From nano- to global scales: properties, processes of formation, and aftereffects of atmospheric aerosol impacts. 2. Field observational experiments. America, Western Europe, and high latitudes

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Analysis is presented of the results of combined investigations into the atmospheric aerosol properties, obtained during field observational experiments in America, Western Europe, and in high latitudes.

Introduction

Part 1 of this paper discusses the results of investigations into the aerosol properties obtained in the field observational experiments in Africa and Asia.[4] The main objective of the Part 2 is analysis of data obtained in America, Western Europe, and in high latitudes. Before addressing these data, consider some general tendencies in the atmospheric aerosol studies. In addition to the combined field observational experiments, these general tendencies include: 1) continuing improvement of observational techniques (both ordinary and satellite ones) with considerable attention paid to remote sensing; 2) development of the global network of aerosol monitoring (in the first turn, with the use of multichannel sun photometers and lidar sensing); 3) justification of more adequate models of the atmospheric aerosol.[1,5,30,64,74–76]

Remote sensing

Techniques of lidar sensing of the atmosphere occupy an important place in the development of observational facilities.

According to the Third Assessment Report of the Intergovernmental Panel on Climate Change (IPCC), global warming forecasts (even those using the best climate models) still include many uncertainties.[3] In particular, a great uncertainty still exists in the estimates of the radiative forcing (RF) due to inadequacy of satellite observations of the Earth's radiative budget (ERB) and the lack of reliable understanding of the roles of clouds, aerosol, and water vapor (as well as interactions between them) in ERB.

In this context, the National Space Development Agency of Japan (NASDA) and the Communications Research Laboratory (CRL), Japan, in co-operation with the European Space Agency (ESA) started the Phase A of the Earth CARE Project. The main goals of this project are satellite observations of three-dimensional fields of aerosol and cloud characteristics, as well as of the ERB components, and further studies of the aerosol and cloud effect on the radiative budget and radiative forcing.

The Earth CARE satellite described by Kimura et al.[40] includes: 1) RADAR-CPR for cloud sensing; 2) LIDAR-ATLID for sensing the atmosphere; 3) MSI multispectral videoradiometer; 4) ÂBR broadband radiometer; 5) FTS Fourier-transform spectrometer. The Doppler radar at the frequency of 94 GHz has a reflector of 2.5 m diameter and is characterized by the sensitivity equal to 38 dB. The vertical and horizontal resolutions are 100 m and 1 km, respectively. The Fourier-transform spectrometer operates in the spectral range from 5700 to 25000 nm at the resolution of 0.5 cm^{-1} and the viewed area of 10×10 km. The Earth CARE project is planned as a 2-year mission to be launched in 2008. Phase-A study would have been completed by the end of 2003.

Bravy et al.[15] have analyzed the possibilities of using a multifrequency pulsed chemical DF laser (10 uniformly distributed channels in the wavelength range $\lambda = 3600–4200$ nm) for remote sensing of chemical composition and microphysical characteristics of atmospheric aerosol and discussed briefly the experience of testing it under laboratory conditions.

To retrieve aerosol properties from satellite spectral measurements of outgoing shortwave radiation, the contributions coming from the surface and various optically active atmospheric constituents need to be separated. This is most easily done over dark surfaces, i.e., water surface. This principle has been applied to the retrieval of aerosol optical depth (AOD) over water from satellite observations in the near-infrared region. The AOD in the UV can be determined both over water and over land, using the same principle of a dark surface.

For longer wavelengths, data obtained with the ATSR-2 satellite for two viewing directions were very significant. In this connection, de Leeuw et al.[49]

discussed the results of testing the dual-view algorithm by use of data obtained over the Indian Ocean (INDOEX area) and South Africa (SAFARI experiment). The results obtained showed that the AOD could be retrieved within reasonable accuracy limits. Apart from the ATSR-2, similar algorithms were developed for AATSR and SCIAMACHY (ENVISAT) and OMI (EOS-TERRA).

Gassó and Hegg[29] proposed a technique for retrieval of column aerosol mass concentration (AMC) and concentration of cloud condensation nuclei (CCNC) from the primary aerosol products of Moderate-Resolution Imaging Spectroradiometer (MODIS). The technique uses direct proportionality between the AMC and aerosol optical depth with the proportionality constant dependent on the MODIS-derived r_{eff}, the parameter η (contribution of the accumulation mode of aerosol to the total radiance), ambient *RH*, and all that under assumption of constant aerosol chemical composition. The CCNC is derived from a parameterization of concentration as a function of the retrieved aerosol volume.

By comparing with *in situ* data, it was shown that retrievals for the case of dry ambient conditions (dust) are improved if using a proportionality constant dependent on r_{eff} and η obtained in the same pixel. In the cases of high humidity, the improvement achieved with new method was inconclusive because of the difficulty in accounting for the vertical profile of relative humidity. In practice, the same results were obtained by using another retrieval technique developed for processing the MODIS Airborne Simulator (MAS) data, though the latter technique proved a little bit more reliable.

Von Hoyningen-Huene et al.[37] proposed a technique for retrieval of the aerosol optical thickness of the atmosphere over land surfaces from measurements of the shortwave radiation intensity using instruments looking to nadir for the ENVISAT radiometers SCIAMACHY and MERIS. The technique is based on lookup tables (LUT) of *a priori* data, characterizing the relation between the AOT and the reflectance of the surface–atmosphere system for the wavelengths shorter than 0.67 μm. The reflectance is determined from calculations of shortwave radiation taking into account the spectral albedo of the surface. Over land, the surface reflectance is estimated by a combined model of bare soil and green vegetation spectra, tuned by the normalized differential vegetation index (NDVI) of the satellite scene. The method has been tested and validated with SeaWiFS data and with aerosol properties of the closure experiment LACE-98 (Lindenberg Aerosol Characterization Experiment). For shortwave channels (412–510 nm), an agreement within 20% was achieved between the retrieved data and data of ground-based observations.

Fussen et al.[28] have developed a combined technique for remote sensing of the atmosphere based on combining the inversion operator, which is applied in the case of significantly noisy observations, and the optimal estimation. The success of realization of this technique depends on the completeness of the available *a priori* information about the state of the atmosphere. Reference 28 exemplifies the processing of "occultation" data of remote sensing of aerosol, illustrating that the technique proposed is quite robust.

Holzer-Popp et al.[36a] described a new technique for retrieval of the aerosol optical depth called Synergetic Aerosol Retrieval (SYNAER). This technique used simultaneous measurements with the ATSR-2 radiometer (Along the Track Scanning Radiometer) and the GOME spectrometer (Global Ozone Monitoring Experiment). Both these instruments were operated from onboard the European Remote Sensing (ERS-2) satellite.

These observations allowed the retrieval of AOT for the boundary layer (BLAOT) and determination of the aerosol type both over land and over ocean. The latter was realized as estimation of the BLAOT percentage of six representative components from the Optical Parameters of Aerosols and Clouds (OPAC) data set (high spatial resolution of ATSR-2 permits accurate cloud detection).

The discussed BLAOT technique of retrieval was based on the comparison of the recorded spectra of outgoing radiation in the visible and near-IR regions (obtained from GOME data after rejection of the cloud effect) with those calculated by different aerosol models. The agreement between the spectra at the least-squares fit determined the choice of the adequate aerosol model. This technique was planned to be used for the future sensor pairs Scanning Imaging Absorption Spectrometer for Atmospheric Cartography (SCIAMACHY) / Advanced ATSR (AATSR) on Envisat and GOME-2 / Advanced Very High Resolution Radiometer (AVHRR) on the METOP satellite.

One of the promising remote sensing techniques for retrieval of optical properties of atmospheric aerosol and clouds is the use of satellite data on the spectral distribution of the outgoing shortwave radiation at high spectral resolution in the O_2 A-band centered near 762 nm. To experimentally test this technique, the NASA Langley Research Center has developed the Langley Airborne A-Band Spectrometer (LAABS) operating in the spectral range 12980–13160 cm^{-1} (~760–770 nm) with the resolution about 0.60 cm^{-1}. Pitts et al.[63] described thoroughly the results of laboratory tests of LAABS capabilities, and the field tests were carried out in July 2001 during test flights over the Chesapeake Bay (USA). The tentative results obtained during three flights proved to be positive. It was planned to validate the instrument against actual *in situ* observations of the aerosol and cloud optical parameters.

Yi et al.[85] described the algorithm for retrieval of the single scattering albedo and the scattering phase function from multiangle sky radiance measured at the ground. The first stage of solution of the problem consists in derivation of the linear equation system from the radiative perturbation theory to approximately represent exiting radiance as a function of single scattering albedo and the coefficients of Legendre polynomial approximating the scattering phase function. However, this linear system is ill-posed. Therefore, regularization is introduced to obtain a stable solution. An important and difficult step of the regularization is the determination of the regularization parameter, especially when iteration is involved in the inversion.

Yi with co-workers[86] have checked the adequacy of this technique of retrieving the single scattering albedo and the scattering phase function of atmospheric aerosol from observations of the angular distribution of the clear-sky radiance. For this purpose, they used the data of synthetic experiment with varying levels of random noise. The results obtained indicated the stability of the algorithm even at the high noise level. Sensitivity studies have been conducted to analyze the influence of errors in the surface albedo and the total optical thickness on retrieval. Errors in both of these factors may have an impact on the retrieval of single scattering albedo. The reliability of retrieval was also analyzed through the comparison with data of real observations being conducted at the aerosol observation network.

Zhao et al.[88] have analyzed the effect from abandonment of the assumption on sphericity of aerosol particles when retrieving the aerosol optical thickness τ from AVHRR data taking into account the nonspherical shape of dust aerosol particles.

The values of τ retrieved with assuming both spherical and non-spherical shape have been compared with the surface AERONET observations at two desert dust sites to evaluate and quantify the effect of non-spherical dust particles on the satellite aerosol retrievals. The errors (especially the random ones) in the satellite-retrieved τ have been reduced as a result of adopting the non-spherical assumption in the AVHRR aerosol retrieval algorithm.

Ellrod et al.[24] proposed a new technique for detection of volcanic ash from the data of meteorological satellites in three infrared (IR) bands centered near 3900, 10700, and 12000 nm. The technique is based on the sum of two brightness temperature differences (BTDs), scaled to maximize the brightness and contrast of volcanic ash in the output image. The physical effects attributed to the observed BTDs that help distinguishing between the volcanic ash and various meteorological cloud types are (1) differential absorption by volcanic ash or sulfur dioxide at 3900 nm, 10700 nm, and 12000 nm and (2) strong reflectance of solar radiation by ash at 3900 nm, which undergoes diurnal variation. The best results of retrieval were obtained during daylight hours over any surface, and at night when the ash cloud was over the ocean or other large body of water.

Alexandrov et al.[6] discussed the results of statistical analysis of the aerosol optical thickness (AOT) variability in time and space on the spatial scales from 0.2 to 1200 km using data sets from the local network of Multi-Filter Rotating Shadowband Radiometers (MFRSRs) located in the Southern Great Plains (USA). Twenty one instruments were arrayed across north-central Oklahoma and south-central Kansas. The data of the ground-based network were compared with AOT derived from MODIS satellite aerosol retrievals. The MODIS retrievals were also used to study large-scale AOT variability. The results obtained suggested that the variability of AOT splits into sub-regimes of three main scales. Three-dimensional turbulent transport dominates in small scales (0–30 km), the influence of 2D turbulence at scales larger than 30 km makes AOT more non-stationary, and finally, in contrast the AOT variations on the scales larger than 100 km make the non-stationary AOT behavior weaker.

An Nd:YAG laser-based portable 3-D-scanning lidar was developed at the Universitat Politecnica de Catalunya (UPC, Technical University of Catalonia, Barcelona, Spain) for retrieval of aerosol properties in the free atmosphere. The lidar simultaneously operated at the 1064-nm and 532-nm wavelengths and was used in its scanning mode from 15° to 70° from zenith with 5° steps. This lidar was used by Sicard et al.[69] to retrieve vertical profiles of various aerosol properties (first of all, the backscatter-to-extinction ratio *b*/*å*) from observations in the Barcelona area, where an important number of pollution and Saharan dust events can be observed in the atmosphere all year round.

The algorithm was based on the variational method used to invert the multi-angular profiles and to retrieve the aerosol optical thickness and the vertical profile of the backscatter coefficient *b* without any assumption on the aerosol type. Quite good results of retrieval of the coefficient *b* (differences less than 20%) were observed in the 0.3–2.5 km region with the mean value of $0.030\ \mathrm{sr}^{-1}$, whereas no agreement were achieved at higher altitudes. The lidar profiles closer to the zenith allowed calculation of the Angström coefficient in backscatter to be done, which was in agreement with that retrieved by the variational method. The difficulties encountered at heights above 1.6 km showed the presence of atmospheric inhomogeneities, which are likely connected with the strong spatial variability of aerosol properties.

In situ aerosol measurements. Considerable progress of remote sensing by no means decrease the significance of direct methods of measuring the aerosol characteristics. This is especially true for the chemical composition of aerosol. In this connection, adequate understanding of the nature and properties of atmospheric aerosol is seriously hampered by the lack of the instrumentation needed. The instrumentation for measurement of the mass concentration and particle number density, as well as the aerosol microstructure, is developed quite well. However, the situation with measurements of the chemical composition of aerosol is different. The most widely used technique now is the technique of chemical analysis of samples collected with filters, but its application faces various difficulties. In recent years, aerosol mass-spectrometry providing for real-time measurement of the aerosol chemical composition has actively been developed. This technique involves sampling of aerosol, evaporation of the aerosol matter, ionization of gas molecules produced, and mass-spectrometric analysis of ions. Evaporation and ionization are carried out with the aid of high-power lasers. The practical use of this technology of obtaining quantitative results is seriously hindered by a number of factors. For example, the role of particles with the size smaller than 0.2 μm is accounted for inaccurately. Desorbtion and ionization assume significant loss of information in the case of complex organic compounds, which is caused by intense molecular fragmentation.

Allan et al.[7] discussed the results obtained by applying a new aerosol mass spectrometer (AMS), manufactured by Aerodyne Research, Inc., in which aerosol is evaporated on a heated surface and then the quadrupole mass-spectrometry with electron impact ionization is accomplished. In this case, it is possible to obtain the quantitative information on the chemical composition and size of volatile and semivolatile fine airborne particulate matter with high time resolution. The data processing includes the conversion from the mass spectrum (MS) mode to atmospheric mass concentrations of chemical species (in μg/m^3) based on calibration data.

Illustrating the advantages of mass-spectrometry, Allan with co-workers[8] discussed the measured results on the chemical composition of atmospheric aerosol obtained using an Aerodyne aerosol mass spectrometer from sampling air in two U.K. cities, namely, in Edinburgh during October 2000 and in Manchester during July 2001 and January 2002. These results include data on the total mass concentrations and size-resolved mass distributions of nitrate, sulfate, and organic compounds. All the three cases evidences have been found of the emissions from local transport, with the mass-modal aerodynamic diameter of 0.1–0.2 μm. This mode is dominated, in its formation, by hydrocarbons showing only little evidence of oxidization. A larger mode consisting of inorganic chemicals and oxidized organics has also been observed, which likely appeared from sources outside the cities and showed evidence of internal mixing. The mass-modal aerodynamic diameter varied between approximately 200–500 nm in winter and 500–800 nm in summer. The summer data also showed an increased mass load without any increase in total particle number. Evidence of material building up and ageing in the atmospheric surface layer during periods of low winds was also observed.

Improvement and wider application of various facilities for aerosol observations have opened wide promises for obtaining data that are more representative and, in particular, for justifying new aerosol models.

Aerosol models. Gong et al.[31,32] described the Canadian Aerosol Module (CAM) developed for use with climate and air quality models. It allows for the major aerosol processes in the atmosphere: generation, hygroscopic growth, coagulation, nucleation, condensation, dry deposition/sedimentation, scavenging in the subcloud layer, and activation. The cloud module explicitly accounts for the microphysical processes in treating the aerosol-cloud interactions and chemical transformation of sulfur species in clear air and in clouds. Gong with co-workers[31,32] optimized the ÑÀÌ algorithm to efficiently solve the complicated size-segregated multicomponent aerosol system and make it feasible to be included in global and regional models. As to the aerosol particles, those were considered an internal mixture for all types of aerosols except for particles of dust and black carbon, which were assumed the external ones being mixed near their sources.

To test the adequacy of the proposed algorithm,[32] emissions of anthropogenic and natural aerosols to the atmosphere were simulated for two aerosol types: sea salt and sulfates. A comparison was made between two numerical solutions of the aerosol algorithm: the process-splitting method and the method of ordinary differential equations (ODE). It was found that the process-splitting method used in this model yields a solution that is about 15% more reliable than that obtained by ODE method, in the case of total sulfate mass concentration and a little bit less reliable (by about 1%) for sea-salt concentration. It is important that computations by process-splitting method are more than 100 times faster than those by ODE technique.

Gong with co-authors[31] have studied the sensitivity of the simulated size distributions to the number of size bins. The diffusion behavior of each individual process was quantitatively characterized by the difference in the mode radius and standard deviation of a lognormal curve fit of distributions between the approximate solution and the 96-bin reference solution. The numerical simulation showed that both the number and the mass size distributions were adequately predicted by a sectional model of 12 bins in many situations in the atmosphere where the sink for condensable matter on the existing aerosol surface is high enough that makes the nucleation negligible in production of new particles. Total mass concentration was adequately simulated using lower size resolution of 8 bins. However, to properly resolve nucleation mode in the size distributions and minimize the numerical diffusion, a sectional model of 18 size bins or larger was needed.

Network observations. Along with the data of satellite and airborne observations, regular ground-based observations, especially, on the AERONET global network considerably increase of the adequacy of the aerosol models.

Since the mid-1990s, with the onset of commercially available, high-quality, low-maintenance automated instrumentation for measurement of direct solar radiation in order to retrieve the aerosol optical depth of the atmosphere the observation network of sun photometers (SP) has extended significantly. In this connection, the comparability of sun photometers used in the exiting networks is an important issue. At the Bratt's Lake Observatory four separate types of sun photometers are operated in cooperation with three different networks: Aerosols in Canada, Global Atmosphere Watch, and the U.S. Department of Agriculture UV-B Monitoring Program. As the instruments used are similar to other networks from around the world, it is believed that the results of obtained with sun photometers in summer 2001 can be transferred, at least partially, to other operational networks. The results obtained by McArthur et al.[54] showed that the errors in retrieval of instantaneous AOT values are within ± 0.01. Over the three-month comparison, the root mean square difference of aerosol optical thickness at 500 nm between the instruments was 0.0069. Thus, the sun photometers are quite suitable for global monitoring of the aerosol optical thickness.

Continuing the developments based on the use of AERONET data, Smirnov et al.[70] studied the properties of atmospheric aerosol over ocean, which can vary considerably, depending on contributions of major aerosol components, i.e., urban / industrial pollution, desert dust, biomass burning, and maritime aerosol as well. Based on the Aerosol Robotic Network (AERONET) data from three island locations: Bermuda (Atlantic Ocean), Lanai, Hawaii (Pacific Ocean), and Kaashidhoo, Maldives (Indian Ocean), Smirnov with co-workers[71] have justified the model of the maritime aerosol component, which can be used, in particular, for solving the problems of atmospheric correction. To retrieve the maritime component, they considered the dataset only when the aerosol optical depth at the wavelength of 500 nm was below 0.15 and Angström parameter was less than 1.

The inferred maritime component in the columnar size distribution, which was found to be very similar for the three case studies, was bimodal with a fine mode at the effective radius $r_{\text{eff}} \sim 0.11–0.14$ μm and a coarse mode r_{eff} of $\sim 1.8–2.1$ μm (these values are quite close to other literature data). The refractive index was independent of wavelength and its estimated value was $1.37–0.001i$ (single scattering albedo of about 0.98). Fractional contributions of the fine and coarse modes to the AOT (500 nm) computed were within the range of $\tau_{\text{fine}} \sim 0.03–0.05$ and $\tau_{\text{coarse}} \sim 0.05–0.06$. The Angström parameter varied from 0.8 to 1.0 (340–670 nm) and from 0.4 to 0.5 (870–2130 nm). Aerosol phase functions were very similar for all three sites considered.

The CERES instrumentation installed aboard a TRMM (Tropical Rainfall Measurement Mission) satellite is a passive radiometer, which has three broad spectral bands (300–5000, 300–100000, and 8000–12000 nm) and measures the Earth's radiation budget. The TRMM instruments include also the VIRS visible and infrared scanning radiometers to measure outgoing radiation in the visible and IR spectral regions. Zhao et al.[89] compared the TRMM / CERES-VIRS data with the AERONET sun photometer observations in order to validate the aerosol optical thickness (τ) retrievals over ocean and to quantitatively evaluate the effects of potential subpixel cloud contamination (using VIRS data) and of the wind near the ocean surface (which determines the ocean surface roughness).

A positive correlation between the surface wind speed and τ has been observed for high speed of the surface wind, which likely represented actual relationship between the surface wind and the wind-driven aerosols rather than the disturbing effect of the surface reflectance associated with the rough ocean surface.[71] After the potential subpixel cloud contamination was minimized and the effects of large surface wind removed, the positive systematic discrepancy between the satellite and ground-based data have been reduced from 0.05 to 0.02 in VIRS channel 1 (630 nm) and from 0.05 to 0.03 in channel 2 (1610 nm). Random errors have also been reduced from 0.09 to 0.06 at 630 nm and from 0.06 to 0.05 at 1610 nm. The remaining discrepancies at small τ are mostly caused by calibration errors, radiometric noise, and inadequate definition of aerosol parameters in the retrieval algorithm. The future works will aim at estimation of the errors of τ retrieval for different types of aerosol.

Taking into account that the particle size distribution (PSD) of aerosol is, as a rule, bimodal, it is possible to extract the optical depths (τ_f and τ_c) due to fine and coarse aerosol fractions from the spectral shape of the total aerosol optical depth ($\tau_a = \tau_f + \tau_c$). O'Neill with co-authors[60] showed that the adequacy of this technique is ensured by: 1) its physical validation; 2) the fact that τ_c variations agree with the photographic evidence of thin cloud events, while the adequacy of data on τ_f variations agrees with the photographic observations under clear sky and haze conditions; 3) the fact that the estimates of τ_f and τ_c correlated quite well with the corresponding values obtained by inverting data on the sky brightness and total extinction of solar radiation. Also, these data allowed the validation of the cloud-screening algorithm to be performed.

In recent years, particular attention has been paid to the investigations of the biogenic component of atmospheric aerosol.[2] Rannik et al.[65] discussed the results of gradient (correlation) observations of aerosol particle fluxes above a 40 years old forest of Scots pine (*Pinus sylvestris L.*) carried out at the Station for Measuring Forest Ecosystem–Atmosphere Relations (SMEAR) in Hyytiälä in southern Finland (61°51′N, 24°17′E, 181 m above the sea level). The program of observations involved the measurements of the aerosol particle size distribution (particle diameter 0.003–0.5 μm) with Differential Mobility Particle Sizer (DMPS) system set at the height of 2 m and the condensational particle counter (CPC) installed at the level of 23 m (the mean height of the forest was 14 m). The CPC data were used to detect the total particle number concentration in the size interval from about 0.01 to 0.5 μm.

Analysis of observations showed that the aerosol particle concentration is often characterized by non-stationary behavior, more frequently than other scalar parameters like temperature, water vapor and carbon dioxide. During stationary episodes most of the flux estimates indicated deposition of particles to forest. Size-dependent deposition rates were estimated from auxiliary measurements of particle size distribution. A semi-empirical deposition model developed in Ref. 65 explained most of the variance observed in the particle fluxes when size spectrum was dominated by particles of the size below 0.1 μm. However, estimates of the deposition rate for particles of above 100 nm sizes remained very uncertain.

1. North America

A number of large-scale field observational experiments involving the investigations of atmospheric aerosol have been carried out in North and South America. On the territory of the USA, the emphasis was, naturally, on the estimation of anthropogenic impact and, in this connection, field experiments in big cities.

Atlanta Supersite Project

In summer of 1999, a super site has been instrumented in the center of the Atlanta city (Georgia, USA) under financial support from the U.S. Environmental Protection Agency (EPA). The Atlanta Supersite Project was located at the existing Southeastern Aerosol Research Characterization Study (SEARCH) / Aerosol Research Inhalation Epidemiology Study (ARIES) site in Jefferson Street in NW Atlanta, Georgia. The "supersite" for aerosol studies under the urban atmosphere conditions was organized within the Southern Oxidants Study (SOS) Project. The short-term primary goal of the Atlanta Supersite Project was to evaluate and compare advanced measurement methods for particulate matter structure (mass), chemical composition, and physical properties of aerosol. The long-term goal was to study the physical and chemical processes in the urban atmosphere under the intense anthropogenic impact.

Three categories of instruments were used at the site to obtain the information about aerosol: 1) time-integrated or discrete filter-based methods like those (PM2.5) used in EPA's Network for monitoring fine aerosols; 2) methods of continuous or quasi-continuous measurements of the chemical composition of atmospheric aerosol and trace gases, most of which are still under development; and 3) single particle mass spectrometers, the most advanced methods looking at the chemical composition of individual particles. Solomon with co-workers[72,73,74] compared 12 filter-based samplers operated in a discrete sampling mode. Samples were analyzed for total aerosol mass, sulfate, nitrate, ammonium, organic carbon, elemental carbon, and some trace elements. As a rule, the concentrations for most species were sufficiently above the detection limits, except for nitrates and some trace elements (for example, As and Đb). In most cases, real differences existed among the data obtained with the samplers tested for most of the species studied, with sulfate and ammonium being the exceptions. These differences were due to various factors, including the sampler design, as well as due to the use of different methods of chemical analysis. Comparability among most of the samplers for a given species was: for mass (±20%); sulfate (±10%); nitrate (±30–35%); ammonium (±10–15%); organic carbon (±20% to ±35–45%); elemental carbon (±20 to ±200%); and minor and trace elements (±20–30%).

Solomon et al.[73] overviewed the instruments for investigation of aerosol and trace components, which were employed within the Southern Oxidants Study (SOS) Project supported by the U.S. Environmental Protection Agency (EPA). The supersite for testing and comparing the aerosol measurement techniques was located at the existing ACE site in the northwestern of Atlanta city. The analyzed technologies included filters and impactors, as well as instruments for measuring the chemical composition of aerosol (sulfate, nitrate, ammonium, organic carbon, elemental carbon, trace elements) and gas-phase precursors of aerosol. Analysis of chemical composition of aerosols and precursor gases has been performed with four mass spectrometers. In addition to these data meteorological data as well as data on volatile organic compounds (VOC), oxidized VOC, and NO_y, that is, data on the conditions of aerosol formation and accumulation in Atlanta. Analysis of the results revealed some differences between data obtained with different instruments, but (with the use of the adequate data processing technique) provided quite comprehensive information about the dynamics of aerosol in Atlanta city.

Since August 6 and until September 1, 1999, an intensive field campaign was conducted at the supersite in Atlanta, Georgia, focused on the characterization of urban particulate matter with the use of an aerosol time-of-flight mass spectrometer (ATOFMS), which enabled measuring continuously the aerodynamic size and chemical composition of individual particles of the fine aerosol fraction (0.2–2.5 μm). Liu with co-workers[52] have analyzed the results obtained (a total of 455444 mass-spectra) and revealed the presence of seven types of aerosol: 1) potassium-containing; 2) potassium-containing with admixture of secondary components (ammonium, nitrate, sulfate); 3) carbonaceous (both EC and OC);

4) carbonaceous with admixture of secondary components (ammonium, nitrate, sulfate); 5) dust; 6) dust with admixture of secondary components (ammonium, nitrate, sulfate); 7) secondary components (ammonium, nitrate, sulfate). The main characteristics of the ion components of aerosol are described in Ref. 52. On the whole, the results obtained demonstrate high complexity and variety of the urban aerosol composition.

Wenzel et al.[82] compared the ATOFMS results with the data obtained with a laser counter and revealed significant discrepancies depending on the chemical composition of the particulate matter. These discrepancies were especially large at enhanced ammonium and sulfate mass concentrations. The missed particles occurred mostly in the range of small sizes (0.35–0.54 μm) that well correlated with the measurements of optical wave scattering. In Ref. 82 a scaling procedure was developed that allows one to account for the ATOFMS chemical bias. The results obtained in this way were compared with measurements obtained using other instruments.

During August of 1999, as part of the Southern Oxidants Study Supersite Experiment, Rhoads with colleagues[66] collected size-resolved measurement data on the chemical composition of aerosol particles with a unique real-time laser desorption/ionization mass spectrometry technique. The rapid single-particle mass spectrometry instrument was capable of analyzing “ultrafine” particles with aerodynamic diameters ranging from 0.01 to 1.5 μm in real time. Under the heaviest loading observed in Atlanta, particles were analyzed at a rate of roughly once per second in sizes ranging from 0.1 to 0.2 μm. Nearly 16000 individual spectra were recorded over the course of one month during both daytime and nighttime sampling periods.

Evaluation of the data obtained indicated that the composition of the ultrafine (less than 100 nm) particles was dominated by carbon-containing compounds. Larger particles showed variable composition, but typically they appeared to have organic carbon characteristics mixed with an inorganic component (e.g., crustal materials, metals, etc.). During the experiment, 70 composition classes were identified. Rhoads et al.[66] reported on the average spectra and correlations with different meteorological parameters for all major compound classes and a number of the minor ones. The major composition classes were identified from the primary peaks in their spectra as organic carbon (about 74% of the particles), potassium (8%), iron (3%), calcium (2%), nitrate (2%), elemental carbon (1.5%), and sodium (1%). Many of these compound classes appeared in repeatable size ranges and quadrants of the wind rose, indicating emission from specific sources.

Between 23 August and 18 September 2000, an ultrafine-particle mass spectrometer (RSMS-II) was employed in Houston, at a point located to the north from major sources of industrial emissions into the atmosphere. Phares with co-workers[62] acquired roughly 27000 aerosol mass spectra, which were classified and labeled based on the spectral peak patterns using the neural networks algorithm. The frequency of occurrence of each particle class was correlated with time and wind direction. Some classes were present permanently, while others appeared intermittently or for very short time. The most frequently detected species at the site were potassium and silicon, with lesser amounts of organics and heavier metals.

In connection with the established harmful effect of coarse urban particulate matter (data of PM-2.5) on the human health, the US Environmental Protection Agency adopted in 1997 the National Ambient Air Quality Standards (NAAQS), which limited the maximum permissible concentration for PM (ÐÌ-2.5) in the atmosphere. According to NAAQS, the PM2.5 standards are 15 micrograms of PM2.5 particulate matter/cubic meter of air for annual mean concentration and 65 micrograms of PM2.5 particulate matter/cubic meter for the daily mean one. However, in May 1999 the Supreme Court of the USA cast doubt on the legality of these standards.

In connection with this, the monitoring network was established in the metropolitan Atlanta, Georgia, to measure fine particulate mass (PM-2.5) and composition. According to the results obtained by Butler et al.,[18] the annual mean mass concentrations ranged (depending on the monitoring site) from 19.3–21.2 $\mu g/m^3$, maximum 24-hour mean concentrations were 44.3–51.5 $\mu g/m^3$, and maximum 1-hour mean concentrations were 73.3–87.9 $\mu g/m^3$. Mass concentrations varied significantly with season and time of day. Approximately 75% of the mass was identified using various chemical analyses. The data obtained showed that carbonaceous material, sulfate, ammonium, and nitrate were the largest constituents of the aerosol substance.

Lim with co-workers[51] carried out an intense field campaign at the Atlanta Supersite Experiment at a ground-based measurement site in Jefferson Street in midtown Atlanta, Georgia, from 3 August to 1 September 1999. Quasi-continuous particulate organic and elemental measurements that were conducted as a part of the experiment using an *in situ* thermal-optical carbon analyzer, an ambient carbon particulate monitor, Radiance Research particle soot absorption photometer, a flash vaporization carbon analyzer, and an AE-16 aethalometer. The “intersampler precision” with which the quasi-continuous particulate total carbon (TC), organic carbon (OC), and elemental carbon (EC) were measured was 7, 13, and 26%. Correlations between pairs of OC measurements were moderate (r^2 = 54–73%), and correlations between pairs of EC measurements were high (r^2 = 74–97%). Differences in the reported OC concentrations were small compared to differences in the EC concentrations. In general, the agreement between quasi-continuous samplers was quite good, especially if taking into account quality of previous particulate carbon comparisons.

In August 1999, five new instruments for quasi-continuous measurements of the fine particles (PM2.5) composition were deployed in the Atlanta Supersite Experiment during an intensive study. The instruments measured the aerosol chemical composition at rates ranging from every 5 min to once per hour. The techniques included a filter sampling system with automated water extraction and ion chromatographic (IC) analysis.

During the one-month study carried out by Weber with co-workers,[81] 15-min integrated nitrate concentrations were low, ranging from about 0.1 to 3.5 μg/m^3 with a mean value of 0.5 μg/m^3. Ten-minute integrated sulfate concentrations varied between 0.3 and 40 μg/m^3 with a mean of 14 μg/m^3. The comparison of results obtained with the use of different techniques (including 24-hour integrated filter measurements) revealed a close agreement, within the deviation from 20 to 30% for nitrate (corresponding to the mass concentration ~0.1–0.2 μg/m^3) and 10–15% for sulfate (1–2 μg/m^3). Within 95% confidence intervals, no biases existed between the quasi-continuous techniques and the 24-hour integrated filter measurements of nitrate and sulfate. However, for nitrate, the quasi-continuous intercomparisons showed significantly lower variability than intercomparisons amongst the 24-hour integrated filter measurements.

Carrico et al.[19] examined aerosol radiative and related physical and chemical properties based on measurement data on PM-2.5 (aerosol particles with aerodynamic diameters d_p less than 2.5 μm) in Atlanta city. Arithmetic means and standard deviations of the light scattering by PM-2.5 (σ_{sp} at 530 nm) and absorption coefficients (σ_{ap} at 550 nm) measured at a controlled relative humidity of 49 ± 5% were $[(121 \pm 48) \cdot 10^{-6}]$ and $[(16 \pm 12) \cdot 10^{-6}]$ m^{-1}, respectively. Although the mean light extinction coefficient (σ_{ep}) in Atlanta is much larger than in the background sites, it is comparable to rural areas in the interior southeast United States highlighting the contribution of a regional haze here. The single scattering albedo (ω_0) in Atlanta is 0.87 ± 0.08 thus being ~10% lower than reported in for rural polluted sites, likely due to emissions of elemental carbon (EC) from mobile sources.

A pronounced dual pattern in aerosol properties was observed with clear influences from mobile sources (morning rush hour maxima in concentrations, particularly soot-related indicators) and atmospheric mixing (afternoon minima). A strong linear relation ($r^2 = 0.80$–0.96) between σ_{sp} and PM-2.5 was observed, and the mean-mass scattering efficiency due to elemental carbon (E_{sp}) ranged from 3.5 to 4.4 m^2/g. EC and σ_{ap} were less correlated ($r^2 = 0.19$) than σ_{sp} and PM-2.5.

Four methods of determining the mass absorption efficiency of EC gave E_{ap} ranging from 5.3 to 18.3 m^2/g. That wide range of values was the result of variability in aerosol properties, uncertainties in the light absorption measurements, and in particular, differences in the EC measurement techniques. The best agreement was found for EC mass distributions measured using a multistage impactor in comparison to σ_{ap} calculated with a Mie code yielding $E_{ap} = 9.5 \pm 1.5$ m^2/g, while the EC mass summed from the impactor stages in comparison to measured σ_{ap} gave $E_{ap} = 9.3 \pm 3.2$ m^2/g. Mie light-scattering calculations using, as inputs, the measured mass and EC size distributions gave geometric mean light extinction coefficients and showed the dominance of the fine aerosol particles to light extinction in the urban environment. The mean PM-2.5 value was (31 ± 12) μg/m^3 and the aerosol optical depth in Atlanta measured at the wavelength of 500 nm τ_a was 0.44 ± 0.22. Based on these data and other radiative measurements, the best estimate of the average direct aerosol radiative forcing at the top of the atmosphere (a measure of the climate significance) in Atlanta was $\Delta F = -11 \pm 6$ W/m^2, which was an order of magnitude greater than global mean estimates.

Middlebrook et al.[55] compared four mass spectrometers used in the measurements of the concentrations of minor gases and the chemical composition of aerosol particles during the Atlanta Supersite Project in the central part of Atlanta in order to study the anthropogenic impact on the chemical composition of minor gases and aerosol. Though all four mass spectrometers were generally classified as similar instruments, they clearly had different characteristics due to their unique designs. Some differences in design were present even in two laser desorption/ionization (RSMS-II) mass spectrometers, but despite these differences, the laser-based instruments gave very close results, although some differences were observed with the results obtained with two other mass spectrometers.

Jimenez et al.[38] described the technique of atmospheric aerosol sampling using the Aerodyne Aerosol Mass Spectrometer (AMS) designed to measure size-resolved mass distributions and total mass load of particles from volatile and semivolatile chemical species. The AMS used an aerodynamic lenses to focus the particles into a narrow beam, a roughened cartridge heater to vaporize them under deep vacuum, and a quadrupole mass spectrometer to analyze the vaporized molecules. Particle size was measured via particle time-of-flight. The AMS was operated in two modes: (1) a continuous mass spectrum mode without size information; and (2) a size distribution measurement mode for selected m/z settings of the quadrupole. Individual particles could also be detected and sized if they had enough mass.

In Ref. 38, Jimenez et al. discussed the results of AMS tests carried out in August 1999 at a ground sampling site near downtown area of Atlanta city and at a suburban location in the Boston area during September 1999. The major observed components of the aerosol at both sites were sulfate and organics with a minor fraction of nitrate, consistent with prior studies and collocated instruments. Different aerosol chemical components often had different size distributions and time evolutions. More than half of the sulfate mass contained in 2% of the ambient particles. Trends in mass concentrations of sulfate and nitrate measured with the AMS in Atlanta compared well with those measured with ion chromatography-based instruments. A marked diurnal cycle was observed for aerosol nitrate in Atlanta. The results observed were generalized in the form of an urban aerosol model.

Based on the data on the atmospheric aerosol composition obtained during the 1999 Atlanta Supersite Experiment since August 3 through September 1 of 1999 near downtown area of Atlanta city, Zhang et al.[87] analyzed the assumption of thermodynamic equilibrium between fine particles (PM-2.5) nitrate (NO_3^-) and ammonium (NH_4^+) and gas-phase nitric acid (HNO_3(g)) and ammonia (NH_3(g)). Equilibrium was tested, first by calculating the equilibrium concentrations of HNO_3(g) and NH_3(g) implied by the PM-2.5 inorganic composition (i.e., Na^+, NH_4^+, Cl^-, NO_3^-, and SO_4^{2-}), temperature, and relative humidity observed at the site. These calculated equilibrium concentrations were then compared with the corresponding one gas-phase concentrations observed.

The observed PM-2.5 composition was based on the 5-min averaged measurements of the Georgia Tech PILS, while the observed gas concentrations were based on the measurements borrowed from other sources. The equilibrium gas-phase concentrations were calculated using the ISORROPIA model. The array of combined data on the PM-2.5 chemical composition and HNO_3(g) and NH_3(g) concentration includes 272 five-minute intervals.

Initial calculations using these 272 data points suggested the absence of thermodynamic equilibrium for the case of calculated concentrations of NH_3(g), which are generally lower than the measured ones (for HNO_3(g) quite opposite situation occurred). However, relatively small downward adjustments in the measured PM-2.5 SO_4^{2-} (or apparent acidity) brought the calculated and measured NH_3(g) and HNO_3(g) into agreement. Moreover, excluding 31 of the 272 data points with either anomalously low observed concentrations of SO_4^{2-} or NH_3(g), reasonable adequacy of the SO_4^{2-} correction has been shown. The average relative corrections required for equilibrium with HNO_3 and NH_3 were −14.1% and −13.7%, respectively; significantly larger than the estimated uncertainty arising from random measurement errors.

The results discussed in Ref. 87 suggest that thermodynamic equilibrium does in fact apply to the inorganic PM-2.5 composition during the Atlanta Supersite Experiment and either (1) the PM-2.5 SO_4^{2-} concentration measured by the PILS was systematically overestimated by 15% or (2) the PM-2.5 PILS data systematically underestimated the concentration of the alkaline components by 15%; and/or (3) the ISORROPIA model systematically underestimated the pH of the PM-2.5 encountered during the experiment.

The main conclusion to be drawn based on the analysis of the Atlanta Supersite Project is that (as would be expected) the properties of aerosol in the atmosphere of a big industrial city (in addition, located in the southern latitudes) are extremely different and highly variable. This seriously complicates the account for the aerosol as a climate-forming element. This conclusion is illustrated by the results of various local and regional studies of atmospheric aerosol in North America, which are considered below.

Regional developments

Measurements of light absorption by atmospheric aerosol are of particular interest in the context of estimating the effect of black carbon (BC) aerosol on the formation of climate. This aerosol is a product of incomplete combustion of carbon-containing substances. If the radiative forcing due to purely scattering sulfate and nitrate aerosol gives rise to cooling and, thus, causes partial (sometimes, full) compensation for warming caused by greenhouse gases, then the presence of BC, absorbing radiation in the visible region, favors intensification of radiative warming. Another aspect is the BC effect on the microstructure and optical properties of clouds, which manifests itself, in particular, in the decrease of the cloud albedo.

Arnîtt et al.[11] measured light absorption by black carbon using two instruments: photoacoustic aethalometer and filter-based particle soot absorption photometer (PSAP). Measurements were conducted during the Big Bend Regional Aerosol and Visibility Observational Study at the Big Bend National Park in South Texas. The aethalometer measurements of black carbon concentration at that site correlated reasonably well with photoacoustic measurements of aerosol light absorption. The light absorption coefficient at that site never exceeded 2.1 M/m during the month of collocated measurements. Similar measurements with the use of the same two techniques were also conducted during the Photoacoustic IOP in 2000 at the Department of Energy Southern Great Plains Cloud and Radiation Testbed site (SGP). The PSAP measurements of aerosol light absorption also correlated very well with photoacoustic measurements, but the slope of the correlation indicated that the PSAP values were larger by a factor of 1.61. The photoacoustic measurements of light absorption exhibited a systematic decrease when the *RH* increased above 70%. This decrease in light absorption with *RH* may be due to features of the measurement technique.

Thus, PSAP data should be interpreted with caution under conditions of rapid relative humidity change.

Using the equipment installed aboard a NASA DC-8 aircraft, Clement with co-workers[21] measured the concentrations of atmospheric gases and condensation nuclei (CN) or aerosol in order to study the processes of aerosol production. Analysis of observations revealed that major fluctuations in the CN concentration correlated with gas-phase species, but these were shown to arise due to mixing of two different air masses. Condensation nuclei originated in a storm outflow air mass and CN concentration before mixing was approximately uniform over a flight distance about 200 km. The formation of aerosol by nucleation followed by growth and coagulation was analyzed in Ref. 21 assuming that it consists of water and sulfuric acid produced locally by the oxidation of SO_2. It was concluded that a 5-min burst of nucleation was followed by growth and coagulation over a period of about 5 hours. Both the mass and number concentrations of the observed aerosol correspond to a timescale consistent with that of the storm. The final number concentration was very insensitive to the initial SO_2 concentration.

More than 60 Interagency Monitoring of Protected Visual Environments (IMPROVE) monitoring sites conduct regular filter sampling of atmospheric aerosol for determination of the total carbon (TC), organic carbon (OC), and elemental carbon (EC) concentrations in PM-2.5. The analysis of the results obtained[20] suggested that even in the presence of carbonates in the samples, they do not affect significantly the measured TC, OC, and EC concentrations. This situation is caused by the fact that: 1) carbonates decompose as samples are heated above 800°C; 2) the maximum possible content of carbonates determined from the calcium (Ñà) concentration is almost always negligibly small compared to the TC, OC, and EC concentrations. Measured carbonate carbon concentrations at most sites were <100 ng/m^3, with many less than lower quantifiable limits. The highest carbonate carbon achieved 420 ng/m^3.

Blanchard et al.[13] considered the results of processing the atmospheric filter samples collected during several field campaigns in 1998/1999 from two locations in and around the greater Toronto area in order to investigate the difference in the composition of the organic fraction of atmospheric aerosols at both urban and rural sites in different seasons. The composition of organic particulate matter, with an interest in organic carbon, elemental carbon, alkanoic acids, *n*-alkanes, and polycyclic aromatic hydrocarbons (PAHs), was investigated using thermal desorption, solvent extraction, derivation, and analysis by gas chromatography/mass spectrometry.

The concentrations measured in Ref. 13 were similar to those in previous similar observations. The levels of PM-2.5 were larger in February at both urban and rural sites due to weather conditions favorable for buildup of particles. Organic carbon/elemental carbon ratios were typical of emission values except for the rural site where a greater potential for biogenic-related particles, as well as secondary production, likely played a role.

A comparison of the chemical species with the total organic carbon revealed that the acids, alkanes, and PAHs account for a very small portion of the mass, demonstrating the need for further analytical developments. Preliminary results of the tracer analyses indicated that at the urban site the distribution of alkanoic acids concentration correlated with the diesel emissions. The *n*-alkanes distribution profile was representative of both anthropogenic sources for the urban site and biogenic sources for the rural site. At the rural site, one sample exhibited a local source of atmospheric aerosol derived from vegetation.

In August 2000, two broadband 13- and 6-channel radiometers for measuring direct solar, diffuse, and net radiances have been established on the roof of a building in the Poker Flat Research Range, Alaska (65.12°N, 147.43°E) in order to assess the impacts of ozone, cloud cover, surface albedo, and aerosol conditions on trends in the ultraviolet (UV) radiation reaching the surface. In March–April 2001, a series of measurements of aerosol characteristics was conducted using instruments providing for determination of the aerosol chemical composition depending on the particle size. Wetzel et al.[83] discussed the results observed during invasion of four air mass types to the region under study: three springtime examples, including an Asian air mass with Gobi Desert dust, aged industrial pollution from the Arctic, and humid marine air, and the air mass invaded in late summer.

The analysis of the observations showed that long-range pollutants transport to central Alaska produced only small to moderate increase in the UV optical depth and limited reduction in the atmospheric transmittance at the UV wavelengths. Marine aerosol in a high-humidity environment produced the largest impact on the extinction of the UV radiation. The Angström coefficients and single scattering albedos (calculated from data on the spectral AOD), absorption of light by aerosol, and data on the radiation fluxes showed that these characteristics are quite different for aerosols of different types. This means that even in the cases of low aerosol concentration, an air mass may cause strong variability of the spectral and angular distributions of UV radiation that ought to be accounted for in modeling the photochemical processes in the atmosphere and the biological effect of the UV radiation.

Natural and anthropogenic emissions of minor gases – aerosol precursors and their following transport into the upper troposphere (UT) form the major source of aerosol in the UT. As part of the INCA project on the study of interhemispheric differences in the properties of cirrus clouds due to anthropogenic emissions of minor gaseous components (MGC) two aircraft field campaigns have been performed by Minikin et al[56] to study aerosol and cirrus cloud properties of the upper troposphere in the midlatitudes of the southern hemisphere (SH) and northern hemisphere (NH). From the data obtained in this study a part was analyzed that included the data on the measured profiles of the number densities of volatile Aitken nuclei and of the accumulation mode of aerosol. This analysis was aimed at revealing differences between the two hemispheres. The analysis performed showed that aerosol number concentrations in the NH are more variable than in the SH and typically are a factor of 2 to 3 higher.

Feingold and Morley[25] have demonstrated the ability of a single-wavelength backscatter lidar to provide information on the uptake of water vapor by aerosols in a well-mixed, cloud-capped boundary layer of the atmosphere. Solving this problem is important in analyzing the effect of aerosols on the Earth's radiation budget. An airborne lidar directed in zenith was used to measure vertical profiles of aerosol backscatter beneath stratocumulus clouds. The change in backscatter was analyzed as a function of relative humidity over the range ~85% *RH* to ~98.5% *RH*. *In situ* measurements of the aerosol size distribution and composition are used to calculate the expected enhancement in backscatter due to uptake of the water vapor.

Comparison between lidar backscatter enhancement as a function of *RH* and that derived from the *in situ* aerosol size distribution and composition measurements showed a good agreement. Aerosol backscatter, likely, tended to be higher in downdrafts than in updrafts for the same *RH* range. This is consistent with the concept of inertia of the higher hydrated particles to growth / evaporation on short timescales but may also be due to a bias in the way that lidar-derived cloud base is interpreted in updrafts versus downdrafts. Calculations of enhancement in total scatter due to water vapor uptake with enhancement in the backscatter suggest that the effects agree within ~20% for $RH < \sim 95\%$ but that they differ significantly at $RH > 95\%$.

On the clear-sky assumption, Weaver with co-authors[80] justified the technique of atmospheric correction of retrievals of the vertical temperature and humidity profiles taking into account the presence of mineral (dust) atmospheric aerosol, absorbing in the IR region, from the TIROS Operational Vertical Sounder (TOVS) High-Resolution Infrared Radiation Sounder (HIRS) data. The dust concentrations and properties were defined based on the Goddard Ozone Chemistry Aerosol Radiation Transport (GOCART) model. Every 6 hours the assimilated temperature and moisture profiles were used to forecast the brightness temperature. The observed minus the forecast (O–F) brightness temperature served a measure of the effect of dust aerosol on the radiance parameters.

These calculations indicate that the O–F difference becomes more negative with increasing dust concentrations. This indicated that the dust affects the brightness temperatures. HIRS channels that are sensitive to the surface temperature, lower tropospheric temperature, and moisture are subject to a 0.5 K or more reduction in the brightness temperature under heavy dust conditions. Therefore, the radiative transfer module used in the retrieval system was modified to account for dust assuming illite to be the aerosol substance. Accounting for dust absorption in the retrieval system yields warmer surface temperatures (by about 0.4 K) and warmer lower tropospheric temperatures in the regions of moderate dust loading over the tropical Atlantic.

2. South America

The combined field observational experiments in South America were mostly connected with the problem of ecodynamics of the Amazonian region, including, in particular, developments concerning atmospheric aerosol.[42,43,45]

Tropics, occupying about a half of the Earth's surface, play a key role as a factor of chemical reactions and physical processes in the atmosphere. Vast areas of highly productive tropical land ecosystems are important sources of aerosol and minor gases, affecting the formation of the radiative and the oxidizing ability of the atmosphere. Vast territories occupied by world's largest tropical rain forests lie in the basin of the Amazon River subject to the growing anthropogenic impact. Biomass burning in tropics in the late 1970s – early 1980s led to atmospheric emissions of great amounts of key minor gases, which affected considerably chemical processes in the global atmosphere. Thus, for example, the biomass burning is responsible for 10–30% of the global budget of carbon monoxide, and it also was a significant source of such minor gases as NO_x, CH_4, COS, and Ñ Î$_2$.

In this context, as part of the Large-Scale Biosphere–Atmosphere Experiment in Amazonia (LBA)-Cooperative LBA Airborne Regional Experiment, Krejci with co-workers[47] conducted, in March 1998, airborne measurements (ten flights) of aerosol properties over the northeast Amazonian rain forest in the altitude range between 0.2 and 12.6 km. Analysis of the results obtained showed that the particle number densities in the free troposphere, normalized to standard temperature and pressure, exceeded number densities observed in the boundary layer by a factor of 2 to 15. Typical number densities in the boundary layer were around 500 cm^{-3}. The large difference in particle number density between the boundary layer and the free troposphere agreed with previous observations. Back trajectory calculations showed that even after more than a week of transport in the middle free troposphere, the newly formed ultrafine particles made more than one third of the total aerosol number concentration.

Aerosol size distributions between 0.006 and 3 μm, divided into six vertical layers, showed a typical S-shaped vertical structure with high concentrations of accumulation mode aerosols in the mixing layer and in the free troposphere above 10 km. The lowest concentrations were observed in the free troposphere between 4 and 8 km. Enrichment in accumulation mode aerosol at altitudes above 10 km was attributed to outflow from deep convective clouds. The results obtained suggest that convection plays the most important role in evolution of aerosol and its chemical composition in the atmosphere over tropical rain forests. The further investigations should include the studies of: 1) cloud properties and effect on the vertical aerosol transport; 2) aerosol scavenging and transformations in clouds; 3) production of new particles in air flowing out from the clouds.

As part of the European contribution to the Large-Scale Atmosphere–Biosphere Experiment in Amazonia (LBA-EUSTACH), aerosols were sampled over representative pasture and primary rainforest sites in Rondonia, Brazil[33a] during the transition from dry to raining season and time for biomass burning in September–October 1999 aiming to study atmosphere–biosphere links in this season. Water-soluble organic compounds (WSOCs) within the samples were characterized using a combination of Nuclear Magnetic Resonance (NMR) spectroscopy and Gas Chromatography–Mass Spectrometry (GC–MS).

The NMR analysis indicated that WSOCs were predominantly aliphatic or oxygenated aliphatic compounds (alcohols, carboxylic acids, etc.), with a minor content of aromatic rings carrying carboxylic and phenol groups. Levoglucosan (1,6-anhydro-β-D-glucose), a well known product of cellulose combustion, was the most abundant individual compound identified by GC–MS (0.04–6.90 μg / m^3), accounting to 1 to 6% of the total carbon (TC) and 2–8% of the water-soluble organic carbon (WSOC). Other anhydrous sugars, produced by hemicellulose destruction, were detected in much smaller amounts, in addition to series of acids, hydroxyacids, oxoacids, and polyalcohols (altogether 2–5% of TC, 3–6% of WSOC). Most of these components correlated well with organic carbon, black carbon, and potassium, indicating biomass burning to be their major source.

A series of sugar alcohols (mannitol, arabitol, erythritol) and sugars (glucose, fructose, mannose, galactose, sucrose, trehalose) were identified as part of the natural background aerosol and are probably derived from airborne microbes and other biogenic material. The bulk of the WSOCs (86–91% WSOC) eluded analysis by GC–MS and may be predominantly high-molecular compounds in nature.

Mayol-Bracero et al.[53] performed chemical characterization of carbonaceous aerosols from Rondonia in the Brazilian Amazon region as part of the European contribution to the Large-Scale Biosphere–Atmosphere Experiment in Amazonia (LBA-EUSTACH). The sampling period included the peak of the biomass burning season of 1999, as well as the dry-to-raining season transition. Characterization of the carbonaceous material was performed using a thermal combustion method. This enabled determination of aerosol total carbon (TC), black carbon (BC), and organic carbon (OC). A significant fraction of the BC material (on average about 50%) seemed to be water-soluble organic material.

A more detailed analysis of the water-soluble organic carbon (WSOC) fraction of the TC was made in Ref. 53, involving measurements of WSOC content, high-performance liquid chromatography (HPLC) separation (with UV detection) of the water-soluble components, and characterization of individual components by gas chromatography / mass spectrometry (GC / MS). According to the data obtained, the WSOC fraction accounted for 45–75% of the OC. This high WSOC fraction suggests an aerosol derived mainly from smoldering combustion. Using GC / MS, many different compounds, containing hydroxy, carboxylate, and carbonyl groups, were detected. The fraction of the WSOC identified by GC / MS was about 10%. Three classes of compounds were separated by HPLC / UV: neutral compounds (N), monocarboxylic and dicarboxylic acids (MDA), and polycarboxylic acids (PA). The sum of these three groups accounted for about 70% of the WSOC, with MDA and PA being most abundant (about 50%).

High correlation (correlation coefficient r^2 between 0.84 and 0.99) of BC_{water} (BC after water extraction) and levoglucosan (both indicators of biomass combustion) with the water-soluble species (i.e., WSOC, N, MDA, and PA), and their increase in concentrations during the burning period provided strong evidence that biomass burning is a major source of the WSOC. Particularly interesting is that PA and therefore, probably, humus-like substances (due to their polyacidic nature) are generated in significant amounts during biomass burning. These substances, due to their water solubility and surface tension-lowering effects, may play an important role in determining the overall cloud condensation nuclei activity of aerosols of the biomass burning and, consequently, could be important in cloud processes and climate forcing. In the following investigations, serious attention will be paid to study of the WSOC fraction and its effect on production of cloud condensation nuclei.

Tropical rain forests in the Amazon River basin form the largest tropical forestland on the globe and, probably, the most significant continental source of plant emissions of biogenic trace gases and aerosol. This region is also subject to the most intense deforestation due to forest fires. According to the IPCC data, on the global scale, the biomass burning is the second, in significance, source of anthropogenic aerosol, and tropics are responsible for about 80% of biomass burning (and South America accounts for about one third of this amount).

The conditions of intense convection connected with the dynamics of the intratropical convergence zone favors the transport of natural and pirogenic emissions in the form of trace gases and aerosol into the higher atmosphere and the long-range transport to higher latitudes. In this connection, as a part of the Large-Scale Biosphere–Atmosphere Experiment in Amazonia–European Studies on Trace Gases and Atmospheric Chemistry (LBA-EUSTACH), Guyon et al.[34] collected size-fractionated aerosol samples over a primary rain forest in the Brazilian Amazon during two field campaigns in April–May and September–October of 1999. These two periods encompassed parts of the raining and dry seasons, respectively. Daytime-nighttime-segregated sampling was carried out at three different heights (above, within, and below the canopy level) on a 54-m tall meteorological tower. The samples were analyzed for up to 19 trace elements by X-ray emission analysis and for carbonaceous components by thermal-optical analysis. Equivalent black carbon (BC_e) and gravimetric analyses were also performed.

The average mass concentrations for particles < 2 μm in diameter were 2.2 and 33.5 $\mu g/m^3$ for the raining and dry seasons, respectively. The elements related to biomass burning and soil dust generally exhibited highest concentrations above the canopy and during daytime, while forest-derived aerosol was concentrated primarily underneath the canopy and during nighttime. These variations can be largely attributed to daytime convective mixing and the formation of a shallow nocturnal boundary layer, along with the possibility of enhanced nighttime release of biogenic aerosol particles. The data on mass scattering (σ_s) and mass absorption efficiency (σ_a) indicated that scattering was dominated by fine aerosol, while fine and coarse aerosol both contributed significantly to absorption during both seasons. The analysis of the data also suggested that components other than elemental carbon were responsible for a substantial fraction of the light absorption.

Graf[33] studied the combined interactions between aerosols produced by fires in Amazonian forests and clouds. This aspect of aerosol investigations will be discussed in Part 3 of this review.

A number of local field experiments in South America also played a marked role in investigations of atmospheric aerosol.

Remote sensing of dust aerosol by a Moderate Imaging Spectroradiometer (MODIS) installed aboard the Terra satellite contributed significantly to the Puerto Rico Dust Experiment (PRIDE) conducted in Roosevelt Roads, Puerto Rico from 26 June to 24 July, 2000 to study the radiative and physical properties of Saharan dust aerosol transported into the region. A significant part of the PRIDE program was the direct comparison of aerosol retrievals from the Moderate Imaging Spectroradiometer (MODIS) with Sun photometer and *in situ* aerosol measurements.[50] Over the ocean the MODIS algorithm allows retrieval of the aerosol optical depth (AOD) as well as information on the aerosol size distribution. During PRIDE, AODs derived by MODIS at red wavelengths (near 660 nm) compared closely with data of Sun photometers. However, MODIS-derived AODs were too large at the blue and green wavelengths (470 and 550 nm) and too small in the near infrared (870 nm). Such errors in the AOD spectral dependence resulted in that the retrieved particle sizes were small compared to *in situ* measurements and smaller than those reconstructed from sky radiance measurements with a Sun photometer. The differences in size distributions may be, in part, associated with MODIS' simplification treating the dust as spherical particles.

Using measurement data at two sites in Mexico City and five urban areas in Germany, Baumgardner et al.[12] analyzed the relationship between black carbon (BC) and carbon monoxide (CO). The correlation coefficient between BC and CO was greater than 0.90 for all sites. The average slope of the linear regression line for BC versus CO was 2.2 μg/mg for German sites and 1.1 μg/mg in Mexico City. The most important factors that affect the BC to CO ratio appeared to be the ratio of diesel to gasoline usage and the combustion efficiency of vehicles in a particular area. The results of this analysis suggested that CO measurements in urban areas can be used to estimate BC mass when direct measurements are not available.

3. Western Europe

One of the most informative field observational experiments in Western Europe was conducted in Germany in 1998.

LACE-98. Lindenberg Aerosol Characterization Experiment 1998 (LACE-98) took place between 13 July and 12 August 1998, near Berlin, Germany (52.2°N; 14.1°E) in order to study the aerosol properties from the data of ground-based and airborne (three aircraft) observations. Measurements were performed with one airborne and four ground-based lidars. The meteorological situations in which intensive observations were carried out included clean and polluted air masses characterized by low and high aerosol optical depths. Ansmann et al.[10] overviewed the LACE-98 instrumentation, and illustrated it by some results of observations. A very remarkable finding was the situation on 9–10 August 1998, when a free-troposphere aerosol layer was observed that originated from forest fires in western Canada.

The measurements and calculations obtained were used to estimate the radiative aerosol forcing at the tropopause level, which was found to range from -4 W/m^2 under relatively clean conditions to -16 W/m^2 in the situations with heavy pollution. The processing of lidar data showed that the single scattering albedo was always less than 0.9 (polluted atmosphere) and amounted to 0.8–0.9 in the presence of an aerosol layer, resulting from forest fires in western Canada, in the free troposphere. According to the ground-based observations, the average single scattering albedo was 0.8.

One of the important tasks of LACE-98 was to conduct a "closed" aerosol–radiative experiment. The combined data on the aerosol properties (microstructure and optical parameters) and radiative fluxes (both integral and spectral) enabled a correct comparison to be done between the results of observations and computer simulation. Petzold et al.[61] provided detailed information on the vertical variability of the particle size distribution, the aerosol extinction coefficient, and the hemispheric backscatter fraction, which is applicable for a realistic representation of the aerosol properties in climate models. The estimated daily mean aerosol short-wave radiative forcing varied from -33 ± 12 to -12 ± 5 W/m^2, and the corresponding aerosol optical depths at λ = 710 nm ranged from 0.18 to 0.06.

In the period of LACE-98, Burdke et al.[16] measured the microphysical and optical properties of atmospheric aerosol particles near the ground using different instrumentation (an aerosol photometer, nephelometer, particle soot absorption photometer (PSAP) for measurements of scattering and absorption coefficients; an integrating plate, and a lidar). A telephotometer and a horizontally looking lidar were used to determine the aerosol extinction coefficient under ambient conditions and its dependence on humidity. Reference 16 presents some results of observations. The single scattering albedo ranged from 0.80 to 0.93.

During the LACE-98 Experiment Trimborn with co-workers[77] used a LAMPAS-2 mobile laser mass spectrometer for particles in the airborne state for real-time size-resolved determination of the chemical composition of aerosol particles. The LAMPAS-2 instrument used continuously analyzed size and composition of individual particles from five size ranges with a time course resolution of 1 hour, (overall, several ten thousand single-particle spectra have been recorded). Statistical evaluation of the mass spectral data measured during LACE-98 resulted in the determination of 10 basic classes of particles, yielding detailed information on the aerosol composition. Among them, four classes defined by the presence of mineral, sea salt, and carbonaceous aerosol are most significant.

Schröder et al.[67] analyzed the results of statistical processing of airborne (obtained from onboard a Falcon aircraft) observations of aerosol number density and microstructure conducted in July–August 1998 near Berlin (13.5–14.5°W; 51.5–52.7°N) as a part of LACE-98 experiment. Analysis of vertical profiles of the number density and the associated variability of nuclei mode and larger aerosol particles suggested that the upper troposphere is an efficient source of new particles generation from the precursor-gases. The observed extreme cases of aerosol evolution were considered as caused by a sequence of nucleation processes, formation of the accumulation mode, and the aerosol effect on clouds. This interpretation implies that aerosol sources (nucleation from the gas phase) are closely related to aerosol sinks (cloud scavenging of aerosol particles). Processes that could modify the proposed aerosol life cycle were considered in detail, and data on the number concentration and surface area of aerosol particles of various types were examined. The dry aerosol surface area ranged from 1 to 20 μm^2/cm^3 in the free troposphere and from 2 to 13 μm^2/cm^3 in the tropopause region. Parameterizations of the aerosol size distributions and total surface area concentrations were provided to facilitate the use of the data in atmospheric models. However, the adequate parameterization is possible only based on accumulation of a far larger (more representative) array of observations.

In the period of LÀÑÅ-98, Ebert et al.[23] conducted impactor sampling of aerosol particles in the size range of 0.1 to 25 μm, and the atmospheric concentrations of the chemical elements from sodium to lead ($11 \le Z \le 83$) were determined by total reflection X-ray fluorescence analysis. Approximately 15500 individual particles were examined by high-resolution scanning electron microscopy and X-ray microanalysis, and about 3800 particles were examined by transmission electron microscopy combined with X-ray microanalysis.

On the basis of morphology and chemical composition the particles were classified into 10 different groups: ammonium sulfates, calcium sulfates, sea salt, metal oxides/hydroxides, carbonates, silicates, soot, biological particles, carbon/sulfate mixed particles, and the residue particles enriched with carbon. On the basis of the size distributions and relative abundance of the particle groups, the average and size-resolved complex refractive index of the total aerosol was calculated in Ref. 23. The real part of the average refractive index varied between 1.52 and 1.57 on different sampling days; the average imaginary part varied between 0.031 and 0.057 depending on the amounts of soot and carbon/sulfate mixed particles. The average complex refractive index deduced from the analysis of individual aerosol particles is in a good agreement with the results of photometer measurements of dried filter samples.

As a part of LACE-98, Wandinger et al.[79] conducted simultaneous measurements with ground-based and airborne lidars and with two aircraft equipped with instrumentation for *in situ* measurements of aerosol optical and microphysical parameters. From the lidar measurements, particle backscatter coefficients at up to eight wavelengths between 320 and 1064 nm and particle extinction coefficients at 2–3 wavelengths between 292 and 532 nm were determined. This information allowed the aerosol microstructure to be retrieved. The use of two different inversion algorithms provided the data on the particle effective radius, volume, surface-area, and number concentrations, as well as the complex refractive index. The single-scattering albedo was obtained from Mie-scattering calculations. The results of retrieval were compared with data of direct airborne measurements of particle size distributions and absorption coefficients.

Reference 79 presents two examples of observations. During the night of 9–10 August 1998 measurements were conducted in a biomass-burning aerosol layer in the free troposphere, which was characterized by a particle optical depth of 0.1 at 550 nm. Excellent agreement between remote-sensing and *in situ* measurements was found. In the center of this plume the effective radius was approximately 0.25 μm, and all methods showed rather high complex refractive indices, ranging from 1.56–1.66 in real part and from 0.05–0.07*i* in the imaginary part. The single-scattering albedo showed low values of 0.78–0.83 at 532 nm. The second case, on 11 August 1998, presented typical conditions of a polluted boundary layer in central Europe. Optical depth was 0.35 at 550 nm, and particle effective radii were from 0.1 to 0.2 μm. In contrast to the first case, imaginary parts of the refractive index were below 0.03*i*. Accordingly, the single-scattering albedo ranged within 0.87–0.95.

Wex et al.[84] performed the comparison between measured and calculated aerosol scattering, backscattering, and absorption coefficients based on the *in situ* ground-based and airborne measurements carried out during the Lindenberg Aerosol Characterization Experiment 1998 (LACE-98) field campaign on the study of aerosol properties near Lindenberg (Germany). For the scattering and backscattering coefficients, the agreement between measured and calculated values was on the order of ±20%. It was shown that the calculated particle scattering and backscattering coefficients are very sensitive to sizing uncertainties in the measured size distributions. Measured absorption coefficients were significantly smaller than the corresponding calculated values, and the largest uncertainty in the calculated absorption coefficients resulted from the size-dependent fraction of elemental carbon (EC) of the aerosol. The use of the Monte Carlo method with varying input parameters for calculation of variations in the considered coefficients led to the conclusion that the measured variability of the coefficients was within the range of uncertainties of the calculations, with uncertainties for the calculated scattering and backscattering coefficients being about ±20% and for the absorption coefficients about ±30%. Thus, to increase the accuracy of calculated scattering, backscattering, and absorption coefficients, it is crucial to further reduce the error in particle microstructure measurements. In addition, further improvement of the techniques for measuring absorption coefficients and the fraction of EC of the aerosol is necessary.

The interaction of tropospheric aerosol particles with water vapor affects the tropospheric lifetime of particles and the mechanisms of their removal from the troposphere. The hygroscopic properties of aerosol particles depend on the chemical composition of the water-soluble fraction. These properties control the water content in particles and, finally, the size of individual particles. As a part of LACE-98, Busch et al.[17] measured hygroscopic properties of atmospheric aerosol particles in the Aitken, large, and giant particle size-range in a rural area 80 km southeast of Berlin. Hygroscopic properties of the Aitken particles were determined for four ranges of particle size (0.05, 0.1, 0.15, and 0.250 μm) using a differential mobility analyzer with the relative humidity ranging from 60 to 90%. At 90% relative humidity, aerosol particles could be classified into two groups ("more" and "less" hygroscopic fractions), but at $RH = 60\%$ this classification was impossible.

The measured average growth factors of the “more” hygroscopic fraction were found to be at 1.43, 1.49, 1.56, and 1.63 for the mentioned classes of particles, respectively. Growth factors of the “less” hygroscopic mode were about 1.1 for all particle sizes. The water-soluble volume fraction of the particles was estimated using a hygroscopic model, assuming that the particles consist of ammonium sulfate with an insoluble core. The corresponding estimated water-soluble volume fractions for the “more” hygroscopic particles were about 0.47, 0.52, 0.59, and 0.68, whereas the estimated water-soluble volume fractions of the “less” hygroscopic particles were on the order of 0.1. At $RH = 60\%$, the measured average growth factors were in the range of 1.15–1.22, the estimated water-soluble volume fractions were in the range of 0.41–0.59.

According to the measurements for quasi-single-size particles 0.4–3.8 μm in diameter (with the differentiation into seven specific particle sizes), up to three classes of water-soluble volume fractions are observed. In some cases, a class of nearly insoluble particles corresponding to the less hygroscopic fraction was identified; the second class corresponded to more hygroscopic fraction with a water-soluble volume fraction about 0.5–0.7. The third class of particles with a soluble volume fraction of 0.85 was usually found as well. Below 0.7 μm particle size, the third class of nearly complete water-soluble particles is strongly dominant; above 0.7 μm, there is an equal distribution of all classes. No significant variation was seen during the LACE-98 campaign in the large and giant particle size range.

Intensive measurements of chemical and physical properties of the atmospheric aerosol have been performed at two sites in central Europe during the Melpitz-Intensive (MINT) in November 1997 and the Lindenberg Aerosol Characterization Experiment 1998 (LACE-98) in July and August 1998 in order to study the effect of atmospheric aerosol on the short-wave radiation transfer. The particle size distribution and size-resolved chemical composition of aerosol particles, as well as optical parameters and radiative fluxes (both integral and spectral in the short-wave region) were measured using different ground-based and airborne instruments.

To evaluate the quality of the information obtained, Neusüss et al.[59] compared different methods for obtaining the mass-size distributions of particles. Gravimetric mass of fine particles was attributed completely to carbonaceous material and ions, and to water due to hygroscopic compounds. For the characterization of coarse particles, which contribute less to the total mass concentration, insoluble material has to be included to the mass balance. Mass concentrations calculated from the particle-size distributions well correlate with the gravimetric mass concentrations; however, the calculated mass is larger, especially for the Aitken and accumulation modes. The mass concentration calculated from the microstructure is most sensitive to the sizing uncertainty of the measured size distribution. The overall uncertainty of the calculated mass was found to be about 10%.

For different types of air masses, Ref. 59 presents lognormal parameters of the size-distributions for the number density, volume, and particle mass. Most of the modal parameters in different air mass types do not differ significantly. Higher mass concentrations are mostly due to an increase in size (of Aitken and accumulation mode) rather than increase in the number of particles of a given mode. Generally, the fraction of carbon mass in the particle mass increases with decreasing particle size. The most pronounced feature of the annual behavior is the increase of carbon content from summer to winter as well as the increase in nitrate content, resulting in a decrease of sulfate. For nitrate, a strong dependence on the direction of air mass invasion is observed. Sulfate and nitrate are predominantly neutralized by ammonium.

In addition to the large-scale LACE-98 experiment, numerous other field studies of atmospheric aerosol have been carried out in Europe.

During the four powerful aerosol pollution outbreaks (plumes at the altitudes above 500 m) taking place in the period from June 16 to July 25 1997 from the European continent out over the Atlantic Ocean, Müller et al.[57] performed lidar sensing in order to retrieve the vertical profiles of physical particle properties and the single-scattering albedo at 532 nm. The results of six-wavelength aerosol lidar observations near Sagres (37°N, 9°W), Portugal, in the framework of Aerosol Characterization Experiment 2 (ACE 2) (June–July 1997) were used to find the vertical profiles of the backscattering coefficient at the wavelengths of 355, 400, 532, 710, 800, and 1064 nm, as well as the extinction coefficient at the wavelengths of 355 and 532 nm.

According to the results obtained, particle effective radii were $0.15 \pm 0.06\,\mu m$, volume concentrations ranged from 6 to 27 $\mu m^3/cm^3$, and surface area concentrations were 80 to 1200 $\mu m^2/cm^3$. The mean imaginary part of the wavelength-independent complex refractive index was $0.009i \pm 0.010i$, that is, the particles in general showed negligible absorption. The mean real part was 1.56 ± 0.07, and the mean value of 0.95 ± 0.06 was obtained for the single-scattering albedo at 532 nm. The results indicate that the major contributor to the observed pollution was nonabsorbing ammonium-sulfate-like material, and only a very small fraction was contributed by absorbing soot-like material. Correlation analysis showed that the effective radius was well correlated with the Angström coefficient. Furthermore, correlations of the single-scattering albedo with the particle extinction-to-backscatter ratio and with the imaginary part of the complex refractive index were found. The similarity of the results obtained from the data of the TARFOX field observational experiment indicates that the processes of internal combustion, being aerosol sources, in Europe differ insignificantly from those in North America. However, the differences with INDOEX and SCAR-B data appeared to be considerable, which was caused, in the first case, by production of carbon compounds due to coal combustion, diesel engine functioning and/or biomass burning and, in the second case, by biomass burning.

Within the Global Atmosphere Watch (GAW) Aerosol Program of the World Meteorological Organization (WMO), the aerosol chemical composition has been continuously measured for 1.5 year since July 1999 at the Jungfraujoch (JFJ) observatory (3580 m asl) by Henning et al.[36] Sampling was performed in two size classes (all particles and particles with aerodynamic diameters smaller than 1 μm). The filters were analyzed chromatographically for major ions, which constituted 30% of the total dry aerosol mass collected. As the annual mean, a total ion mass concentration of 1.04 $\mu g/m^3$ was observed. Sulfate, ammonium, and nitrate were the major components of the fine aerosol fraction, while calcium and nitrate were two major water-soluble components in the coarse mode. Single particle analysis confirmed the internal mixture of calcium and nitrate in the coarse mode. The total ion mass concentration showed strong seasonal differences, with 1.25, 1.62, 0.70, and 0.25 $\mu g/m^3$ for spring, summer, fall, and winter, respectively. The variability was stronger for sulfate, ammonium, and nitrate than for calcium. The reason for this was believed to be local sources of calcium, along with Saharan dust episodes, which occasionally happen over the whole year, independent of season.

In spite of the intense efforts in the evaluation of air quality, many problems are still to be solved, which is mostly caused by the insufficiently combined character of the developments. Therefore, Vautard with co-workers[78] carried out the Atmospheric Pollution Over the Paris Area (ESQUIF) experiment, whose major objective was a thorough study of atmospheric pollution in the large urban area of Paris with the following use of the observations to check the adequacy of air quality models, reconstructing the atmospheric pollution due to photooxidants.

The ESQUIF involved the studies of: 1) sensitivity of the processes of surface ozone production to various factors (long-range transport, levels of urban emissions, ratio of anthropogenic to biogenic emissions); 2) adequacy of data on inventory of emissions ($NO_{\tilde{o}}$, CO, VOC, etc.); 3) cloud and aerosol effect on the rate of photolysis; 4) role of photochemical processes in episodes of enhanced NO_2 concentration in winter; 5) ways of improving measurement and simulation techniques; 6) specificity of spatiotemporal variability of aerosol in the region of Paris.

In the period of 1998–2000, a series of measurements were conducted in different areas of the city of Paris, the main part of which was represented by 12 intensive observation periods (IOĐs) mostly in summer. The conditions of observation were characterized by a wide variety of meteorological situations, especially, those, when the strong pollution of the atmosphere over Paris and its suburbs occurred. The system of observations included circular flights around the city in combination with the direct *in situ* measurements and remote sensing. The combined observations allowed reconstruction to be done of the 3D evolution of the pollution (in general, the Paris atmosphere cannot be considered as strongly polluted).

In Ref. 78, the observations for different IOĐs are compared from the viewpoint of revealing the specificity of different atmospheric pollution events, including, in particular, assessment of the role of long-range transport. In addition, the observations are compared with the results of computer simulation (using the techniques of conjugate equations and Monte Carlo). The goal of this comparison was to evaluate the role of correct account for the boundary conditions, setting the emissions of atmospheric pollutants (including biogenic ones), and correct reconstruction of photolysis processes.

The analysis of observations during IOĐs showed, in particular, that the episodes of the atmospheric pollution occur very quickly under conditions of stagnant atmosphere. In these cases, the oxidant concentrations peaked at the high or low altitude of the boundary layer and even under conditions with no marked temperature inversions. The highest peaks of pollution were formed as a consequence of local pollution under the meteorological conditions favoring the accumulation of pollution, but in some cases the long-range transport also contributed considerably.

Chemical analysis of urban aerosol samples collected at Gent, Belgium, conducted by Kubátova et al.[48] using the techniques of gas chromatography and mass-spectrometry revealed the presence of about 100 individual organic compounds. Aerosol samples from winter and summer campaigns were analyzed in order to examine seasonal differences. The major part of the organic carbon in both winter and summer aerosols was found to be from automobile exhausts. Among the prevailing organic compounds resolved in both seasons, there were *n*-alkanes and fatty acids, but the distribution patterns of individual components within each class showed seasonal differences. The *n*-alkane pattern for summer aerosols was clearly affected by emissions from the vegetation, while that of fatty acids revealed a lower relative abundance of unsaturated fatty acids in summer than in winter that can be related to more intense atmospheric oxidation of unsaturated fatty acids during summer. Concentrations of dicarboxylic acids and related products that are believed to be the products of hydrocarbons and fatty acids oxidation were the highest in summer aerosols. Some individual compounds in the latter class could only be detected in summer samples and showed the highest concentrations on hot summer days that were characterized by maximum temperatures above 25°C and increased surface ozone concentrations. The latter compounds included novel, recently identified glutaric acid derivatives, namely, 3-isopropyl and 3-acetyl pentanedioic acid, likely originating from atmospheric oxidation of reactive monoterpene or sesquiterpene precursors that still need to be identified. Several compounds were found that are markers of wood combustion, including diterpenoic acids, lignin pyrolysis products, and levoglucosan. The quantitative results obtained for diterpenoic acids and lignin pyrolysis products indicated that contributions from wood combustion are more important in winter than in summer. The data obtained evidenced that both softwood and hardwood burning contribute to the organic aerosol, but burning of hardwood prevails during winter. Polyaromatic hydrocarbons were also prevail in winter.

De Tomasi et al.[22] discussed the results of retrieval of vertical profiles of water vapor and aerosol concentration using regular monitoring with a combined Rayleigh-Raman lidar based on a XeF excimer laser (351 nm). This lidar installed in southern Italy (40°20′N, 18°06′E) functioned as a part of the European Aerosol Research Lidar Network. Synchronous measurements of vertical profiles of the aerosol backscattering ratio $R(z)$, the aerosol extinction coefficient $\sigma_{\lambda_0}^{\text{aer}}(z)$, and the water vapor mixing ratio $w(z)$ have been performed during several months to investigate the correlation of $R(z)$ and $\sigma_{\lambda_0}^{\text{aer}}(z)$ with water vapor and to characterize the aerosol optical properties. A strong correlation has been found between the spatial and temporal evolution of $R(z)$ and $w(z)$ both in summer and in autumn. The experimental results have revealed that $R(z)$ increased with increasing $w(z)$ and that the aerosol and water vapor content were lower in autumn than in summer. The analysis of the data by use of the 4-day analytical backward trajectories has revealed that the dependence of $\sigma_{\lambda_0}^{\text{aer}}(z)$ and $R(z)$ on $w(z)$ was affected by the typical advection patterns over the lidar site. A growth of the contribution from aerosol to backscattering ratio and extinction coefficient as $w(z)$ increases have been observed when air masses came from northern and eastern European countries. Under these conditions, the average values of the lidar ratio S increased to 50–63 sr. Average S values ranging from 48 to 74 sr have been retrieved for air masses advected from North Africa.

Kiss with co-workers[41] collected atmospheric fine aerosol ($d_p < 1.5$ μm) at a rural site in Hungary from January to September 2000. The analysis of these samples showed that the total carbon concentration ranged from 5 to 13 $\mu g/m^3$ and from 3 to 6 $\mu g/m^3$ in the first three months and the rest of the sampling period, respectively. On average, water-soluble organic carbon (WSOC) accounted for 66% of the total carbon concentration independent of season. A variable fraction of the water-soluble organic constituents (38–72% of WSOC depending on the sample) was separated from inorganic ions and isolated in pure organic form by using solid phase extraction on a copolymer sorbent. This fraction was experimentally characterized by an organic matter to organic carbon mass ratio of 1.9, and this value did not change with the change of seasons. Furthermore, the average elemental composition (molar ratio) of C:H:N:O ≈ 24:34:1:14 of the isolated fraction indicated the predominance of oxygenated functional groups, and the low hydrogen to carbon ratio implied the presence of unsaturated or polyconjugated structures.

Such conclusions were confirmed by UV, fluorescence, and Fourier transform infrared (FTIR) studies. On the basis of theoretical considerations, the organic matter to organic carbon mass ratio was estimated to be 2.3 for the nonisolated water-soluble organic fraction, resulting in an overall ratio of 2.1 for the WSOC. In order to extend the scope of this estimation to the total organic carbon, which is usually required in mass closure calculations, the aqueous extraction was followed by sequential extraction with acetone and 0.01 M NaOH solution. As a result, a total organic matter to total organic carbon mass ratio was estimated to be of 1.9–2.0.

Based on analysis of aerosol samples collected at Puy de Dome (1465 m above sea level, France) in 2000–2001, Sellegri et al.[68] investigated the dependence of the aerosol chemical composition on the particle size. Aerosols have been sampled using low-pressure cascade impactors and analyzed for inorganic (Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^-, and SO_4^{2-}) and organic ($HCOO^-$, CH_3COO^-, and $C_2O_4^{2-}$) ions, organic and elemental carbon (OC and EC), insoluble dust, and total mass. Under cloudy conditions, the sampling included aerosol particles in side the clouds. The residue of evaporated cloud droplets was also analyzed.

The results obtained correspond to air masses, which can be classified into three different categories according to their aerosol load and composition: background (BG), anthropogenic (ANT), and specific events (EV) that include advection of Saharan dust and upward transport from the polluted boundary layer. Depending on the presence or absence of coarse sea-salt particles, further classification permitted distinguishing between air masses that have or have not been over the ocean. A closed mass balance was achieved on submicron ranges (within the error of 18.5%) for the three main air mass categories, providing a reliable description of main aerosol types in the west European free troposphere. The total aerosol mass at 50% relative humidity was close to 2.7 ± 0.6 $\mu g/m^3$ in BG, 5.3 ± 1.0 $\mu g/m^3$ in ANT, and 15 to 22 $\mu g/m^3$ in EV air masses. The aerosol mass distribution generally exhibited two submicron modes (Acc1 at 0.2 ± 0.1 μm and Acc2 at 0.5 ± 0.2 μm geometric mean diameter) and a supermicron mode (2 ± 1 μm).

Aerosols exhibited a high degree of external mixing with carbonaceous (EC and OC) and ion species associated with Acc1 and Acc2. Concentrations of light carboxylates and mineral dust never exceeded 4% of the total content of analyzed compounds, except for the Saharan dust event, during which the contribution of insoluble dust reached 26% of the total aerosol mass. Depending on the sampled air mass, bulk water-soluble inorganic species accounted for 25–70% of the total mass. The OC fraction was higher in air masses with low aerosol load (53%, 32%, and 22% for BG, ANT, and EV, respectively). Conversely, the EC fraction was enhanced from 4% in BG to 10% in ANT and 14% in EV. The inorganic fraction was more abundant in EV (55%) and ANT (60%) than in BG (40%) air masses because of the enhanced nitrate and, to a lower degree, of the sulfate and ammonium content.

Organic carbon observed in the atmosphere is produced by several sources, including emissions of organic carbon due to combustion of fossil fuel, biomass, and gaseous products of the processes of photochemical oxidation of both anthropogenic and biogenic origin. Since chemical composition of the atmospheric aerosol is an important indicator of the processes of organic carbon production, Alves et al.[9] performed analysis of aerosol samples collected in two Portuguese urban areas (Lisbon and Aveiro) and in a Finnish forested site. The particulate matter extract was separated by flame chromatography into aliphatics, aromatics, carbonyls, alcohols, and an acidic fraction, and analyzed by gas chromatography–mass spectrometry. An organic / black carbon analyzer was used to evaluate the carbonaceous matter, the water-soluble fraction, solvent extractable material, and the content of different organic classes.

Results obtained in Ref. 9 showed that extraction with dichloromethane was able to dissolve less than 50% of the particulate organic material, but the use of water, as a solvent, enabled one to dissolve from 70% to 90% of organic carbon. Normally more than 70% of the extracted material comprise organic compounds, among which dominate acids and alcohols, especially for particles with diameters less than 0.49 μm. The organic compounds identified were represented by primary compounds of both anthropogenic and biogenic origin. The latter ones are mainly from vegetation waxes and from petrogenic sources, and secondary products resulting from the oxidation of volatile organic compounds. The water-soluble fraction contains oxocarboxylic and dicarboxylic acids, and cellulosic constituents.

As to the anthropogenic components, nonpolar fractions constituted up to 24% of the extracted organic carbon in Lisbon and presented high levels of petroleum markers, while in the forest station these compounds made only 8%. The oxidized organic compounds accounted for 76–92% of the extracted carbon in samples from Aveiro and Finland. Owing to favorable photochemical conditions during the sampling campaign, secondary organic constituents presented higher levels in Aveiro. The forested site was the one where the water-soluble components showed greater variability and concentrations.

With the instruments installed aboard a C-130 research aircraft owned by the UK Met Office, Formenti with co-workers[26] performed airborne measurements of sulfur dioxide and aerosol chemical/microphysical properties over the Aegean Sea in August 1998. The SO_2 mixing ratios up to 18 ppb and aerosol sulfate levels of up to 500 ppt were measured near Thessaloniki. The highest concentrations were observed at altitudes between 1 and 2 km, while near the surface much lower SO_2 mixing ratios between 1 and 4 ppb were found. The observations showed that pollution was transported southward over the Aegean Sea as far to the south as Crete, where SO_2 mixing ratios of up to 5 ppb level were observed. The combined results of the SO_2, aerosol sulfate, and particle size distribution measurements indicated air masses containing emissions with different ages, which range from a few hours to several days.

Pollution due to fossil fuel burning observed over northern Greece was transported from Eastern Europe (Bulgaria, Romania, and Turkey). Additionally, the measurements indicated significant contributions from forest fires, in particular in the haze layers that formed ten days earlier due to fires over the Northwestern Canadian territories, that had crossed the Atlantic and passed over Europe. At the altitudes above 1 km, CO layers of 1 to 2-km depth were observed; often they had a double-layer structure with a secondary CO layer above 3.5 km. The estimates showed that the contribution of biomass burning to the CO measured over Greece was four times higher than that of fossil fuel. The lower layer had a significant number of particles N_p in the accumulation mode (size range 0.1–1 μm) at the ratio N_p/CO of the order of 2–6 cm^{-3}/ppb, while in the upper tropospheric layers N_p decreases due to convection and wet scavenging. The Aitken particle mode (size range 0.005–0.1 μm) was decreased both in the lower and in the upper layers. This suggests that the Aitken particles originally present in the lower layers had already grown to the accumulation mode size when detected.

Chemical, physical, and optical measurements of aerosol particle properties within an aged biomass-burning plume were performed on board a C-130 research aircraft during a descent profiling over a ground-based site in the northeastern Greece (40°24′N, 23°57′E), where continuous measurements of the spectral downwelling solar short-wave irradiance (global, direct, and diffuse) have been being made.[27] The aerosol originated from biomass burning in the forest fires in Northwestern Canada and then transported over the Atlantic Ocean.

According to the ground-based measurements during the time of that flight, the aerosol optical depth was significantly enhanced (0.39 at the wavelength of 500 nm) due to a haze layer between 1 and 3.5 km altitude. The dry particle scattering coefficient within the layer was around 80 M/m (1 Ì = 10^{-6} m^{-1}), and the particle absorption coefficient was around 15 M/m, giving a single scattering albedo of 0.89 at 500 nm (dry state). The black carbon fraction was estimated to account for 6–9% of the total accumulation mode particle mass (< 1 μm diameter). The increase of the particle scattering coefficient with increasing relative humidity at 500 nm was of the order of 40% for the relative humidity varying from 30 to 80%. With the dry, altitude-dependent, particle size-distribution function and optical properties used as input parameters, the radiative transfer was calculated for the spectral short-wave, downwelling irradiance at the surface. The agreement between the calculated irradiances and the experimental results from the ground-based radiometer was within 10%, both for the direct and diffuse components (at 415, 501, and 615 nm). Calculations of the net radiative forcing at the surface and at the top of the atmosphere (TOA) showed that due to particle absorption the effect of aerosols was much stronger at the surface than at the TOA. Over the sea, the net short-wave radiative forcing (daytime average) between 280 nm and 4 μm was up to −64 W/m^2 at the surface and up to −22 W/m^2 at the TOA. Thus, the radiative forcing at the surface is roughly threefold as large as that at the TOA, which agrees with the INDOEX results.

Johansen and Hoffmann[39] discussed the results obtained from analysis of the atmospheric aerosol samples for labile Fe(II) and other trace metals. The ambient aerosol samples were collected over the Arabian Sea in March of 1997, aboard *Sonne* German research vessel, as part of the Joint Global Ocean Flux Study (JGOFS) Project. Earlier analogous measurements have been conducted over the Arabian Sea during two different seasons. The samples were analyzed for ferrous iron immediately after collection, while trace metals, anions, and cations were determined upon return to the laboratory.

The main crustal component of the aerosol was governed by the average crustal composition in the region and amounted to 5.94 ± 3.08 μg / m^3. An additional crustal constituent of clay-like character, rich in water-soluble Ca^{2+} and Mg^{2+}, was seen in the fine fraction in air masses of the Arabian origin. Total ferrous iron concentrations varied from 3.9 to 17.2 ng / m^3 and averaged to 9.8 ± 3.4 ng / m^3, with (87.2 ± 3.37)% of Fe(II) present in the fine aerosol fraction. The Fe(II) concentrations accounted, on the average, for 1.3 ± 0.5% of the total Fe. The anthropogenic contribution to the aerosol, as traced by Pb, Zn, and some anions and cations, was found to be considerably larger, especially during the first 10 days of this cruise, than in the samples collected previously during the time from the inter-monsoon and southwest monsoon of 1995.

4. High latitudes

The investigations of aerosol in the air of high latitudes attract considerable attention for a long time, and the corresponding results were widely discussed in the literature, including recent monographs.[14,44] Therefore, here we restrict our discussion only to some comments.

Based on the data of airborne impactor and filter observations of the properties of atmospheric aerosol in the Arctic troposphere, obtained during Arctic Study of the Tropospheric Aerosol and Radiation (ASTAR) campaign in March–April 2000 in the region of Spitzbergen (Svalbard), Hara with colleagues[35] analyzed the mixing states of aerosol constituents. The processing of observations showed that sulfate and soot were major aerosol components under both Arctic haze and background conditions, and sea-salts are major only in the lower troposphere (< 3 km above sea level). Mineral / dusts and unknown species were obtained as minor constituents.

Airborne aerosol measurements have been carried out under the Arctic haze (23 March), and aerosol-enriched (20 March and 12 April) conditions. The highest relative abundance of soot (~94.7%) was observed in the free troposphere on March 23, when the heaviest Arctic haze condition was transported directly from Russian industrial regions to the area under study for several days. Analysis of the aerosol samples suggested that whereas the external mixing of soot and sulfate dominated under the Arctic haze and aerosol enriched conditions, the internal mixing dominated under the background conditions. On the other hand, most of the aerosol particles containing sulfate had the external mixing states with soot and other aerosol constituents in the free troposphere under both the Arctic haze and background conditions. Sea-salts were dominant only in the lower troposphere (sometimes sea-salt particles were modified (Cl^- content dropped down) in the lower troposphere (< 3 km)), although a few sea-salt particles were observed in the middle-upper troposphere (3–7 km).

The chemical composition of atmospheric aerosol in many regions over the globe (including urban, maritime, and polar atmospheres) is characterized by the presence of homologous series of low molecular weight (LMW) dicarboxylic acids, which are produced due to combustion of fossil fuels and biomass, as well as in the processes of photochemical oxidation of both anthropogenic and biogenic organic precursors. Nowadays LMW are considered as significant components of water-soluble organic aerosol. Their important function is that particles of such aerosol can serve cloud condensation nuclei and, thus, exert the direct and indirect influence on the formation of the aerosol radiative forcing. Recent observations showed that during polar sunrise the concentration of LMW in the atmosphere increases drastically (5 to 20 times), and this increase can be interpreted as a consequence of photochemical formation of dicarboxylic acids through oxidation of their anthropogenic precursors coming to the Arctic atmosphere from middle and high latitudes. If the volatile organic compounds are the main precursors, then it is natural to suppose possible important role of gas-phase processes in the formation of the LMW in submicron aerosol particles. Another one possibility is caused by heterogeneous reactions as a mechanism of LMW formation on supermicron particles. However, the observations of the particle size distributions of acid aerosol in the Arctic atmosphere are still absent.

To obtain such data, Narukawa et al.[58] collected samples of fine ($d_p < 1$ μm) and coarse ($d_p > 1$ μm) aerosol particles at Alert, Canada (82°27′N, 62°30′W) in the period of polar sunrise since March 29 through April 14 of 1997, as a part of the Polar Sunrise Experiment (PSE-1997). Samples were analyzed for low molecular weight dicarboxylic acids (C_2–C_{11}) using gas chromatography with flame ionization detector (GC–FID) and GC / mass spectrometry (GC / MS).

The results of the analysis showed that more than 80% of the LMW were concentrated in the fine fraction, suggesting the production by gas-to-particle conversion in the Arctic. In both fractions, oxalic acid was the dominant LMW species followed by succinic and malonic acids. Shorter chain dicarboxylic acids (C_2–C_5) showed the concentration maximum on 5–7 April; however, longer chain dicarboxylic acids (> C_6) did not show a clear peak in the same periods.

A significant depletion of ozone was observed during the experiment, where an anticorrelation was found between the concentrations of the ozone and shorter chain dicarboxylic acids (C_2–C_5) in both fine and coarse aerosols. The enhanced concentration of filterable bromine was also observed in both modes. Peaks of dicarboxylic acids in both coarse and fine aerosols during ozone depletion events indicate that heterogeneous reactions occur on the coarse particles and possibly on fine particles as well. Dicarboxylic acids may be produced by the oxidation of precursor compounds such as glyoxal and glyoxylic and other ω-oxocarboxylic acids that contain aldehyde (hydrated form) group, being involved with ozone and halogen chemistry in the Arctic marine boundary layer.

In the periods of January 5–20, 2000 and January 1–26 2001, Koponen with co-workers[46] measured the total particle number concentrations and particle size distributions of atmospheric aerosol at Antarctic research station Aboa (73°03′S; 13°25′W), situated about 130 km off the coastline at the altitude of 480 m above sea level. During the above periods of the austral summers, the observed total particle number density varied between about 200 and 2000 cm^{-3} for particles with the diameter ranging within 0.003–0.8 μm. On the average, the concentrations in marine/coastal air masses were higher than in the continental air masses. Concentrations in excess of 300–400 cm^{-3} were attributed to the presence of small (< 0.02 μm) nucleation mode particles.

Measured size distributions were fitted with two to four lognormal modes. All the spectra displayed an accumulation mode peaking at about 0.07–0.15 μm and an Aitken mode peaking at about 0.03–0.05 μm. The average mean diameter of the accumulation mode was much lower in continental air masses, whereas that of the Aitken mode was almost the same for the two air mass types. A nucleation mode was present in more than a half of the measured spectra, and occasionally two nucleation modes could be identified. Evidence on recent new particle formation with subsequent particle growth up to about 0.04 μm was observed in the marine/coastal air masses only. The particle growth in such cases occurred with the rate of 1–3 nm/h, and the particle size several times exceeded that corresponding to the Aitken mode. This means that climatically important Aitken and accumulation mode particles can be produced not only in the middle and upper troposphere, where the conditions are favorable for their continuous growth, but also in the boundary layer.

The results discussed in Ref. 46 demonstrated that the newly formed particles contribute significantly to the overall particle budget of the Antarctic boundary layer, and they can also be a source of climatically important Aitken and accumulation mode particles. However, it is still unclear whether the new particles are really produced. In air masses formed at the center of continents, the nucleation mode was not observed. In addition, it was noticed very rarely in the remote marine boundary layer over the open ocean. On the other hand, the presence of the nucleation mode was usually seen in shipborne measurements if approaching the Antarctic coast. Consequently, the production of new particles may be attributed to coastal processes. This property can be determined by the meteorological conditions of the coastal zone or by emissions of minor gases in pack-ice regions. Further investigations are needed to answer these questions.

Conclusions

The considered results of investigations into the aerosol properties in different regions of America and Western Europe, obtained within the framework of field observational experiments, have demonstrated once again a very wide variety of physical properties and chemical composition of aerosol, which call for further systematization of observations in order to justify more adequate, than existing, aerosol models. Only the use of such models can permit obtaining more reliable estimates of the possible effect of aerosol on climate, air quality, and human health.

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