, the spectral coefficients of total radiation attenuation were measured in the wavelength range of 0.45–12 μm, as well as polarization scattering phase functions in the visible wavelength range [20–23]. These data were used to create one-parameter models of the aerosol extinction coefficients [21] and angular characteristics of scattering for the visible wavelength range [22].

Arid zone. In 1984–1988, a cycle of integrated research into the optical and microphysical characteristics of aerosol was performed in the near-ground layer of the atmosphere of the arid zone of Kazakhstan for three seasons of the year: spring, summer, and fall. The seasonal behaviors of submicron $\alpha_f(\lambda)$ and coarse $\alpha_c(\lambda)$ components of the aerosol extinction coefficient are found to be well pronounced in the arid zone in the wavelength range $\lambda = 0.44–11.5$ μm [24]: maximal (minimal) $\alpha_f(\lambda)$ values are observed in spring (summer). The average values of aerosol extinction coefficients in the arid zone in the absence of dust storms are shown to be approximately 2–5 times less than in other climatic zones. The measurements were used to develop a two-parameter model for calculating the aerosol extinction in the wavelength region $\lambda = 0.4–12$ μm; the model input parameters are the extinction coefficients at the wavelengths of 0.48 and 0.69 μm [25]. Comparison of extinction coefficients $\alpha(\lambda)$ on extended paths with measurements in local volumes (aureole scattering phase function and aerosol particle size distribution function) showed that their values can be used as input parameters of empirical models [26], which is important for operationally assessing the optical state of the atmosphere.

Active spectronephelometry in integrated studies. The creation of few-parameter models of optical characteristics and their microphysical interpretation has made it possible to identify the basic laws of variations in characteristics which, in Rosenberg's precise figure of speech "are stored by the atmosphere" among a large number of diverse optical-meteorological situations occurring in the near-ground layer. The understanding, gained at this stage, has determined the direction of further development of our work on studying the aerosol life and creating dynamic empirical models. Rephrasing an accurate thought of Rosenberg, we stress that "...the main permanent property of atmospheric aerosol is its permanent variations in space and time" [5]. Hence, even statistically representative average (optical or microphysical) characteristics have a limited applicability range for solving practical problems, suggesting that an empirical model should have measurable or predictable input parameters.

The microphysical composition of particles in a specific atmospheric situation is influenced by many internal aerosol and external environmental processes. In turn, these processes have their own spatiotemporal rhythms and a certain interrelation; hence, a small number of measured characteristics, which show a different character of time variability, and in which the total response of all processes shows up most strongly, can be identified as input parameters. The basic microphysical composition of submicron particles (their microstructure and optical constants, henceforth the "dry base" of aerosol), observed at a specific time, is formed for a relatively extended period of time and depends on the lifetime and trajectory of the motion of an air mass. Among the external factors that determine rapid transformation of particle microphysical parameters, a leading role is played by the relative air humidity, the value of which is not related directly to microphysical characteristics of aerosol particles within a specific air mass, and the variations of which are regulated by relatively transient processes. We note that the condensation activity of aerosol is determined by the relationship between soluble and insoluble substances and their chemical composition in dry basis of aerosol; therefore, in the general case it depends indirectly on the processes that determine the particle microphysical composition. Based on these views on characteristic (spatiotemporal) aerosol variability scales, characteristics of "dry aerosol" and its condensation activity for an artificial controllable change in relative air humidity should be studied separately in addition to in situ observations [27].

STUDIES OF TROPOSPHERIC AEROSOL (AIRCRAFT LABORATORY) 1985–1995

In formulating the plan of a new series of experimental works, we were guided by the following considerations: studying regional-scale phenomena requires either multipoint measurements or the use of highly

mobile means capable of covering vast territories in a limited time. Since the processes of transformation of characteristics of aerosol particles cannot be understood without knowing their spatiotemporal variations (as regards stable air masses, their vertical variations come into foreground), the problem of creating an aircraft laboratory was posed and solved.

In mid-1970s, the aircraft sensing was episodic in character and mainly aimed at developing onboard means of measuring the atmospheric parameters and improving the methods for sensing the characteristics of the air and the underlying surface. However, in 1985, we built a set of instruments for the aircraft laboratory comprising meteorological and microphysical measurement complexes, an active nephelometer with thermo- and hygrooptics [28], a spectrophotometric complex, and an onboard recording system. As a consequence, from 1985 to 1991 the aircraft sensing was performed in almost all physical-geographic and administrative regions of the former Soviet Union [28].

ODAEKS-87 experiment. In collaboration with the IAP of the USSR Academy of Sciences, in the coastal region of Odessa we performed an integrated experiment with the participation of the aircraft laboratory of the IAO and a large ground-based complex of instruments. The ground-based complex comprised two nephelometric installations, an instrument for measuring the spectral optical depth of the atmosphere, an installation for measuring scattering phase functions of brightness in the region of circumsolar halo (all owned by the IAP), a nephelometric installation with thermo- and hygrooptical devices, halo laser nephelometer, aerosol particle counter, aspiration installation for collecting aerosol samples on filters for chemical analysis, an instrument for measuring the horizontal spectral transparency (IAO), as well as a complete suite of instruments at the experimental meteorological proving ground of the basic scientific-research laboratory at the Odessa Hydrometeorological Institute (OHMI, Odessa). This research revealed for the first time the differences in thermo- and hygrooptical characteristics of aerosol for different air masses, detected an increase in the parameter of condensation activity with height, analyzed intraseasonal variability cycles of scattering coefficients and concentrations of particles in different size ranges [29].

DUNA experiment. Properties of dust aerosol from the deserts of Central Asia were studied by carrying out a preparatory experiment in fall 1988 and the DUNA Soviet-American experiment in September 1989, which was implemented through the participation of many research groups from leading organizations in the Soviet Union and United States. The aircraft laboratory of the IAO performed sensing of the vertical profiles and spatial distribution of the optical-microphysical characteristics of aerosol, its chemical composition, as well as the meteorological parameters of the atmosphere in the study area with recurrent control landings (or their imitations) in the airports of Dushanbe, Kurgan-Tube, and Termez. Separate patrol transects (for one-two altitudes) along the paths Dushanbe–Tashkent–Chimkent–Dushanbe, Dushanbe–Chardjou–Bukhara–Dushanbe were carried out to estimate the general optical state of the atmosphere in the region and the spatial scales of the dust storms. In this experiment we obtained a large amount of information on microphysical and chemical composition of dust particles, as well as on the dynamics of their dispersal in space, which made it possible to study in detail and for the first time the vertical profiles of the optical and microphysical characteristics of aerosol in the altitude range from 0 to 6 km for dust storms in Central Asia [30, 31].

An extensive dataset, obtained in 1985–1988 from regular aircraft sensing of optical and microphysical characteristics of aerosol in the atmosphere of the western Siberian region (over 600 vertical profiles, measured in all seasons of the year in diverse meteorological and synoptic situations), served as a basis for creation of the first version of the empirical regional model of aerosol optical characteristics [32, 33].

Atlantic-88 expedition. The ship-based expedition to study near-water aerosol using active spectronephelometry, the first in our practice, was carried out from March to June 1988 in the Atlantic Ocean with a wide coverage of different geographic zones (from 25° to 68° N). We were first to show that thermo- and hygrooptic characteristics and microstructure of aerosol in open ocean conditions in the absence of a marked continental effect are similar for different climatic zones [34, 35].

Ecology 1988–1995 cycle. Onboard the An-30 Optik-E aircraft laboratory of the IAO we performed a large cycle of integrated ecological investigations of the territories of the Kamchatka Peninsula, Baikal region and Lake Baikal, Buryatia, Megion and Samotlor fields, as well as an assessment of the ecologic states of air basins of Nizhny Tagil, Nizhnevartovsk, Pavlodar, Ust-Kamenogorsk, Amursk, Khabarovsk, Komsomolsk-on-Amur, Ulan-Ude, and Kemerovo [36].

AT THE TURN OF THE CENTURY

In 1990s, our Institute, like all domestic science, was in a very difficult situation. However, the undoubted international authority and titanic efforts of our leader, Academician V.E. Zuev, toward saving the IAO SB RAS allowed us to survive this period. It should be recalled that we were the first among foreign participants to receive support from the project (leaders V.E. Zuev and G.A. Titov) in the new fundamental Atmospheric Radiation Measurement (ARM, Pacific Northwest National Laboratory, United States, https://www.arm.gov) program. Many participants of annual ARM team meetings and presentation of results on the entire spectrum of IAO SB RAS activities (e.g., [37]) have determined our continued partic-

ipation in the integrated project, implemented in collaboration with IAP RAS under the leadership of Academician G.S. Golitsyn.

A very important role in this period had been played by decisions of the SB RAS leaders: Academician V.A. Koptyug, who managed to support a number of aircraft experiments in the most difficult years; Academician N.L. Dobretsov, who initiated the decision on regular support of expeditions (primarily those dealing with a continuation of long observation time series), as well as the operation of stationary sites and observatories. The formation of the Russian Fund for Basic Research (RFBR) in 1992 had been one of the key factors ensuring the existence of operable scientific groups, maintaining and, then, developing their material and technical basis. For instance, the project "Aircraft study of the effect of anthropogenic sources and forest fires on climatic and ecological state of the region of Lake Baikal" was implemented in 1995, which could barely be possible without the support under RFBR grant [38]. The availability of the operational aircraft laboratory has determined the conclusion in 1997 of a perpetual agreement with the National Institute for Environmental Studies (Japan, http://www.nies.go.jp) on organizing regular sensing of the troposphere in western Siberia in the framework of a division of the International Geosphere-Biosphere Program headed by Gen Inoue. This event can hardly be overestimated because, since then to the present, only our aircraft laboratory is regularly sensing the tropospheric composition in Russia [39]. Of special note is that long-term aircraft studies of the biological component of the tropospheric aerosol were initiated in 1998 in collaboration with the Vector State Research Center of Virology and Biotechnology [40], and to date a unique set of data and results has been obtained, which does not have analogs in world practice [41].

In those difficult years the participation of the Institute in implementation of the programs of the Presidium of the RAS, integration, and multidisciplinary projects of the SB RAS, and then the emergence of a special program for import substitution and a competitive system of acquiring costly instrumentation from budgets of the SB RAS and RFBR presented an opportunity for gradually developing and broadening the instrument base.

Analyzing the whole the second half of the twentieth century, we note that the advent of lasers, the rapid development of various systems and devices operating in the optical range through the atmosphere (sensing, ranging, navigation, observations of Earth from space, etc.) in the world scientific community initiated large-scale aerosol studies. At that time, researchers had accumulated extensive experimental material that laid the foundation for a number of models of atmospheric optical characteristics, well known to specialists (see, e.g., [42–49]). At this stage, the main efforts of scientists were concentrated on studying the scattering properties of atmospheric particles, because the spectral ranges of instruments were selected to lie outside the absorption lines of atmospheric gases. Much less information at that time was obtained about the aerosol absorption, primarily because of the lack of the relevant instruments. However, as early as the end of the century, in regard to the observed climate change, the world scientific community had been aware of the pressing need for a detailed study of the scattering and absorbing properties of atmospheric aerosol for calculating the radiative properties of the atmosphere and underlying surface (an especially urgent problem for Arctic and Antarctic regions). It is this circumstance that determined the intense development of the instrument base and the beginning of a new cycle of large-scale integrated experiments and routine observations in the world scientific community.

MODERN STAGE (COMPLEX MONITORING AND EXPEDITION STUDIES IN POLAR AND MARINE REGIONS)

The accuracy of accounting for the main optical characteristics of aerosol and parametrizing their variations under the influence of an intricate combination of synoptic and meteorological processes required to solve the radiation problems cannot be achieved only on the basis of a limited series of observations. Even within a separate region, the creation of a branched network of stations, where all the required complex of aerosol characteristics would be observed in a monitoring mode, is an unachievable task for reasons economic and technical in character.

We developed the following approach to organizing research to study the basic physical processes and take into to account the effect of geophysical factors that determine the "aerosol weather" [50]. It envisages a small number of stations, operating in a research monitoring mode, combined with large complex experiments in which the maximum available number of systems is concentrated, providing measurements of the most climatically significant parameters of the atmosphere [51, 52]. An integral part of this is regular expedition measurements using mobile systems based on an aircraft laboratory and research vessels.

We note that the term "research" monitoring is taken to mean a permanently developed system of routine automated measurements of the aerosol optical and microphysical characteristics. Important tasks of research monitoring include: the regular processing of the entire complex of data, retrieval of the total set of the aerosol optical characteristics in situ and their comparison with results obtained directly in the atmosphere on horizontal and slant paths. On this basis we develop new methods, implement them instrumentally, and integrate them into the measurement complex.

THE NEAR-GROUND ATMOSPHERIC LAYER

In 1992, we initiated measurements of the complex aerosol characteristics in the near-ground atmosphere at our first Tropospheric Ozone Research (TOR) monitoring station in Tomsk Akademgorodok (http://lop.iao.ru/RU/tor/) [53]. Currently, the station carries out measurements of the concentrations and distribution functions of particles in two size ranges: 3–200 nm and 0.25–32 μm. The aerosol station (http://aerosol.iao.ru) began operating in Akademgorodok in 1997. Scattering coefficients, the mass concentration of soot in aerosol, and the mass concentration of absorbing material (henceforth black carbon, or M_{BCeq}) are automatically recorded every hour. We explain that the instrument for measuring the absorbing material is calibrated using soot particles and measurements are reduced to the equivalent of the black carbon mass concentration M_{BCeq}(μg/m^3). A cycle of measurements is performed every day at the station, using an active spectronephelometry installation [27] for determining the parameter of hygroscopic particle activity and the fractional content of volatile compounds in the aerosol. Measurements of the extinction coefficients on the open horizontal path in the wavelength region $\lambda = 0.45–3.9$ μm began at the station in 2008; and routine recording of the scattering coefficients in the halo region in the angular range $\varphi = 1.2°–20°$ became operational in 2010 [54]. It should be specially noted that the soot diffusion spectrometer, developed and created in collaboration with the Institute of Chemical Kinetics and Combustion (ICKC) SB RAS, for the first time in the world has provided monitoring measurements of the distribution of absorbing material (M_{BCeq}) by size in submicron aerosol [55]. In 2013, the soot diffusion spectrometer was installed at Aerosol Station; and now every 2 h it is automatically measuring the soot distribution over sizes in the size range 10–1000 nm in the near-ground air layer.

To determine the regional background state of aerosol characteristics, since 1995, in different periods of the year, we performed series of synchronous measurements in Akademgorodok and at the background proving ground of IAO SB RAS, located 60 km southwest of Tomsk. Taking into account the pressing need for continuous monitoring under background conditions, as the material base develops, in 2008 the Fonovaya Observatory was organized at this site, where the distribution function of particles over size is measured in a wide size range. Instruments for recording the scattering coefficients and mass concentration of soot were introduced into the monitoring mode in 2014. We note that southwesterly and westerly winds predominate in the region of Tomsk; thus, air masses first pass the Fonovaya Observatory, and then Tomsk and Akademgorodok. The first cycles of observations (1995–2005) showed that anthropogenic urban sources have an insignificant effect on the state of near-ground aerosol. Based on data analysis, we draw the conclusion on the predominant effect of mesoscale processes on aerosol weather formation in the near-ground atmospheric layer on a regional scale [56], where the prehistory of air masses is a determining factor of variations in aerosol characteristics.

A comparative analysis of data of simultaneous measurements of aerosol optical depth (AOD) in Akademgorodok and in the background region in 1997–2008 [57] showed that the variations in characteristics of atmospheric turbidity are mainly caused by synoptic processes. It was found that AOD values in the two regions are statistically indistinguishable in the summer period; while in the cold period a higher atmospheric turbidity was observed in the suburban zone in separate cases, seemingly because of accumulation of aerosol particles below the near-ground temperature inversion and the spread of the urban pollution "cap." Comparisons of continuing cycles of synchronous observations at the two sites indicated that no significant differences in the disperse composition of aerosol was observed as far back as 2008 [58]. But intensive development of infrastructure in Tomsk near Akademgorodok in the subsequent period led to a marked increase in the anthropogenic aerosol load [59, 60].

Summarizing briefly the main results of multiyear measurements in the near-ground atmospheric layer, we note that they were used to determine stable patterns of interannual, seasonal, and diurnal variations in the scattering coefficients, concentration, and size distribution function of particles, concentration of the absorbing material of particles, their parameters of condensation activity and thermo-optical characteristics (see, e.g., [52–63]). We identified for the first time the conditions of nanoparticle formation (nucleation bursts) in the near-ground atmospheric layer of the boreal zone of western Siberia, classified the bursts, and determined the main characteristics of the bursts and the statistics of their frequency of occurrence [64–66].

STUDY OF ATMOSPHERIC AOD IN SIBERIA

Based on the concept of the "optical weather", developed in our Institute [67], and the ensuing need for integrated regular observations, we note that one of the most important aspects of studying different scales of its spatiotemporal variations is the need for information on aerosol characteristics in the entire atmospheric column. The AOD value for different wavelengths of the solar spectrum is used as such a characteristic. Based on data from ozonometer stations in the country [68], IAO researchers developed maps of the distribution of spectral atmospheric transparency and aerosol extinction for the entire territory of the Soviet Union [69]. For each month of the year and for six wavelengths in the range $\lambda = 0.344–0.627$ μm, the authors of that work showed that a complex, multizonal AOD distribution is observed on the territory of the country, identified regions with elevated AOD val-

ues, and also determined the zones with minimal AOD values. The maps and the results of aircraft sensing of particle number concentration were used to analyze the state of the aerosol atmosphere in different pressure structures.

Scientific and practical needs motivated the development of instrumental base, and sun photometers have been widely used for studying the optical characteristics of the atmospheric column and for a subsequent retrieval of the microstructure parameter, asymmetry factor of the scattering phase function, and single scattering albedo of aerosol (see, e.g., [70]).

Measurements of spectral AOD in the region of Tomsk (Akademgorodok and Fonovaya Observatory) had begun in 1992 with using the SP-type sun photometers (wavelength range of 0.34–4 μm), developed at the IAO SB RAS [71]. Owing to their wider wavelength range, these photometers made it possible to obtain for the first time the data on spectral dependence and variations of AOD in the near-IR spectral range [72]. A significant event for development of AOD studies had been the meeting in 2001 with Brent Holben, the leader of AERONET international network (https://aeronet.gsfc.nasa.gov/) [73]. His enthusiasm and vigorous activity had led, as early as 2002, to the signing of an agreement with NASA (GSFC, United States) on the creation of an IAO-headed branching AEROSIBNET network of photometer measurements on the territory of Russia (Tomsk, background regions near Yakutsk, Ussuriysk, Irkutsk, Yekaterinburg, and Zvenigorod).

Analysis of AOD variations on different variability scales [74–76] showed that: there is no trend component in the multiyear AOD variations in the postvolcanic period (after 1994); the largest (about 75%) variations are caused by changes in air masses and pressure structures; the seasonal behavior is the next in significance; and diurnal oscillations make a minimal (10–20%) contribution. Multiyear data are used to detail the average annual behavior of the AOD [77]: the first maximum ($\tau^a_{0.5} = 0.25$) is observed in early May, the second maximum in July, and a minimum ($\tau^a_{0.5} = 0.05$) in November. Availability of regular observations in different geographic regions has made it possible not only to identify the specific features of the spatial AOD distribution in the Asian part of Russia, but also to develop the method of fluid location for retrieval of the aerosol turbidity field of the atmosphere and its transformation in the motion of the air mass [78].

OPTICAL-MICROPHYSICAL CHARACTERISTICS OF SMOKES FROM SIBERIAN FOREST FIRES

Problems of the climatic consequences of nuclear war, actively debated in 1970s and 1980s, were the impetus for starting our research into the optical characteristics of aerosols from biomass burning [79]. In turn, in Siberia the smokes from wildfires in the warm period of the year are an integral part of aerosol weather and, as such, an important climatically significant factor (based on data of monitoring at all stations, smoke haze in this period is observed for about 40 days, on average) (see, e.g., [80, 81]). For the past decades, the IAO SB RAS has performed a large cycle of research into the optical properties and microstructure of smoke aerosol under controllable conditions in the Large Aerosol Chamber (LAC) when smoke plumes from Siberian forest fires come to the region. Based on integrated experiments in the LAC, we found for the first time that the temperature of the burn regime is a determining factor in the formation of optical and microphysical properties of smoke aerosols [82]. It is shown that the flaming combustion phase of forest combustible materials is the main source of particle generation with a high content of absorbing material (single scattering albedo up to $\omega \approx 0.3–0.4$) and a high condensation activity parameter of particles $\gamma \approx 0.5$. Conversely, the pyrolysis (smoldering) phase produces a large number of particles with a weak ($\omega \approx 0.9–1.0$) absorption and a low ($\gamma < 0.1$) condensation activity [83].

Recent studies of the physicochemical characteristics of smoke confirmed that the combustion phase plays a determining role in the formation of the morphology and chemical composition of smoke aerosols. It was found that particles from open flaming-phase smokes stand out in high concentration of fine soot particles with the morphology of agglomerates of the primary particles and a low ratio of organic carbon (OC) to elemental carbon (EC), explaining the high absorptivity of visible radiation. Smoldering-phase smokes show weak absorption because of the dominant group of organic particles with the high OC/EC ratio [84, 85].

Measurements of aerosol scattering and absorption characteristics in smoke plumes from Siberian forest fires, entering the Tomsk region, showed that the arrival of smoke haze is accompanied by a decrease in the relative content of absorbing material (soot) in particles $P = M_{\mathrm{BCeq}}/M_{\mathrm{A}}$, ($M_{\mathrm{A}}$ is the mass concentration of submicron aerosol in dry particle base). It was suggested to use P values below 0.05 as an information feature for indicating the arrival of smoke haze in the measurement region. We apply this criterion in the daily analysis of monitoring data [86].

STUDY OF OPTICAL PROPERTIES AND MICROPHYSICAL COMPOSITION OF AEROSOL OVER THE OCEAN

A large amount of research into the generation of particles from the surface of sea water was performed as early as the twentieth century (see, e.g., [15, 17]). However, considering that the ocean plays an exclusively important (and, possibly, decisive) role in the Earth's climate system [87], it should be noted that aerosol models, proposed before, are already unable to

provide an estimate of radiative characteristics of atmospheric aerosol for solving climate problems. Occupying more than 70% of the Earth's area and possessing high heat capacity, the ocean is, on the one hand, an inertial medium that accumulates thermal energy; and, on the other hand, it serves the main source of the strongest greenhouse gas (H_2O) entering the atmosphere. It is these properties that determine the highest accuracy required of the calculations of radiative fluxes reaching the sea surface for solving climatic problems. We note that most of the earlier data from measurements of the optical and microphysical characteristics of the marine aerosol and the aerosol models contain no information on the absorbing properties of aerosol both in the near-water layer and over the entire atmospheric depth.

Given the relevance of this problem, our Institute, in collaboration with the Arctic and Antarctic Research Institute (AARI) and the Institute of Oceanology (IO) RAS, has been conducting expeditionary studies of aerosol characteristics in various regions of the World Ocean for 30 years (Fig. 1). The aerosol parameters (AOD, particle size distribution function, and concentration of absorbing material) are measured using a set of instruments comprising portable sun photometers (SPM and/or Microtops II), AZ-10 particle counters, and MDA aethalometers. In the first stage (1989–1996), we studied the specific features of the spatial AOD distribution in different regions of the Atlantic (10° S, 60° N), the effect of outflows of continental aerosol, and the dynamics of the aerosol cloud after the eruption of the Pinatubo volcano. We obtained for the first time the statistically justified data on spectral AOD dependence in a wide (0.34–4 μm) wavelength range and carried out AOD zoning over the northern and central Atlantic [88, 89]. The ship-based measurements were used to validate the satellite algorithms of retrieving the AOD over the ocean in the framework of international cooperation [90–93]. Further developments included participation in the Maritime Aerosol Network (MAN) program [94], which is a component of AERONET network. The basis for creating the MAN/AERONET database was the results of our Microtops photometer measurements of the AOD in 2004. Data from measurements obtained by various researchers in approximately 400 expeditions (cruises) in different regions of ocean have been accumulated to date at the MAN/AERONET website. This information is used in problems on satellite sensing and serves a basis for developing and testing models of atmospheric aerosols [95].

Currently, the database created is most representative in the totality of aerosol characteristics (spectral AOD, aerosol and black carbon concentrations, and ion composition) and geographic coverage: 46 expeditions in four oceans and measurements at polar stations (Barentsburg, Tiksi, Cape Baranov, Mirny, and Vostok). Analysis of the data showed that part of the variations in aerosol characteristics were due to the dependence on wind speed and humidity; however, the main effect on interdiurnal variations and spatial inhomogeneities is exerted by outflows of continental aerosols of different types to the marine atmosphere. Based on multiyear studies, we determined for the first time the average latitudinal distribution (with the step of 5°) of aerosol characteristics in the eastern Atlantic from the English Channel to Antarctica [96] and suggested empirical models of zonal distribution [97].

Under the conditions of rapid climate changes, the most pressing problem is to determine aerosol physicochemical characteristics in high-latitude regions of the ocean [98]. Special attention is devoted to the study of absorbing aerosol [99], which reduces both the single scattering albedo of aerosol and the albedo of snow cover. Based on results from a few expeditions, we obtained the first estimates of the average aerosol distribution over Arctic seas (from the Barents to Chukchi Sea) [100]. It was shown that most characteristics decrease from west to east: the AOD (0.5 μm) decreases from 0.078 to 0.028, aerosol concentrations from 3.2 to 1.4 cm^{-3}, and black carbon concentrations from 40 to 20 ng m^{-3}. In 2009 and 2011, we carried out for the first time the unique observations of characteristics of near-ground aerosol at the Russian Antarctic station Vostok. It was found that aerosol and black carbon concentrations were minimal (0.43 cm^{-3} and 10 ng m^{-3} respectively) in June–August, and maximal (0.99 cm^{-3} and 19 ng m^{-3}, respectively) in November–April [101].

AIRCRAFT LABORATORY IN AEROSOL STUDIES IN THE TROPOSPHERE OF WESTERN SIBERIA AND THE RUSSIAN SUBARCTIC

As already indicated above, a cycle of monthly integrated studies of tropospheric composition was initiated in 1997 using the aircraft laboratory [38] in the south of Novosibirsk oblast (Zavyalovo settlement). In contrast to aircraft measurements carried out in the period 1986–1988, these studies included measurements of absorbing material in the composition of the submicron fraction of aerosol. Comparison of data from two cycles of observations, 1986–1988 and 1997–2016, showed that the altitude profiles of the aerosol scattering coefficient agree well with each other. This fact allowed us to conclude that the data on absorbing properties of aerosol also describe quite adequately the aerosol characteristics in the study region, reflect the main features of aerosol weather, and can be used in empirical model development [102–105].

To study aerosol in the troposphere of the Russian subarctic, two round-trip airborne sensing cycles at subpolar latitudes were carried out in 2008 as part of the International Polar Year and POLARCAT projects (in collaboration with the Norsk Institutt for Luft-

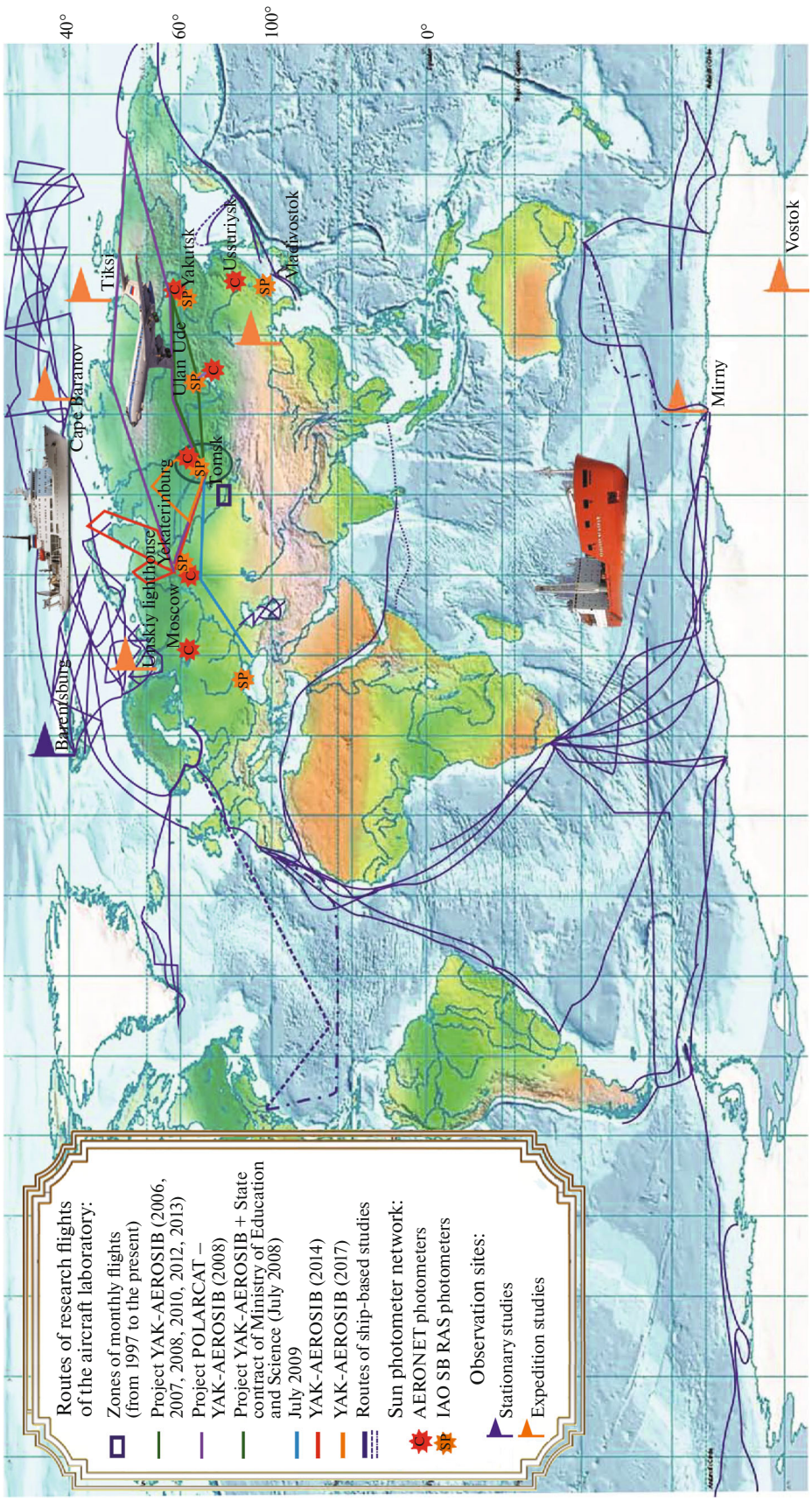

Fig. 1. Geography of aerosol studies in IAO SB RAS.

forskning, Norway) and YAK-AEROSIB [106] (in collaboration with Centre National de la Recherche Scientifique, France). The first flight (from June 7 to 12) was along the route Novosibirsk–Salekhard–Khatanga–Chokurdakh–Pevek–Chokurdakh–Yakutsk–Mirny, and the second flight (from July 21 to 29) along the route Novosibirsk–Mirny–Yakutsk–Lensk–Bratsk. We note that on the subpolar route segments Khatanga–Chokurdakh and Chokurdakh–Pevek–Chokurdakh the flights took place under conditions of extremely high transparency. In these flights at all altitudes we recorded extremely low levels of aerosol concentration (less than 1 $\mu g/m^3$) and soot (about 0.05 $\mu g/m^3$). An interesting feature of the distribution of scattering coefficient and mass concentration of soot over altitude, which we had not encountered previously, was a profile, inverse in character, with these parameters growing with altitude from the near-ground layer up to 7 km. Moreover, minimal ratios of mass concentration of absorbing aerosol to the total aerosol mass (parameter P) were observed in the lower atmospheric layer; and above 3 km we recorded separate layers where P reached 0.10–0.15. These facts indicate that (1) there are no sources of absorbing aerosol in the near-ground layer at these latitudes (70°–72° N), and (2) high P values at high altitudes suggest that polluted air is supplied to this region from remote industrial sources (recall that $P < 0.5$ in wildfires [86]). The cycle of YAK-AEROSIB studies continued in April 2010 and in July 2013 along the route Novosibirsk–Mirny–Yakutsk–Lensk–Novosibirsk showed that aerosol characteristics recorded in these regions under spring and summer conditions corresponded well to the average regional state of the atmosphere.

On October 15–17, 2014, we conducted four flights of the aircraft laboratory toward the Russian subarctic and toward the Kara Sea (55.0°–74.8° N, 61.3°–82.9° E): flight 1 along the route Novosibirsk–Salekhard; flight 2 along the route Salekhard–Kara Sea (eastern sector, northward of the Dikson settlement)–Salekhard; flight 3 along the route Salekhard–Kara Sea (western sector, southward of Novaya Zemlya island)–Salekhard; and flight 4 along the route Salekhard–Novosibirsk. We note that, like in the 2008 flights, in 2014, in the flights on all segments of the route there were maxima in the parameter $P \approx 0.10$–0.18, indicating that industrial pollution plumes entered the atmosphere over the Kara Sea water basin.

Unique results were obtained in the aircraft expedition from July 31 to August 1, 2012, along the route Novosibirsk–Tomsk–Yakutsk–Novosibirsk, which was carried out during severe wildfires on the vast territory of Tomsk oblast, Krasnoyarsk krai, and Yakutia. During these flights we collected information on the complex of optical and microphysical characteristics, not only under the background conditions of the atmosphere of western Siberia and in the Arctic regions of the country, but also under the influence of fires with different intensities [107].

The aircraft measurements of the optical and microphysical characteristics of aerosol during the fire period have made it possible to estimate quantitatively the strengths of the sources and evolution of smoke plumes [108]. The detailed information on concentration and distribution of soot particles over altitude [109] was used to analyze the sources, spatio-temporal variations in the air of the northern regions of Russia [110, 111], and to verify the satellite methods of estimating the soot supply during the Siberian forest fires [112]. The results of numerical simulation, performed on the basis of retrieved radiatively significant aerosol characteristics in the troposphere according to empirical ground-based and aircraft data for background and smoke-laden states, showed that the diurnal deficit of the total solar radiation, caused by the occurrence of optically dense smoke at the underlying surface level, is about 13 MJ/m^2 [113], and, hence, the smoke haze acts as a cooling factor. At the same time, the presence of a large number of absorbing particles in the atmosphere leads to a heating of the central part of the smoke layer at altitudes of 3–4 km, where the diurnal temperature changes are about 2.5°C at $\tau_{smoke} = 2$, reaching 5.5°C at $\tau_{smoke} = 4$, thus favoring an increase in the lifetime of smoke haze, which should be taken into consideration in predictions of smoke haze dispersal [114].

CONCLUSIONS

In recent decades, scientists have concentrated special attention on studies of the contribution of organic substances to aerosol composition under real atmospheric conditions, i.e., on the problem still poorly studied (see, e.g., [115]). Since 2013, in collaboration with the Institute of Petroleum Chemistry (IPC), ICK, and the Institute of Computational Mathematics and Mathematical Geophysics (ICM MG) SB RAS, scientists in the IAO SB RAS also implemented the program "Organic Aerosol in the Atmosphere of Siberia and the Arctic" [116–120]. As results of the modern studies show (see, e.g., [116]), the percentage of organic compounds in the mass concentration of submicron particles varies from 20 to 70% and may reach 90% (!) in the ultrafine size range. It was found that the major organic component of these particles contains water-soluble compounds (see, e.g., [121]). These facts force us to reconsider the interpretation of certain well-established viewpoints on the sources and nature of particle production in the gas–particle process (in a theoretical description of this process, the role of water vapor at the initial stage of nucleus formation and a subsequent clustering stage is still unclear).

We turn to the quotation from the work by Rosenberg: "Data from chromatographic measurements of F.W. Went, which proved that, at any rate, a consider-

able part of Aitken nuclei is biogenic or anthropogenic, suggest that the average concentration of decomposition products terpenoids in the atmosphere is about 104 μg/m^3 (i.e., about 106 tons in the entire atmospheric volume), and that their total production on the globe is close to 5–10^8 tons per year .... Seemingly, the study of the nature and origin of Aitken nuclei and how they grow in the presence of atmospheric moisture thereby becomes one of the most important problems not only in the optics, but also in the thermodynamics and chemistry, of atmospheric aerosol because, as is now clear, the Aitken nuclei act as one of the most important factors, determining the course of condensation and, thereby, weather formation processes" [5]. We stress that the comprehension of this problem and of its importance for understanding aerosol life was discussed in the work published in 1968.

The first results from measurements of natural photochemical hazes were published in 1976 [121]. However, many more long years were still required before large-scale studies were initiated and reliable results obtained [122]. In our opinion, this example shows very clearly how multifaceted the research subject is, and how much time and labor are required of the scientists to create an adequate instrumental basis and carry out long-tem measurements before new results can be achieved.

We conclude this review with the hope that this material and bibliography will be useful to young staff scientists beginning their career in searching for interesting and important problems on atmospheric aerosol study.

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CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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