Aerosol and climate studies: current state and prospects. 2. ACE-Asia field observational experiment

K.Ya. Kondratyev

Scientific Research Institute Center for Ecological Safety, Russian Academy of Sciences, St. Petersburg
Nansen International Environmental and Remote Sensing Foundation, St. Petersburg

Received September 16, 2005

The results of complex studies of atmospheric aerosol and its conversion in the process of long-range transport are discussed; the studies are performed as part of the observational field experiment ACE-Asia.

Introduction

Naturally, (with the exclusive importance of numerical modeling) the understanding of real climate change relies heavily on analysis of observation data, especially data of regional complex observational field experiments. In the context of aerosol–climate problem, an important recent milestone has been the publication of thematic issues of the journal *Atmospheric and Oceanic Optics* devoted to developments under the "Aerosols of Siberia" Program. In this regard, of special importance are the results of implementation of field observation program Aerodyne Characterization Experiment-Asia (ACE-Asia), devoted to studies of atmospheric aerosol and aerosol effects in the regions of South-East Asia and North-Western Pacific; this research has been performed as part of ACE program of field experiments aimed at studying aerosol properties (other analogous programs were discussed in Ref. 18). Analysis of results obtained during ACE-Asia project makes up the primary contents of this overview; however, before discussion of this problem, it is necessary to briefly mention some other aspects of analogous developments that have made up the context for ACE-Asia. These developments cover almost the entire wide range of problems, from the processes associated with formation of individual aerosol particles and evolution of their properties to the dynamics of global aerosol distribution.

In 2002–2003, Texas A&M University, College Station, Texas, regularly exploited mass spectrometer, designed for measurements of the chemical composition of separate particles of ultrafine atmospheric aerosol of different sizes. In December–March, this site performed three series of observations separated in time by 6 weeks. A total of 128 thousands of mass spectra were recorded for particles with aerodynamic diameter from 30 to 300 nm, and these spectra were then subjected to subsequent classification.

Glagolenko and Phares[9] have identified and described 15 statistically significant classes. The most frequently observed species were ions of potassium nitrate, carbon nitrate, and silicon nitrate/oxide. The presence of nitrate in most particles can be explained by the effect of agricultural activity near the observation site. The frequency of detection of nitrate correlates with the near-ground air temperature and relative humidity. Another particle class, identified as amine, is characterized by the presence of strong dependence on relative humidity, though manifested only under conditions of low relative humidity. Seemingly, some of the recorded particles formed in big cities were then coated with film of nitrate, sulfate, and organic species in the process of long-range transport.

Using two Tandem Differential Mobility Analyzers (TDMAs), on August 9–12, 2002, at the College Station, Texas, Santarpia et al.[32] performed measurements of mobility of charged aerosol particles for analysis of the state of aerosol hydration as a function of particle size. In the process of observations the aerosol was moistened in air with the relative humidity $RH = 80\%$ before and soon after sunrise (RH in the atmosphere ranged from 50 to 70%). Atmospheric aerosol particles in the size range from 160 to 320 nm were studied. Analysis of measurement results has shown that the hygroscopic particle growth was below the level calculated for such inorganic compounds as ammonium sulfate, suggesting that the particles were composed of mixture of soluble and insoluble components. Those particles that were characterized by the presence of hysteresis of hygroscopic growth were almost always metastable aqueous solutions.

Analysis of data of observations of non-sea-salt (nss) sulfate aerosol in remote (background) marine boundary layer (RMBL) of the atmosphere indicates that a substantial fraction of nss aerosol may be in the form of coarse (supermicron) sea-salt aerosol. The nss aerosol formation can only partly be explained by mechanisms taking into account the influence of aqueous phase. Sievering at al.[39] have processed samples of atmospheric aerosol collected at Baring Head, New Zealand, under clear-sky and normal cloudiness conditions; these results have made it possible to estimate the contribution of oxidation

0235-6880/06/05 333-18 $02.00

mechanism, caused by near-ground ozone (O_3), to formation of coarse fraction of nss particles.

The observation data suggest that the mass concentration of coarse aerosol under clear-sky conditions was higher than 8 nmol nss/m^3, while under average cloud conditions it was less than 3 nmol nss/m^3. These data were obtained for strong wind (~11 m/s) conditions, minimizing the lifetime of coarse aerosol and, hence, the possibility of its interaction with clouds. In all cases, the aerosol composition was characterized by excessive calcium (Ca) content, 200 times higher than normal, and, correspondingly, by a factor of 30–40 higher alkalinity of aerosol than that of the bulk seawater. These conditions enhanced aqueous-phase, O_3-caused oxidation, which may account for the presence of coarse fraction of non-sea-salt aerosol.

Data of satellite remote sensing indicate that Southern Hemisphere ocean, upwind of the Baring Head, has approximately three times higher annual primary productivity (with corresponding enhancement of biogenic Ca content) than the open ocean surface water. The presence of powerful source of biogenic Ca and strong wind determine anomalous character of RMBL in the region of Baring Head. Shipboard observations under conditions of more typical RMBL in western Pacific of the Southern Hemisphere indicate that marine biogenic sources may account for only from ~1 to ~2.5 times higher alkalinity of sea-salt aerosol (in comparison with bulk seawater). Coarse aerosols typically experience intense deposition onto the ocean surface, resulting in rapid recycling of ocean-derived sulfur. However, earlier models overlooked this process of sulfur budget in the RMBL. One serious consequence of this is inadequate treatment of the contribution of biogenically derived oceanic sulfur of RMBL to new particle formation, because ozone-caused oxidation in sea-salt aerosol particles turns out to be energy-wise more favorable than the homogeneous nucleation.

Biomass burning has significant regional and global effect on chemical processes in the troposphere. The existing estimates suggest that the biomass burning contributes about 15 to 30% to global CO budget, it, correspondingly, contributes ~9% to NO_x and ~14–18% to O_3 budgets. On the global scale, the biomass burning is a dominating source of organic carbonaceous (OC) aerosol, and has approximately two times larger contribution to OC budget than that from emissions due to fossil fuel combustion. The role of the two sources is approximately identical in the case of carbon black (CB) aerosol. It is also important that in the process of biomass burning, different greenhouse gases are emitted to the atmosphere. In this context, it is an urgent problem to estimate the role of forest fires as a factor of regional atmospheric pollution.

De Bell et al.[6] have considered the cases of forest fires, which appeared in Quebec province, Canada, in early July 2002 on the area about 10^6 ha. The resultant smoke plume was seen in satellite images of the region of U.S. East Coast. Simultaneously, extremely high CO mixing ratios were observed at the Atmospheric Investigation, Regional Modeling, Analysis and Prediction (AIRMAP) network sites in New Hampshire and at the Harvard Forest Environmental Measurement Site (HFEMS) in Massachusetts. The observed level of CO mixing ratio was 525 to 1025 ppbv higher than that in the preceding and subsequent periods.

The presence of biomass burning source was also confirmed by observation data exhibiting increase of such components in aerosol composition as K^+, NH_4^+, NO_3^-, and $C_2O_4^{2-}$. Additional information on the abundances of K, organic carbon, and elemental carbon in aerosol composition indicated that the smoke plume affected much of the U.S. East Coast from Maine to Virginia. Comparison with observations of CO mixing ratio at the stations with 10-year or longer measurement series has made it possible to consider this event as the largest biomass burning episode observed on the U.S. East Coast over the past 10 years. From observations of CO mixing ratio, as well as from scattering coefficients calculated from data on number concentration of aerosol particles with diameters less than 2.5 μm (PM-2.5), it follows that this event is comparable in scale to large events of anthropogenic pollution and haze formation, which affect, from time to time, the rural regions of New England. The enhancement of contents of O_3, NO_y, NO_3^-, NH_4^+, and SO_4^{2-}, in the plume from forest fires, significantly depended on the elevation and latitude, probably because of the variability of long-range transport and processes of the surface deposition.

Eldering et al.[8] derived vertical profiles of volume concentration of sulfate aerosol particles and extinction coefficient from data collected in 1992, 1993, and 1994 with high-resolution infrared spectrometer under the Atmospheric Trace Molecule Spectroscopy (ATMOS) experiment conducted from onboard Space Shuttle missions. Since the observations were conducted in the period following powerful eruption of Mt. Pinatubo in June 1991, the analysis of obtained results has made it possible to trace the evolution of volcanic sulfate aerosol during three post-eruption years. Eldering et al.[8] present the results on vertical profiles of volume aerosol concentration derived for three regions of the globe in the altitude range from 10 km (polar latitudes) to 20 km (subtropical latitudes) and above. The aerosol volume concentration reached 2–3.5 $\mu m^3/cm^3 \pm 10\%$ in 1992, decreasing to 0.2–0.5 $\mu m^3/cm^3 \pm 20\%$ in 1994, in agreement with other analogous observations. Also, a good agreement of these reconstructed results is obtained in the case of vertical profiles of extinction coefficient.

Ramachandran[30] discussed the results of shipboard observations of variations and trends of aerosol optical depth (AOD) in the wavelength range 0.4–0.85 μm, performed during 1996–2000 missions in periods of north-eastern winter monsoon in the regions of Arabian Sea (AS) and tropical Indian

Ocean (TIO). Very strong latitudinal gradient of AOD was revealed while the ship cruised off the Indian coast (IC). Increasing AOD trends are observed in the vicinity of IC and AS; however, in the TIO region, the AOD systematically decreases throughout the 5-year observation period. Monthly mean AOD values for December–April increase in IC and AS regions, but decrease in the TIO zone. The AOD trends are analyzed and compared for two inland observation sites [Trivandrum (TVM) and Vishapakatnam (VSK)] on Indian peninsula and at two points in oceanic region, located in direction of predominating winds blowing from Indian subcontinent. For 5-year average AOD the latitudinal gradient is less pronounced in all cases, and the AOD has close values. For four-year period in TVM, the increasing AOD trend is recorded; however, in VSK no marked trend has been detected. At the two above-mentioned points of the region of Indian Ocean, AOD trend is decreasing and analogous to what is observed for the entire ocean. In December–April, increase of monthly mean AOD values is observed, both in the inland regions and over the ocean.

Using the data of shipboard observations of spectral aerosol optical depth over Arabian sea and tropical Indian Ocean in the periods of north-eastern winter monsoon in 1996–2000, Ramachandran[31] calculated Angström parameters α and β to analyze their spatiotemporal variations (optical depth $\tau = \beta\lambda^{-\alpha}$, where λ is the wavelength). For 5-year average value of the Angström exponent, α, the calculations provide approximately 1.5 for regions of coastal India, AS, and TIO, while 5-year average of turbidity parameter β is approximately 0.14 over IC, 0.10 over AS, and 0.05 over TIO. Behavior of α and β is characterized by the presence of interannual variations and changes of these parameters from month to month. The 5-year average values of α and β at two inland sites [Minicoy (MCY) и Kaashidoo (KCO)] are approximately 1 and 0.2, respectively. For 1996–2000, at the wavelength of 0.5 μm the dependence of AOD on diurnally mean wind velocity is distinctly seen. The estimated distance, at which the continental aerosol significantly influences the atmospheric optical properties over Indian Ocean, is 1500 km. Also, 5-year average anthropogenic contribution to AOD variations at 0.5 μm wavelength was estimated to be more than 90% over IC and AS, and somewhat smaller (74%) over TIO. For the MCY and KCO, this contribution is less than 70%.

Since the dust aerosol, formed primarily due to the influence of dust storms in deserts, is an important component of global aerosol, Martonchik et al.[21a] undertook comparisons of Multi-angle Imaging Spectro Radiometer (MISR) retrievals of aerosol optical depth (AOD) with the data of four AErosol RObotic NETwork (AERONET) stations, obtained using sun photometers located in deserts, for the period from December 2000 to November 2002. The comparison of the two data series showed good agreement (with no systematic biases), indicating the adequacy of MISR AOD data in the visible and near IR spectral regions over surfaces with high albedo. The random retrieval errors of MISR AOD are 0.08 at 17.6 km spatial resolution. If the spatial resolution is degraded to 52.8 km, the error decreases to 0.05. Thus, the MISR AOD data can safely be used for reliable quantitative estimates of variations in atmospheric dust content in desert regions.

Schaap et al.[36] proposed a model of fine atmospheric aerosol for West Europe applied to 1995 conditions taking into account CB; it is based on recent inventory of anthropogenic primary aerosol particulate matter. Annual CB emissions over West Europe and former Soviet Union for 1995 were estimated to be, respectively, about 0.47 and 0.26 Tg, with maximum contributions to aerosol abundance coming from transport and households. Based on the proposed model, the CB concentration varies in the range from ≤ 0.05 μg/m^3 in remote background regions to >1 μg/m^3 in densely populated regions. The model values of concentration are about 25% of the total primary aerosol concentration.

To reproduce the concentration field of total anthropogenic fine aerosol, Schaap et al.[36] combined the primary aerosol fields with the previously calculated distribution of secondary aerosol concentration. Consideration of these data has led to conclusion that the modeled CB contributes only 4–10% to the total fine aerosol mass; whereas the contributions of nitrate and sulfate are 25–50% and 5–35%, respectively. Comparison with observation data has demonstrated that the model underestimates PM-2.5 aerosol concentration, primarily due to underestimation of the total organic substance (both CB and OC) by approximately two times. This underestimation can be partly due to incomplete account of local emissions, observation uncertainties, inadequate representation of natural sources, and wet deposition. However, the main source of discrepancies may be insufficient reliability of the CB (and total PM) emission inventory. In comparison with previous data, the new estimates of CB emissions turned out to be about two times lower, primarily because of more reliable determination of different factors of emissions, though the corresponding estimates still need further improvement.

Recent observations have led to conclusion that, for the period from 1979 to 1998, in the New York State, the atmospheric concentrations of SO_4 and total sulfur ($SO_2 + SO_4$) have decreased, with linear relationship observed between SO_4 concentration and SO_2 emissions in eight states upwind of and contiguous with New York State. Primary SO_2 source in these eight states was coal combustion, which also entailed ash emissions to the atmosphere, containing different trace and volatile compounds. The condensation of the latter compounds has led to formation of aerosol particles. The natural aerosol

components included soil particles, sea-salt particles, and forest fire products.

Husain et al.[12] have analyzed daily aerosol samples (for about 20-year period), collected at Mayville and 350 km downwind at Whiteface Mountain (altitude of 1.5 km above sea level), to determine K, Sc, Mn, Fe, Zn, As, Se, Sb, Hg, and Pb concentrations. For the entire observation period, there were data on [SO_2] and [SO_4] concentrations, whose analysis has shown that total sulfur concentration at Mayville was approximately four times higher than at Whiteface. From 1979 through 2002, the concentration of [SO_4] decreased by 59% at Whiteface, while at Mayville it decreased by 30% from 1984 to 2002. The decrease of SO_2 concentration for the period from 1979 until 2002 in the above-mentioned eight states was found to be 49%. The observations at the two sites indicated that a linear relationship existed between [SO_4] concentration and the total sulfur concentration. These data suggest that a further reduction in SO_2 emission will lead to a proportional decrease of [SO_4] and total sulfur concentration in New York State and possibly throughout the northeast of the United States. From data for Whiteface it follows that, beginning in 1997, the decrease of [SO_4] and total sulfur, relative to SO_2 emissions, could be faster than before.

Like [SO_4] and total sulfur concentration, the abundance of trace elements was found to be about 2 to 5 times higher at Mayville than at Whiteface, with decreasing trends of concentration observed at both sites, most strong for such elements as Hg (16 and 10%/yr at Mayville and Whiteface, respectively) and Pb (14 and 10%). In other cases (K, Mn, Sc, and Fe), with the exception of Sb, the decrease was in the range of 3 to 5%/yr. Trends of Sb at Whiteface and Mn at Mayville could not be determined (seemingly, due to the presence of emission sources nearby). Undoubtedly, the consequence of reduction of SO_2 and aerosol emission levels has been the decrease of trace elements in the atmosphere and improvement in air quality.

A characteristic phenomenon in Arctic, appearing in winter and spring, is the formation of "Arctic Haze" whose important constituent is carbon black (CB), forming predominately under the impact of anthropogenic pollution sources, located in Europe, former Soviet Union, and North America, and then undergoing long-range transport to the Arctic. The carbon black, suspended in the atmosphere (in aerosol composition) or residing in the snow cover, has a significant effect on radiative forcing, caused by the Arctic atmosphere. Sharma et al.[37] discussed results of regular observations of CB, contained in atmospheric aerosol, at the station Alert (82.5°N; 62.5°W) since 1989.

Intensive observations have been carried out during three years in order to compare the results obtained through thermal analysis and by the method of optical absorption, which afforded, in particular, the possibility of studying annual behavior of the "operational" value of the aerosol absorption cross section. Analysis of the obtained time series has shown that the variations of carbon black concentration are characterized by the presence of strong annual variations superimposed upon a long-term trend. The latter amounted to a decrease of CB concentration in 1989–2002 at Alert by approximately 55%. Estimates of these factors, responsible for the aforesaid variations, such as emissions to the atmosphere and long-range transport, suggest that the decreasing trend was largely due to decrease of CB emissions in the former Soviet Union. Contributions of long-range transport from regions of North America and West Europe were less significant.

Results of satellite remote sensing presently become increasingly more important source of information on the global aerosol properties. At the same time, there appears a problem of their verification. Solution of this problem, based on comparison of ground-based, aircraft, and satellite observations, was discussed recently in a number of publications such as Refs. 1, 2a, 12a, 19a, 20a, 31a, and 41a; their main conclusion is recognition that serious discrepancies exist, and that their removal requires further development.

Some results of recent studies, mentioned above, illustrate fragmentary character of many developments whereas solution of the problem of climate change requires complex approach to atmospheric aerosol study. Let us now turn to achievements of ACE-Asia program as one of the most successful complex field campaigns.

1. Field observational experiment ACE-Asia

1.1. Meteorological conditions

Merrill and Kim[24] discussed main features of the meteorological situation in period March 30 to May 4, 2001 observed during the field aerosol observation experiment ACE-Asia intensive observation period (IOP). The observed anomalies of the structure of atmospheric circulation were found to be in the middle of the interannual variation range, with most anomalies spatially confined and small in magnitude. The occurrence and spatial distribution of midlatitude cyclones, affecting the atmospheric circulation and spatial structure of long-range aerosol transport, are close to the climatological mean. Analysis of observation data on aerosol revealed the presence of strong coarse-mode aerosol scattering at low altitudes in post cold-frontal circulations during intense dust outbreaks to the atmosphere. Also, a very strong scattering is associated with mixed-mode aerosol microstructure in zones of dust plumes from polluted areas.

To study the relation between composition of atmospheric aerosol and meteorological conditions, Park et al.[28] analyzed data of ground-based observations for 10-year period (from March 1992 to

February 2002), obtained at Gosan, Korea. Gosan is situated in the western part of Jeju island, about 100 km away from the southern part of Korean Peninsula (approximately at 500-km distance from West China and 200 km east of Japanese islands). Jeju island has very clean atmosphere, so the observations on it are highly representative in analysis of impact of the air masses invading from continent on the atmospheric properties. The observations show that the average concentration of non-sea-salt sulfate is 6.74 μg/m^3, higher than in other background areas in the world. It was demonstrated that the observed high level of sulfate is due to the air transport from outside the site. Close relationship between concentration ratios non-sea-salt (nss) sulfate/ammonium/nss-potassium, sodium/chloride/magnesium, and nss-calcium/nitrate has been revealed, suggesting that they have common emission sources and/or similar long-range transport pathways. Quite probably, there is a decrease in the nss-sulfate/nitrate concentration ratio, associated with the increase of nitrate concentration. It is assumed, that this trend is caused by the dynamics of emissions in China. Backward trajectory calculations have led to conclusion that about half of the air mass trajectories originated from northern China, and a quarter from southern China. Based on the cluster analysis, it is shown that air masses coming from China have the highest concentration of nss-sulfate, ammonium, and nitrate.

1.2. Microstructure, chemical composition, and optical characteristics of aerosol

During the field observational experiment ACE-Asia, aimed at studying atmospheric aerosol properties in the region covering territories and water basins of China, Korea, and Japan, five sessions of synchronized observations from Terra satellite instruments (Multiangle Imaging Spectroradiometer, MISR), two research aircraft, and ground-based AERONET network stations have been performed. The observation conditions included cases of clean, polluted, and dusty atmosphere. Kahn et al.[13] performed detailed comparison of the results obtained and analyzed the discrepancies occurred in the data. As expected, within a few lower kilometers of the atmosphere above the surface, a mixture of dust and anthropogenic (largely industrial) aerosol dominated. In the five cases studied, microstructure of medium- and coarse-mode particles varied little; however, aerosol and optical thickness of atmospheric column varied by a factor of 4, while variations of the dust proportion in the near-surface layer were in the range from 25 to 50%. The amount of absorbing component in the submicron aerosol (single scattering albedo of 0.8) was found to be maximum when wind crossed the Beijing region and Korean Peninsula.

Rapid industrial development of China stimulates the research and development aimed at studying how emissions of different trace gases and aerosol into the atmosphere influence the environment and ecosystems, with the primary focus on such consequences of these emissions as formation of acid deposits, "photochemical" near-surface ozone, and regional haze. The estimates available point that in the period from early 1980s to mid-1990s, there was a growth of anthropogenic emissions of such trace gases as NO_x and SO_2. However, measures to improve technologies, undertaken in China, ensured reduction of emissions: observations revealed decreasing trends of NO_x and SO_2 concentrations in 1995–2000. In this regard, Wang et al.[47] discussed the observations of concentrations of different trace gases and fine aerosol in Lin'an, located in rural region of eastern China, in the period from February 18 to April 30, 2001, coinciding with the timeframe of the ACE-Asia field experiment.

The observation program included measurements of concentrations of O_3, CO, NO, NO_y^*, SO_2, CH_4, C_2–C_8 non-methane hydrocarbons (NMHCs), C_1–C_2 halocarbons, and the chemical composition of aerosol PM-2.5. The average hourly mixing ratios (± standard deviation) of CO, SO_2, and NO_y^* were, respectively (677 ± 315), (15.9 ± 14.6), and (13.8 ± 7.2) ppbv. The mean daytime mixing ratio of ozone was (41 ± 19) ppbv. The most abundant NMHC was ethane (3189 ± 717) ppbv), followed by ethyne (2475 ± 1395), ethene (1679 ± 1455), and toluene (1529 ± 1608) ppbv. The average concentrations of particulate organic matter (POM, as organic carbon, times 1.4) and elemental carbon in PM-2.5 were, respectively, (21.5 ± 7) and (2.5 ± 7) μg/m^3, and sulfate and nitrate levels were, respectively, (17.3 ± 6.6) and (6.5 ± 4) μg/m^3.

Relatively high correlation was revealed between the mixing ratios of CO and NO_y^* ($r^2 = 0.59$), OC ($r^2 = 0.65$), CH_3Cl ($r^2 = 0.59$), soluble potassium ($r^2 = 0.53$), and many NMHCs, thus pointing to the presence of such a source as biofuel/biomass burning. There is also correlation between concentrations of CO and such an industrial tracer as C_2Cl_4, signifying the influence of industrial emissions. On the other hand, SO_2 concentration correlated well with EC ($r^2 = 0.56$), reflecting the influence of coal combustion. Ammonium was sufficiently abundant (seemingly, due to agricultural activity) to neutralize completely the sulfate and nitrate. Silicon and calcium typically have poor correlation with iron and aluminum, implying the presence of Si and Ca sources other than soil. Analysis of the concentration ratios C_2H_2/CO, C_3H_8/C_2H_6, nitrate/(nitrate + NO_y^*), and sulfate/(SO_2 + sulfate) has led to conclusion that the observation site was located in a relatively fresh air mass. Comparison of the observed concentration ratios with the corresponding data of inventories showed that, while the ratio SO_2/NO_y^*, observed in March (1.29 ppbv/ppbv), differed from inventory-derived data by 20%, for CO/NO_y^* the observed average value (37 ppbv/ppbv) was about

200% larger. The CO/NMHC ratio (taken with inclusion of ethane, propane, butane, ethene, and ethyne) also signified the presence of the CO excess, suggesting that CO concentrations obtained earlier for the eastern China were underestimated.

Arimoto et al.[1a] discussed the results of analysis of ion and elemental composition of atmospheric aerosol, based on regular observations at Zhenbeitai (ZBT) (39.3°N; 109°E), China, in the period April 8–30, 2001, and at (GOS) (34.28°N; 126.17°E) during two time periods, March 25 – May 1, 2001, and May 1 – September 30, 2001 (only on seven chosen days), as part of ACE-Asia program. From analysis of samples of fine aerosol PM-2.5 at ZBT it follows that the concentration of 24 studied elements correlates with Al (an indicator of dust aerosol), and that ratios of concentration of all studied elements to Al concentration are close to those in a loess certified reference material. However, the second group of studied elements was enriched over the first group, probably because of anthropogenic emissions to the atmosphere. The concentrations of water-soluble (WS) cations (Na^+, K^+, Ca^{2+}, Mg^{2+}) generally correlated with Al concentration at both observation sites except for WS K^+ at ZBT, where biomass burning may have had an effect.

The observations showed that the percentage of soluble calcium approached 100% at ZBT versus ~60% at GOS. The ratio WS Ca^{2+}/Al was also higher at ZBT. Molar ratio of sulfate concentration to water-soluble Ca^{2+} was ~0.1 at ZBT, but increased to almost unity at GOS, where the ratio nitrate/soluble Ca^{2+} was found to be tenfold and even hundredfold higher than that at ZBT (presumably due to anthropogenic influence). The observed differences of aerosol characteristics at the two considered sites can be interpreted as the final result of contributions of different components combined with complex process of gas-particle conversion, size-dependent fractionation, and aerosol mixing.

One of the regions of northwest China, which is an important source of dust (mineral) aerosol (DA), is Gobi desert. In the framework of the ACE-Asia field observational experiment in the period from March 30 to May 1, 2001, at the monitoring site devoid of local pollution sources and located 5 km north of Yulin (38°20′N; 109°43′E, 118 m above sea level), owned by Institute of Earth Environment of Chinese Academy of Sciences, complex observations were performed to study chemical, physical, and radiative characteristics of aerosol delivered to the atmosphere under desert conditions.[48]

The mean values and standard deviations (in parentheses) of the observed absorption coefficient σ_{ap}, scattering coefficient σ_{sp}, and single scattering albedo were 6 Mm^{-1} (11 Mm^{-1}), 158 Mm^{-1} (193 Mm^{-1}), and 0.95 (0.05). Values of σ_{ap} and σ_{sp} were characterized by the distinct diurnal behavior, caused by diurnal behavior of turbulent mixing in the atmospheric boundary layer, as well as by contributions of pollution sources from fossil fuel combustion in morning hours and intensification of dust emissions in the afternoon. Xu et al.[48] revealed the existence of two distinct populations of aerosol mass scattering efficiency Escat_2.5, one for aerosol dominated by DA (~1.0 m^2/g), and the other for aerosol from local pollution sources (~30 m^2/g). During the observations, three episodes of dust storm occurred lasting, on average, a few days.

A consequence of most powerful dust storm was increase of 24-hour-average particle mass concentration PM-2.5 of fine aerosol fraction to 453 $\mu g/m^3$, with peak σ_{sp}, reaching 2510 Mm^{-1} on April 8. The mean particle concentration PM-2.5 for period of all the three dust storms was about 169 $\mu g/m^3$, about 4 times greater than the mean value (44 $\mu g/m^3$), observed in the periods of influence of local pollutions. Under these conditions, when the local sources become a dominant contributor to fine particulate mass, the major component of aerosol composition had been organic matter (OM), contributing 41% to the PM-2.5 particulate mass, followed by crustal material (29%), sulfate (17%), and elemental carbon (13%). During sandstorms, about 51% of the mass of fine aerosol was accounted for by crustal component, followed by CO_3^{2-} (11%) and OM (9.5%). Determination of element enrichment factors has led to conclusion that significant local pollution sources are coal combustion, biomass burning, as well as different mobile sources.

Bates et al.[2] discussed observations of aerosol chemical composition and microstructure, performed aboard the Research Vessel (R/V) *Ronald H. Brown* from March 14 to April 20, 2001, as part of the Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia). They analyzed primarily the samples, collected in the Sea of Japan in conditions of prefrontal and postfrontal air masses between April 6 and 15, 2001, with focus on determination of contrasts in aerosol properties for different meteorological conditions in atmospheric marine planetary boundary layer off the East coast of Asian continent. The prefrontal air mass had a dominant accumulation aerosol mode composed mainly of pollution products and volcanic aerosol. Predominating constituents in aerosol composition were ammonium sulfate and organic carbon, and in minor amount, there was dust aerosol as a result of precipitation of aerosol brought by long-range transport after being injected to the atmosphere in Takla Makan desert. Also, the detected sea-salt submicron and larger aerosol particles were highly depleted in chloride due to reactions with sulfuric and nitric acid vapor. The passage of a powerful low-pressure center, surrounded by airborne atmospheric dust, on April 10 led to an increase in the concentration of submicron and larger dust aerosol particles from Gobi desert. The supermicron dust particles were characterized by high concentration of sulfate, nitrate, organic, and elemental carbon.

Measurements conducted during ACE-Asia intensive observation period (IOP) in spring 2001 included complex observations of physicochemical characteristics and radiative properties of dust aerosol (DA), formed after three dust storms in China. The observations were performed in Kwangju (South Korea) on March 22, April 11–13, and April 25–26, 2001, and included continuous measurements of DA extinction, scattering, and absorption coefficients with the use of transmissometer, integrating nephelometer, and aethalometer. The results of these observations were compared with data of analogous measurements under conditions of the clean marine and haze-contaminated urban atmosphere.

DA characteristics in the first dust event were quite typical for dust storms observed in the desert region of the northwestern China. In the case of the second dust storm, the DA properties significantly changed in the process of long-range transport under the influence of mixing with polluting components in the atmosphere, which led to an increase of sulfate and organic carbon concentrations in the aerosol composition. Kim et al.[15a] analyzed the specific features of aerosol chemical composition and optical properties under conditions of clean continental, southeastern marine, and stagnant polluted atmosphere. For the three dust storm events, they determined mass scattering coefficient and single scattering albedo for fine and coarse aerosols. Aethalometer is used to measure the CB concentration in the composition of fine and coarse aerosol. It is found that CB concentration in the coarse mode increased due to agglomeration of CB particles and high concentration of DA. Under conditions of the three dust storms considered, the single scattering albedo increased to 0.93, 0.90, and 0.84, respectively, while in other times the average value was 0.85.

Results of quasi-Lagrangian airborne measurements, considered by McNaughton et al.,[23] provided information on characteristics of the processes of formation of the secondary aerosol and its condensational growth in the atmosphere over the Yellow Sea, East China Sea, and Sea of Japan during Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia) and Transport and Chemical Evolution over the Pacific (TRACE-P) experiment. Under conditions of the discussed airborne observations, the specific surface area of mixed-composition aerosol particles, including pollution products and mineral dust, reached 1200 $\mu m^2/cm^3$. Concentration of sulfuric acid vapor generally was insufficient for binary nucleation. However, data of observations and numerical simulations show that ternary nucleation, H_2SO_4–H_2O–NH_3, mechanism works. Growth rate of ~2 nm/h can be explained by the influence of condensation of sulfuric acid at a rate of $(2 \pm 1) \cdot 10^6$ molecules/($cm^3 \cdot s$). Aerosol volatility suggested intensification of neutralization of the aerosol during particle growth.

From measurements of aerosol microstructure it follows that a characteristic feature of the atmosphere of the considered region was weak ability to produce new particles (mean number concentration of the condensation nuclei (CN) of 3 to 13 nm size was about 500 cm^{-3}). However, in postfrontal air masses, associated with the offshore flows under clear-sky conditions, new production of particles was enhanced by as much as an order of magnitude (mean concentration of CN of 3 to 13 nm size reached 5000 cm^{-3}). Fogs and clouds played an important role in the regional-scale processes of nucleation by washing out secondary aerosol, as well as causing depletion of gas-phase precursors through heterogeneous chemical reactions. Results obtained by McNaughton et al. demonstrate that only 10–30% of the total aerosol population consists of aged secondary aerosols about 2 days after onset of long-range transport of aerosol from its source. Despite its high productivity during nucleation events, the secondary aerosol advected to the region of Pacific Ocean, has only small influence on indirect radiative forcing (RF), and produces almost no effect on the direct RF compared with the primary aerosol emissions and trace constituents that condense on these aerosol particles.

The results of McNaughton et al.[23] are in contrast with the data on the formation of the secondary aerosol in the remote (background) free troposphere. Under these conditions, the airborne nucleation, in regions where cloud-induced aerosol scavenging took place, is followed by a slow particle growth accompanied by slow coagulation, which determines the important role of thus formed particles as a supplement to the existing CN, and also (sometimes under conditions of clean atmosphere) as a factor of light extinction due to scattering. This dictates the necessity to take into consideration secondary nucleation, in numerical modeling of the aerosol effect of radiative forcing, which turns out insignificant under conditions of polluted atmosphere.

So far the information on the properties of atmospheric aerosol remains fragmentary and, particularly, undoubtedly insufficient for verification of results of numerical simulation of spatiotemporal variations of aerosol properties (microstructure and chemical composition), especially organic components of aerosol and elemental carbon. It is for this reason that in the ACE-Asia field observational experiment, performed in spring 2001 in Asian Pacific region, and primarily focused on the study of aerosol outbreaks from Asian continent to Pacific water basin, special emphasis was put on airborne observations (C-130 research aircraft) of the abundance of elemental carbon (EC) and OC in aerosol composition. Aerosol samples were collected on quartz filters whose use turned out quite reliable, despite serious evaporation of samples detected on about one third of the filters.

Results of sample analysis, obtained by Huebert et al.,[11] have led to a conclusion that the concentration of total carbon (TC) and OC was in general higher in the boundary layer (medians of 7.6 $\mu gTC/m^3$ and 5.8 $\mu gOC/m^3$) than in the free

troposphere (3.1 μgTC/m^3 and 3.9 μgOC/m^3). The same appears to be true for EC, though the bulk of these data was insufficient to obtain reliable quantitative estimates. Data of observations also show that OC concentration turned out to be much more variable than that of the EC. Under conditions of free troposphere the concentration ratio of TC to non-sea-salt (nss) sulfate aerosol varied in the range from 3.2 to 6.0, that is indicative of that the carbonaceous aerosol was primarily concentrated in the upper troposphere in contrast to the nss sulfate aerosol.

When mass of dust (mineral) aerosol was included, relative abundance of organic matter in fine aerosol was in the range from 0.1 to 0.7. The TC/EC ratios ranged from 2 to 15, with medians being ~ 4–5. The atmosphere in the regions of Yellow Sea, the Korea Strait, and the Sea of Japan was generally characterized by higher OC concentration than East China Sea, or the Pacific south and east of Japan. The data of observations demonstrate very strong variations of concentration of different aerosol components at all altitudes. Variation of the ratio of maximum to minimum concentration reached several orders of magnitude.

Maxwell-Meier et al.[22] collected aerosol samples in the free troposphere from onboard a C-130 research aircraft and then analyzed the samples in the laboratory for the presence of inorganic components. Maxwell-Meier et al.[22] discussed the results obtained during three flights as part of ACE-Asia field experiment in planetary boundary layer (PBL) over Yellow Sea. The purpose of processing the observation data was to study the processes of DA plumes formation, observed in spring 2001 under conditions when these processes were governed by combination of intense frontal activity in such a vast arid region as Gobi desert, as well as dry agricultural fields prepared for sowing. Dust storms, occurring under such conditions, led to formation of large amount of DA, and its long-range transport determined the significant influence of aerosol on radiation regime and climate.

Analysis of aerosol samples has shown that the main components of water-soluble mineral dust were Mg^{2+} and Ca^{2+}. Study of ion balances of fine and total aerosol has demonstrated that a significant fraction of Mg^{2+} and Ca^{2+} ions existed in the form of carbonates. In polluted air mixed with dust that advected from large cities in roughly 0.5 to 1 day, much of the fine aerosol, containing nitrate and sulfate (approximately 80%), was associated with ammonium or calcium, with the latter being likely associated with mineral dust. Only air masses that spent 2 to 5 days over Yellow Sea had clear evidence of Cl^- depletion. The discussed observation data can be explained by assuming initial mass accommodation coefficients less than 0.1 for uptake of SO_2 or HNO_3 by DA particles in urban aerosol plumes containing components of emissions due to fossil fuel combustion and biomass burning. The observation data suggest that the accommodation coefficient depends on the relative air humidity.

Kline et al.[17] discussed the results of ion chromatographic analysis of filter and impactor samples of aerosol, collected onboard a C-130 research aircraft during ACE-Asia field observational experiment in spring of 2001 in the region of Southeastern Asia. The results considered bear information on composition (analysis for the presence of sulfate, ammonium, nitrate, calcium, potassium, magnesium, and oxalate) and concentration of coarse and fine aerosol fractions. These results exhibit the presence of significant differences between aerosol composition in the free troposphere (FT) and the boundary layer (BL) of the atmosphere. For instance, the averaged molar ratio of nitrate to soluble calcium was 1.8 in BL, but only 0.2 in FT. Nitrate and calcium were frequently characterized by size distribution identical to that of coarse aerosol, while sulfate and ammonium typically had distributions identical to that of fine aerosol.

Dust aerosol typically experiences conversion of NO_y to coarse-mode nitrate aerosol. Whereas sulfate abundance in FT aerosol is close to concentration of ammonium bisulfate (half neutralized), the non-sea-salt (nss) sulfate in BL usually suffers almost complete neutralization to ammonium sulfate. In the presence of DA, up to 50% of nss sulfate is in the coarse-mode fraction (probably, due to SO_2 uptake by $CaCO_3$ contained in DA). Averaged content of soluble calcium in the coarse-mode DA fraction is 5–8%. Generally, the specific features of chemical composition of aerosol in BL are not representative indicators of ion composition of aerosol. An important feature of aerosol in Asian Pacific region is very strong spatiotemporal variation of its properties, determined by the presence of distinct aerosol layers alternating with the clean atmosphere. Under these conditions, the averaged aerosol characteristics cannot be considered as representative information. Similarly, the results of ground-based observations cannot quantify aerosol properties in the boundary layer of the atmosphere and in the free troposphere.

To estimate the adequacy of different methods of measurement of inorganic aerosol chemical composition in the troposphere, the TRACE-P and ACE-Asia field observational experiments in spring 2001 deployed three research aircraft, equipped with instruments of five types: filters, five-stage micro-orifice impactor (MOI), mist chamber (MS), and particle into liquid sampler PILS. Ma et al.[20] compared the data of identical PILS, installed onboard two aircraft, and revealed high correlation between them (e.g., $r^2 = 0.95$ for sulfate aerosol), but, on the other hand, systematic discrepancies (10 ± 5)% have also been revealed. Comparison of PILS and MS measurement data on fine sulfate aerosol has led to $r^2 = 0.78$ at the systematic discrepancy of (39 ± 5)%. The MOI and PILS measurements of ion composition of aerosol agree within (14 ± 6)% at $r^2 = 0.87$. For most ion types,

the discrepancies were within ±30%. Overall, it can be concluded that the data of aircraft observations of fine aerosol composition agree within 30–40% accuracy.

Zhang and Iwasaka[50] compared sizes and chemical composition of separate particles of atmospheric aerosol before and after their water dialysis, made to remove water-soluble components in sample of aerosol having arrived from south-western Japan and Asian continent. The comparison has shown that, after dialysis, the aerosol microstructure, representing a mixture of mineral (dust) and sea-salt aerosol, underwent shift toward more abundant fine particles in comparison with aerosol studied before making dialysis. This shift increased with growth of sea-salt content in the particles; whereas the process of dialysis did not result in marked changes of sizes or morphology of particles provided that they contained no sea salt. Studies of all dust aerosol have led to conclusion that its mixing with sea salt entailed shift of microstructure toward larger particle sizes, reaching 0.4–0.8 μm. Thus, the interaction of dust particles with sea salt seems to be an important factor of change of their size and composition during long-range transport, which, in its turn, influence the radiative transfer in the atmosphere and the process of mineral dust deposition onto the underlying surface.

In spring 2001, in the region of Asian Pacific coast, two large-scale field observation programs were performed: TRACE-P field experiment, aimed to study long-range transport and chemical transformation of air masses over Pacific Ocean (sponsored by NASA), and ACE-Asia field experiment, targeting studies of aerosol characteristics (sponsored by National Science Foundation, USA). Both were aimed at studying the consequences of pollution of the atmosphere over Pacific water basin by emissions of trace gases and aerosol, caused by such factors as biomass burning, urban and industrial pollutants, dust aerosol formed under the impact of dust storms in China deserts.

TRACE-P concentrated on airborne measurements of trace gases and aerosol in March–April using instrumentation installed onboard two research aircraft, DC-8 and P3-B, based in Hong Kong and Yokota Air Force Base, Japan. ACE-Asia field experiment focused on airborne observations of aerosol characteristics and radiative fluxes in April–May and involved NSF C-130 aircraft, based in Iwakuni Marine Corps Air Station, Japan.

Moore et al.[25] compared results of the above-mentioned airborne measurements of aerosol characteristics: number concentration, microstructure, chemical composition, and optical properties. Best agreement (generally within the RMS instrumental uncertainties) took place in comparisons of microphysical characteristics of submicron aerosol, including condensation nuclei number concentration, scattering coefficient, as well as size distribution of the accumulation mode measured with a differential mobility analyzer and an optical particle counter (OPC). Large discrepancies (exceeding the instrumental RMS uncertainties) were frequently observed in the case of microstructure parameters of the coarse aerosol. This concerns the data on scattering and absorption coefficients, as well as on the concentration of soluble components of chemical composition, usually contained in large aerosol particles (e.g., Na^+, Cl^-, Ca^{2+}, and Mg^{2+}). Some of the observed discrepancies are associated with specific character of aerosol sampling and specific features of instrument configuration and design. For horizontal legs of different lengths, means and variances of simultaneously measured parameters were calculated, with subsequent analysis of their correlations, which was important, in particular, for assessment of instrument performance.

Starting from January 1, 2001, mass concentration of chemical composition of fine (PM-2.5) and coarse (PM-10) atmospheric aerosols was measured every Wednesday and Sunday during 18 months at three key ACE-Asia sites (Hong Kong, Cheju Island in South Korea, and Sado Island in Japan). Data obtained by Cohen et al.[4] show that diurnally mean (for the entire observation period at the three sites) concentrations were, respectively, 29, 16, and 9.1 μg/m^3 (PM-2.5) and 33, 14, and 11 μg/m^3 (PM-10). The corresponding maximum loadings are 109, 81, 78 μg/m^3 (PM-2.5) and 101, 162, and 253 μg/m^3 (PM-10). Chemical analysis of aerosol samples was also made to quantify the content of the following elements: F, Na, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Br, and Pb, with detection limits lower than 1 ng/m^3. The average PM-2.5 percentage composition by weight for all the three sites was estimated to be: (8.4 ± 4)% carbon black, (7.7 ± 7)% soil components, (43 ± 14)% ammonium sulfate, (11 ± 16)% organic matter, (10 ± 12)% sea-salt aerosol, and (0.6 ± 0.3)% trace elements.

Cohen et al.[4] identified soil fingerprints of aerosol composition in East Asian region based on measurement data on the content of oxides of Al, S, K, Ca, Ti, Mn, and Fe. The results obtained indicate that the coarse fraction was dominated by wind blown soil particles (23%) and sea salt aerosol (48%). Mass concentration of sulfur, predominately (98%) of anthropogenic origin, at the three sites was correspondingly 8.7, 4.8, and 2.9 μg/m^3 having a pronounced annual behavior (especially in Hong Kong) with the wintertime maximum. Average fraction of sulfate aerosol varied in the range 30–32% (with respect to the total mass), while the mass concentration of sulfate component PM-2.5 at the three sites was found to be 30, 19, and 12 μg/m^3. For carbon black concentration, the average values were 1.8, 1.1, and 0.77 μg/m^3 with very strong annual behavior and wintertime maximum. The fraction of carbon black varied from 8 to 9% of the total aerosol mass. Averaging of data for mass concentration of PM-2.5 yielded the following results, characterizing the weight fraction of separate components: (8.4 ± 4)% carbon black, (7.7 ± 7)% soil

component, (43 ± 14)% ammonium sulfate, (11 ± 16)% organic matter, (10 ± 12)% sea salt aerosol, and (0.6 ± 0.3)% trace elements. Average mass ratio PM-10/PM-2.5 is 2.1 ± 0.4. The coarse fraction was dominated by sea salt aerosol (59 ± 25)% and wind blown dust (17 ± 11)%.

In ACE-Asia field campaign period in spring from March to May 2001, Shimizu et al.[38] performed continuous sensing of atmospheric aerosol with polarization lidars in Beijing, Nagasaki, and Tsukuba (using second harmonic of a Nd:YAG laser radiation at 532-nm wavelength). Results of these observations were used to classify the vertical profiles of dust aerosol (more precisely, "non-spherical" aerosol) and spherical particles, as well as to determine such characteristics as frequency of occurrence of the events of elevated atmospheric dust content, aerosol concentration, and parameters of cloud cover at altitudes up to 6 km. Frequency of the dust occurrence was maximum in Beijing for the entire altitude range. In Nagasaki, there was a maximum of dust occurrence near the surface. Tsukuba was characterized by frequent occurrence of dust layers in the free troposphere. Data on the depolarization ratio were used to calculate the contributions of DA and spherical aerosol particles to the total backscattering coefficient (assuming that both of the aerosol types considered are external mixtures).

Data on the vertical profiles of DA backscattering coefficient and spherical particles exhibit the diversity of aerosol characteristics. Monthly mean backscattering coefficients of DA near surface were 0.03 $km^{-1} \cdot sr^{-1}$ (Beijing), 0.001–0.002 $km^{-1} \cdot sr^{-1}$ (Nagasaki), and 0.0006 km^{-1}/sr^{-1} (Tsukuba). Average backscattering coefficient of spherical particles was 0.002–0.004 $km^{-1} \cdot sr^{-1}$ at all observation sites. Comparison of observed backscattering coefficients with those calculated using Chemical Weather Forecasting System (CFORS) model showed that the model quite reliably reproduces the vertical structure of DA distribution and the increase in the content of spherical particles throughout the observation period. Shimizu et al.[38] described in detail the results of simultaneous sensing from five lidars in Japan on May 16–19, 2001.

Simoneit et al.[40] analyzed aerosol samples collected aboard a C-130 research aircraft under conditions of the polluted troposphere in the east Asia/Pacific region. The primary goal was to study the organic compounds of different types using capillary gas chromatography–mass spectrometry. More than 70 organic species were detected in aerosol and classified into several functional groups including *n*-alkanes, polycyclic aromatic hydrocarbons, fatty acids, dehydroabietic acid, alkanols, water-soluble sugars (including glucose, sucrose, mycose, and levoglucosan), monocarboxylic and dicarboxylic acids, urea, and phthalates. It is noteworthy that water-soluble compounds (with concentration in the range 72–133 $ng \cdot m^{-3}$) were found to make up from 16 to 50% (34% on average) of the total identified compound mass (TCM). The organic compounds were further categorized by sources.

Fossil fuel combustion was most important TCM source, contributing from 33 to 80% (with average of 50%) of TCM, followed by soil resuspensions (5–25%, 19% on average) and secondary oxidation processes (4–15%, 9% on average). In contrast, the contributions of natural sources such as plant wax and marine lipids (fatty acids and alkanols) was relatively small (correspondingly 3.4 and 9.4%, on average). In the case of Asian aerosol during spring, the biomass burning products were found to have only minor contribution (1.4%, judging from data on levoglucosan). However, it is possible that levoglucosan suffered partial hydrolysis and/or oxidation during long-range aerosol transport, so the measured values characterize its low concentration limit. The considered composition of organic compounds in the aerosol samples studied differs from the composition of aerosol particles in the atmosphere over Atlantic Ocean in terms of the abundant presence of such water soluble components as saccharides, anhydrosaccharides, and the secondary dicarboxylic acids. The obtained results convincingly demonstrate the possibility of long-range transport of organic components in aerosol composition, and help in making more reliable identification of the sources of organic aerosol components in the Asia/Pacific region.

Simoneit et al.[41] analyzed content of organic tracer components, as well as organic carbon and elemental carbon, in aerosol samples acquired during ACE-Asia in spring 2001 in Gosan, Jeju Island, Korea, in Sapporo, Japan, and in Chichi-jima Island in the western Pacific of Southern Hemisphere, as well as on the National Oceanic and Atmospheric Administration *Ronald H. Brown* R/V. Application of the method of gas chromatography–mass spectrometry has made it possible to discriminate polar and aliphatic compounds. Total aerosol and its components such as organic matter, and lipid and saccharide compounds, recorded during Asian dust episode in early April, 2001 appeared to be much higher than in other time periods.

Organic matter was apportioned among seven sources and oxidation producing secondary organic products during long-range aerosol transport. Natural background compounds, revealed at land according to observation data, represented vascular plant wax lipids released to the atmosphere by direct emission or as part of dust aerosol formed during dust storms. Also, there were no doubt about the presence of fossil fuel combustion products emitted by transport and industries. Saccharides make up the major polar (water-soluble) carbonaceous fraction originating from soil resuspension (agricultural activities). All samples in all observations contained products of biomass burning contributing up to 13% of all water-soluble components. Burning of waste was one of the sources of organic compounds in aerosol particles.

Variable levels of marine lipids were superimposed on the basic aerosol composition during long-range aerosol transport over ocean. Content of secondary oxidation products in aerosol composition increases in the processes of long-range transport. An important fact is that the aerosol, studied during ACE-Asia campaign, is not a product of only desert dust storms, but also it includes such components as soil aerosol, smoke from biomass and waste burning, and emissions from fossil fuel combustion under urban conditions.

It is well known that the aerosol radiative forcing (ARF), determining the level of aerosol effect on climate, is comparable with greenhouse radiative forcing (RF) (the globally mean ARF ranges from 0 to -2 W/m^2). However, ARF estimates are still very uncertain, largely due to strong spatiotemporal variations of aerosol concentration and properties. Since the aerosol characteristics in the atmosphere over eastern Asian region (microstructure, aerosol optical depth, Angström exponent α, and single scattering albedo) are highly specific, Kim et al.[14] analyzed their variations using ground based observations of direct solar and scattered radiation at the seven wavelengths: 315, 400, 500, 675, 870, 940, and 1020 nm, in 1998–2000 at eight stations located in China, South Korea, and Japan.

Analysis of observations has shown that the annual behavior of aerosol optical depth (AOD) typically exhibits spring maximum and fall minimum. In comparison with observation sites located in desert regions of China (Dunhuang, Mandalgovi), data obtained at Sri-Samrong and Yinchuan show more distinct annual behavior of AOD. Generally, aerosol microstructure appears to be bimodal, and is characterized by the presence of fine (with characteristic particle size of about 0.2 μm) and coarse (2–5 μm) modes. Like AOD and α, the size spectra of volume concentration ($dV(r)/d\ln r$, cm^3/cm^2) also strongly vary as functions of location of observation site and time of the year. Thus at Dunhuang the coarse aerosol fraction behaves uniformly in all seasons, whereas at Mandalgovi and Sri-Samrong strong annual behavior of the total volume of fine-mode particles was observed.

Aerosol single scattering albedo (SSA) over east Asia at wavelength 0.5 μm was close to 0.9, that well agree with data of AERONET observations, but exceeding earlier results obtained in the east Asian region (0.63–0.89). Aerosol optical properties in the regions of South Korea and Japan were found to be quite similar to those observed in dust source regions such as Dunhuang and Mandalgovi, although the AOD values were smaller (especially in periods of dust storms in deserts of China). Undoubtedly, the aerosol properties observed in South Korea and Japan were strongly influenced by long-range transport of anthropogenic aerosol from China (especially under conditions of west–east transport). Thus, the variable aerosol properties in the considered region are determined by the processes of mixing and long-range transport of aerosol of various origins (dust, urban, and biomass burning aerosol).

During ACE-Asia field experiment conducted in spring 2001, complex observations of aerosol optical properties were performed on board the *Ronald H. Brown* R/V to study the effects of atmospheric aerosol on atmospheric chemistry and climate. Measurements of aerosol properties in western Pacific in the Northern Hemisphere included determination of the aerosol microstructure and chemical composition, as well as aerosol optical parameters (scattering and absorption coefficients and hemispheric backscattering coefficient). In addition, optical depth and aerosol backscatter coefficient were measured.

Quinn et al.[29] analyzed the obtained results and concluded that the aerosol within the ACE-Asia study region is a complex mixture whose chemical composition results from many aerosol sources (marine, industrial, volcanic, and dust storms). Quinn et al.[29] reproduced and discussed in detail the observations of the following quantities: fractional mass concentration of the components dominating in the aerosol chemical composition, contribution of each component to aerosol scattering, mass scattering coefficient of individual components, aerosol scattering and absorption coefficients, single scattering albedo, Angström exponent, AOD, and vertical profile of aerosol extinction (all obtained for the relative humidity of $55 \pm 5\%$).

Overdetermination of the results obtained favored comparisons of measured and calculated aerosol characteristics, and assessment of internal consistency and error sources of the discussed data. Comparisons of measured and calculated scattering coefficients, performed by adjusting the measured microstructure to take into account the particle nonsphericity, revealed quite satisfactory agreement (within measurement errors). However, the corresponding differences in absorption coefficients have been substantial, and can be due to both inadequacy of Mie calculations, and assumption of the internally mixed homogeneous spherical aerosol particles. Values of mass scattering coefficients of non-sea-salt sulfate aerosol and sea salt aerosol, as well as submicron particles of organic matter and dust agree with ACE-1, Aerosols-99, and Indian Ocean Experiment (INDOEX) data. A unique feature of ACE-Asia data was a large fraction of dust aerosol, which determined its dominant contribution to aerosol optical properties, as well as an important role of interaction of dust and soot aerosol.

Since large uncertainty remains in the data on aerosol radiative forcing, Lee et al.[19] analyzed variations of aerosol properties using Sea-Viewing Wide Field-of-View Sensor (SeaWiFS) satellite observations, performed during the ACE-Asia intensive observation period in spring 2001. The parameters retrieved are AOD and Angström exponent. These were retrieved using Bremen aerosol retrieval (BAER, Germany) technique for regions over land and ocean with the spatial resolution of

1×1 km. SeaWiFS-derived AOD data were compared with ground-based AERONET AOD observations. The validation of satellite data, made in such a way, gave the correlation coefficient $r > 0.89$ for the period of ACE-Asia IOP. The SeaWiFS-retrieved AOD values in the wavelength interval 443–670 nm were high (about 0.8), while Angström exponent was 0.51 during an Asian dust case of 13 April, 2001, over Gosan, South Korea. After the dust episode, AOD has decreased to 0.30, while Angström exponents became 1.12.

As was indicated by Satsumabayashi et al.,[35] on July 8, 2000, Miyake volcano started to erupt; it is located in the northwest Pacific Ocean 200 km of the Tokyo metropolitan area. The maximum SO_2 emissions to the atmosphere from this volcano reached $6–10^4$ t/day, which approximately corresponded to the level of anthropogenic SO_2 emissions in northeastern Asia and was a factor of 20 larger than emissions in Japan. Two years before this event, regular (daily) observations of aerosol, precipitation, and certain gaseous pollutants (HCI and HNO_3 vapor) were initiated in this region (Happo ridge, 330 km away from the volcano). Processing of these observations revealed increase of concentration of sulfur dioxide (SO_2) by a factor of 3.8, while SO_4^{2-} concentration in aerosol and precipitation during observations has been, respectively, 1.5 and 1.7 times higher. At the same time, there was a decrease of NO_3^- and Cl^- concentrations in aerosol, caused by influence of volcanic emissions. These processes were associated with formation of SO_4^{2-} during SO_2 transport. The SO_4^{2-} produced, first of all, exhausted gaseous ammonium concentration to form $(NH_4)_2SO_4$-containing aerosol and then, $(NH_4)_2SO_4$ replaced NO_3^- and Cl^- in the aerosol composition, what sometimes manifested itself as a sulfuric acid mist formed. This process can be described as establishment of multi-component equilibrium in the gas–aerosol particles system. As to removal of HNO_3 and HCl from the atmosphere, it might occur through wet and dry deposition, favoring environmental acidification.

The Twin Otter research aircraft, operated in spring 2001 as part of ACE-Asia field observational experiment, served observation platform equipped with two types of instruments for spectral measurements of shortwave radiation at different altitudes in the troposphere. The Spectral Solar Flux Radiometer (SSFR), used for spectral measurements of upward and downward hemispheric fluxes of shortwave radiation, provided measurements in the wavelength range 300–1700 nm with the error about 1 to 3%. Ames Airborne Tracking Sunphotometer (AATS-14, 14 channels) performed measurements of direct solar radiation with the purpose of a subsequent calculation of aerosol optical depth. Bergstrom et al.[2a] discussed the observation results obtained for two cases: 1) a moderately thick aerosol layer (April 12); and 2) relatively thin aerosol layer (April 16). On both days, the Twin Otter repeatedly measured vertical profiles of radiative fluxes in the region of Korean Strait near Gosan Island. Analysis of results, obtained on 12 April, has shown that there was increase of single scattering albedo with wavelength, from 0.8 at wavelength 400 nm to 0.95 at 900 nm, then remaining essentially constant between 950 and 1700 nm. In data acquired on April 16, the aerosol absorption was very low; whereas SSA somewhat decreased with growing wavelength in the visible spectral range.

The results of observations are interpreted in the context of the presence of two absorbing aerosol components: mineral dust and carbon black. Thus on April 12, the effect of mixture of mineral dust and carbon black was observed, while on April 16, the dominating contribution to aerosol absorption seemingly was from carbon black mixed with non-absorbing aerosol of anthropogenic origin. Comparison of data on April 12 with the data of SeaWiFS satellite observations, measurement data acquired from aboard Twin Otter research aircraft, and shipborne measurements aboard *Ronald H. Brown* R/V, as well observations in Gosan, South Korea, and in Japan, have led to conclusion about quite complex nature of aerosol absorption. This is probably caused by the combined effect of the mixture of industrial aerosol (including carbon black), contained in anthropogenic pollutants of the atmosphere, and natural mineral dust. It is an important problem to identify and quantify the contributions of the two aerosol components to absorption of shortwave radiation. In this regard, it is quite important the good agreement between data on spectral downward flux of shortwave radiation in the wavelength range 300–1700 nm, obtained at 2-km altitude (above aerosol layer) on April 12, 2001, and the calculated ones.

The Transport and Chemical Evolution over the Pacific (TRACE-P) field observational experiments and ACE-Asia campaign, performed in spring 2001, involved complex observations of number concentration, microstructure, state of mixing of aerosol particles and optical properties of different aerosol types (dust, carbon black, and other aerosol components in the plumes formed under impact of urban/industrial pollutions and dust storms in China deserts). The use of optical particle counters in combination with the method of thermal heating ensured extraction of volatile components and resolution of particle sizes of dust aerosol and soot, usually dominating in absorption of shortwave radiation.

The observation data, processed by Clarke et al.,[3] showed that the CB particles were internally mixed with volatile aerosol in ~85% of accumulation-mode particles and made up about 5 to 15% of the mass of all particles. The presence of these optically active particles determined the microstructure of soot and dust aerosol, and constrained the imaginary part of complex refractive index to $k = 0.0006 \pm 0.0001$.

Correspondingly, the single scattering albedo ω at the wavelength 550 nm, for the dust aerosol varied from 0.99 (particle diameter $d_p < 1$ μm) to ~0.90 ($d_p = 10$ μm) with (size-integrated) campaign-average of 0.97 ± 0.01. Typical mass scattering coefficient for DA was approximately 0.3 m^2/g, and mass absorption efficiency MAE = 0.009 m^2/g. Less DA amount in the atmosphere south of 25° N and stronger biomass burning signatures resulted in lower values of $\omega \sim 0.82$ in the tropospheric pollution plumes.

The concentration of elemental carbon correlated with CB-caused absorption ($r^2 = 0.40$), while the volume of soot particles 0.1–0.5 μm in size was characterized by high correlation ($r^2 = 0.79$) with absorption of shortwave radiation. The value of MAE for CB reached $-7 + 2$ m^2/g and exceeded the calculated value MAE = 5 m^2/g. The observed increase of mass fraction of soot aerosol and CB in the pollution-caused aerosol in the presence of dust layers in the free troposphere was due to uptake of the volatile components onto coarse DA particles. Under these conditions, ω decreases for accumulation mode from 0.84 down to ~0.74. The calculations based on the model of long-range transport, taking into account the chemical reactions, showed good agreement with observed absorption by CB for most of the south-eastern Asia, as well as plumes from biomass burning. However, calculations underestimated, by approximately three times, the CB concentration in the exhaust of internal combustion engines in the region north of 25°N.

During the TRACE-P observational field experiment in March 2001 Mari et al.[21] performed two series of airborne observations of atmospheric pollution in two cold front episodes (flights out of Yokota, Japan). The observation data were analyzed through comparison with results of mesoscale three-dimensional numerical modeling. Special attention was paid to analysis of impact of cyclonic systems on the long-range transport of pollutants from Asia, including estimates of the relative role of convection and updrafts in the warm conveyor belts associated with the considered cyclones.

Although favorable conditions for transporting polluted air masses out from Asian continent (i.e., occurrence of warm conveyor belt, WCB, near the source region of emissions to the atmosphere) were observed during one of the flights, numerical simulation did not predict any marked pollution of the lower and middle troposphere of the mid Pacific. The development of long-range transport of polluted air masses was prevented by efficient ventilation of WCB by convection near the coast, the advection by the anticyclonic flow above the 700-hPa level, and by the downward motions in the region of Pacific high in the remote Pacific. During the other flight, the conveyor belts were located in remote ocean region. The pollution plume was split by rising air in the warm conveyor belt, which transported CO-poor air northward, as well as by the oceanic convection responsible for transport of clean air upward. These mechanisms led to dilution of East Asian pollutants in WCB en route of long-range transport to North America, and determined the episodic character of Asian outflow of polluted air masses by fragmenting the pollution plume.

2. Long-range transport and transformation of aerosol properties: numerical simulation

The research and developments completed in the past decades have caused a considerable progress in the studies of spatiotemporal distribution and properties of aerosol. These studies were mainly motivated by the need in reliable estimates of aerosol effect on climate and the role of aerosol as atmospheric pollution responsible, in particular, for degradation of visibility conditions. The most significant contribution to understanding of the processes of aerosol formation and aerosol properties came from satellite remote sensing of aerosol observed using such instrumentation as Stratospheric Aerosol and Gas Experiment (SAGE) sensors, MODerate-resolution Imaging Spectroradiometer (MODIS), and Multi-angle Imaging SpectroRadiometer (MISR), and in recent years from space-based lidar sensing. Pioneering works at the initial stage of remote sensing were performed by astronauts onboard manned spacecraft. Large-scale studies with the use of usual (ground-based, airborne, and balloon-borne) measurement facilities have played an important role in collection of complex information on aerosol, including data on aerosol microstructure and chemical composition.

An important step toward determination of interrelations between usual and satellite observation data, as well as spatiotemporal generalization of the results of ordinary observations, has been numerical simulation using three-dimensional regional- and global-scale models. Special concern has been given to the dynamics of sulfate aerosol (SA), formed predominately by oxidation of such gas precursors as sulfur dioxide (SO_2), dimethylsulfide (DMS), hydrogen sulfide (H_2S), and carbonyl sulfide (COS).

Kittaka et al.[16] undertook numerical simulation of long-range SA transport from Asian continent to the Pacific water basin using University of Wisconsin Nonhydrostatic Modeling System (UWNMS) in combination with the aqueous sulfur chemistry module. The calculations have been made for conditions of TRACE-P field observational experiment performed in winter–spring 2001 with the aim to study long-range transport, taking into account chemical conversion of aerosol and trace gas constituents, from North-Eastern Asia to the Pacific water basin. They considered data acquired during the period from March 7 to April 3, 2001, obtained in the regions of eastern half of China, Korean peninsula, southeastern Russia, Japanese islands, and Northwest Pacific.

The model covers 20-km atmospheric depth with the horizontal (vertical) resolution of 110 km (0.4 km). Comparison of numerically simulated results with observation data showed that the model overestimated the SA mixing ratio by 20% in the layer 1–6 km and underestimated it by about 30% near surface and by about 50% above 6 km. The calculations made for the case with convective clouds turned out to be in excellent agreement with the observations of background SA, but revealed underestimation of SA mixing ratio (by about two times) in the regions with the cloudy atmosphere. Numerical simulation showed that a large amount of SA (total mass of sulfate in the entire atmospheric volume considered varied in the range $(1.5–7.5)\cdot 10^6$ kg is released into the atmosphere after being produced in clouds through the oxidation of sulfur dioxide by hydrogen peroxide. Instantaneous SA mass in clouds was in the range $(0.2–3.5)\cdot 10^6$ kg, while the mass of sulfate washed out by precipitation was $(0.01–1.8)\cdot 10^6$ kg. Sulfate mass in ice clouds was two orders of magnitude smaller than that in water clouds. Results obtained by Kittaka et al.[16] indicate that the sulfate input to the atmosphere of Eastern Asian region exceeds the sulfate removed from the atmosphere through precipitation. Thus, this region in spring can be an important source of SA whose presence (with long-range transport taken into account) may compensate for the sulfate removal from the atmosphere (through aerosol scavenging by precipitation) in other regions of the globe.

Han et al.[10] developed atmospheric aerosol microstructure model that includes the main physical processes (aerosol formation, long-range transport, and dry and wet deposition). In combination with Regional Air Quality Model (RAQM), this model was used to simulate dust storms in Asia during the period March 15–24, 2002. Meteorological parameters were calculated using nonhydrostatic MM-5 mesoscale model. The numerical results are verified using available meteorological data and showed satisfactory model reliability in terms of adequate precalculation of evolution of dust aerosol and its long-range transport, as well as size-segregated aerosol mass distribution. One of the important features, both reproduced by numerical simulation and observed in experiment, is that, during long-range transport from DA source, the aerosol microstructure shifts the bulk of mass concentration from coarse to fine mode. The calculated maximum DA concentration, averaged over 10 days, in the region of China–Mongolia border was 3000 $\mu g/m^3$. Total dry DA deposition in the Gobi desert (along China–Mongolia border) reached 30 g/m^2. The spatial distribution and magnitude of dry deposition strongly varied and changed DA concentration and size-segregated dry deposition rate and the rate of scavenging. It is worthy to note that whereas dry deposition dominated in the process of removal of particles from the atmosphere in or near DA source, the role of the wet deposition intensified along the pathway of the long-range DA transport, with more efficient removal of coarse-mode particles (> 2 μm) and lower efficiency in the case of accumulation mode. Some 43.2 megatons, about 71% of the total mass of DA emissions, were redeposited onto the underlying surface by the dry deposition, and only 6% by the wet deposition. The remaining 23% stayed suspended in the atmosphere or experienced long-range transport.

Starting from 2000, in Japan a regular occurrence were events of elevated mineral DA content. The DA sources were dust storms in China deserts, though, on the other hand, the frequency of dust storms was found to decrease starting from the second half of 1970s. Mukai et al.[27] undertook a numerical simulation of the formation of dust storms and subsequent long-range DA transport in the region of East Asia in the period from 1981 to 2002. The calculations are made based on Spectral Radiation-Transport Model for Aerosol Species (SPRINTARS), developed in the Center for Climate System Research at the University of Tokyo and National Institute for Environmental Studies at the University of Kyushu. Evolution of fields of wind velocity, specific humidity, soil moisture, and the snow cover thickness is specified based on European Centre for Medium-Range Weather Forecasts reanalysis data.

Analysis of the results obtained showed that the dust emissions are substantially influenced by near-surface wind field, while in early spring the state of snow cover also turns out to be an important factor. The numerical simulation suggest that DA emissions to the atmosphere in Northeastern China strongly influence the DA mass concentration in the atmosphere of downwind regions, including cities of northeastern China, Korea, and Japan. When the frequency of dust events occurrence was high in Japan, a low-pressure system tended to develop over the northeast China region, favoring intensification of wind velocity. The numerical simulations indicate that in 2000–2001 the DA emissions in Takla Makan desert and Northwestern China decreased, while in Gobi desert and Northeastern region of China they increased. Therefore, it is worth considering that long-range DA transport was primarily from the second region due to the westerly transport to downwind areas in spring as was actually observed. Despite this general agreement between calculated and observed data, there are serious discrepancies in the estimates of frequency of occurrence of the atmospheric dust events and enhanced DA concentration that calls for further model refinement. Desertification taking place in the Northeastern China might also play a known role.

Growing ecological activity and resulting emissions of trace gases and aerosol to the atmosphere of the eastern region of Asia heighten the concern about the effect of anthropogenic emissions to the Pacific atmosphere, most clearly manifested in spring. Tu et al.[45] discussed the data of observations of long-range sulfur dioxide (SO_2) transport from East Asia in spring 2001 to the Central Pacific in the

Northern Hemisphere; the data were obtained during regular commercial flights under the NASA-supported mission on studying Transport and Chemical Evolution over the Pacific (TRACE-P). Among significant features of the SO_2 spatial distribution is the existence of layers with enhanced SO_2 content associated with low water vapor content and weak turbulence, usually isolated from marine boundary layer (MBL).

Very important factor of the spatial SO_2 distribution in March–April 2001 was the atmospheric dynamics. Analysis of long-range SO_2 transport trajectories has led to a conclusion that these SO_2-enriched layers could be the result of volcanic activity or anthropogenic SO_2 sources in East Asia. The trajectory data also indicate that air parcels usually ascend at about 2 km above the source, and then move to the east at midlatitudes (30°–60°N), reaching the Central region of the Pacific Ocean in 2 to 3 days. Sulfur dioxide, having undergone long-range transport at altitudes 2 to 4 km, dominates in the SO_2 concentration field over Central Pacific. Comparison with the numerical results, based on long-range transport model taking into account chemical conversion, has shown that SO_2 removal from the atmosphere is primarily caused by the processes occurring in clouds. Therefore, under clear-sky conditions in the atmosphere the distance of long-range transport will be larger if the trajectories are decoupled from the MBL. In March–April, 2001, an important factor influencing SO_2 concentration field was SO_2 emission by Miyake-jima volcano, whose contribution at times exceeded emission levels of the six anthropogenic SO_2 sources in East Asia.

Using regional chemical weather forecasting system (CFORS) model of long-range aerosol transport, taking into account aerosol chemical conversion along the transport pathway, Satake et al.[33] calculated the transport, chemical composition, and optical depth of tropospheric aerosol (in particular, such components as dust sulfate, carbonaceous species, and sea salt, contained in aerosol composition) for conditions of ACE-Asia field observational experiment performed in spring 2001. The calculated results were carefully compared with data of *in situ* measurements (PM-10 concentration, sulfate, and total carbonaceous aerosol), lidar sensing, and satellite observation data.

The comparison appeared, overall, to be quite satisfactory, and the results obtained revealed, in particular, the presence of latitudinal gradient of aerosol concentration. Analysis of the fields of aerosol and AOD, averaged over 2 months (March–April), even more clearly confirmed the presence of this latitudinal gradient, as well as the fact that dust aerosol is concentrated in latitude belt from 30 to 45°N. In contrast, the fields of sulfate and carbonaceous aerosol components experienced dominant influence of the aerosol sources in Central China and Southeastern Asia north of Japan (25–45°N latitudes). Analysis of horizontal aerosol transport demonstrated its close correlation with wind variability field, with each aerosol component being characterized by its own specific features of long-range transport. Main DA flow was oriented eastward along 45°N latitude, and was located in the free troposphere.

Sulfate and carbonaceous aerosols were concentrated in the atmospheric boundary layer with the presence of clockwise and divergent flow pattern over central China, which determined intensive arrival of polluted air masses at northern latitudes, and constrained continental outflow of air masses at southern latitudes. The specific long-range transport pathways along 30°N from Thailand and Laos were typical for carbonaceous aerosol at heights from 2 to 6 km. Estimates of regional budget of tropospheric aerosol led to conclusion that the total emissions of different aerosol types were: 105 Tg (dust), 8.3 Tg of SO_2 (73% due to industrial activity and 27% from volcanoes), and 3.07 Tg of carbonaceous aerosol. Dry deposition, gravitational settling, and northward outflow of dust accounted for 33%, 27%, and 14% of the total emissions, respectively. Wet deposition, eastward outflow, and dry deposition of sulfur accounted for 33%, 27%, and 21%, respectively. Regarding carbonaceous aerosol, its outflow to the east turned out to be most significant (49%), followed by dry deposition (16%) and outflow to the north (14%). Average contributions of different components to AOD were: 25% (sulfate), 24% (DA), and 15% (sea salt aerosol).

The regular dust storms in China are characterized by the presence of distinct interannual variability. Anomalous number of dust storms was recorded in 2000–2002 for total dust emissions in Eastern Asian region amounting to 100–200 Tg, equivalent to approximately 10% of the global emissions. Dust aerosol typically has distinct annual behavior, determined primarily by maximum soil erosion of Gobi desert in spring. Analysis of interaction between DA and aerosol formed due to industrial and agricultural activity becomes increasingly important nowadays. In the context of the field observational experiment (ACE-Asia), during the intensive observation period in spring 2001, Uno et al.[46] have developed a technique for numerical modeling of the long-range transport and chemical transformations of aerosol occurring during the transportation aimed at reproducing and interpreting the ACE-Asia observation data.

The regional "System for chemical weather forecasting (CFORS)" model was applied in combination with such auxiliary observation data as surface weather reports, satellite-derived Aerosol Index (AI)), results of ground-based measurements of aerosol characteristics, and data of aerosol lidar sensing. Analysis of CFORS data demonstrated that they quite reliably reproduce many observed features of long-range aerosol transport. In particular, the calculated vertical aerosol distribution well agrees with satellite data on AI and on data of observations with TOMS spectrometer used for mapping the total ozone content. The aerosol is transported in

accordance with meandering of synoptic-scale temperature field at the 500-hPa level.

The calculated aerosol optical depths differ from lidar data on AOD by no more than 20% in most of the cases. Analysis of numerical results has made it possible to analyze in detail the structure of "ideal" dust storm observed in early April. This dust storm consisted of two boundary layer components and one elevated-dust feature in the free troposphere above 6 km height, resulting from the effect of two large cyclonic systems. Consideration of calculated variations of DA concentration fields made it possible to correctly determine the onset time of dust storms for each observation site, however model results overestimated DA concentration at low latitude sites. Estimates of the dust flow due to horizontal DA transport along 130°E longitude in March–April gave the value 55.2 Tg (68% of DA at this time were emitted during three dust storms).

Based on modified Taiwan air quality model (TAQM-2), Tsai et al.[44] calculated masses of coarse (particle diameter greater than 1 μm) and submicron (diameter < 1 μm) aerosol fractions in order to reproduce the results of airborne observations of aerosol properties, in the framework of ACE-Asia in spring 2001 in the Northwestern Pacific. Comparison with observation data showed that the calculated vertical profiles of submicron aerosol concentration reasonably well agree with observations during 19 flights.

During dust storm events and at the level of maximum DA concentration, the calculated dust fraction in coarse aerosol content usually exceeded 90%, while DA was concentrated at the altitudes in the lower troposphere mainly below 6 km. Even in the absence of dust storms, the dust component of coarse aerosol fraction was still dominant above the ABL. The numerical results suggest that during the airborne observations the dominating components in the submicron particle composition were products from atmospheric pollution. The calculated vertical profiles of submicron particle concentrations were sensitive to the emission inventory of air pollutants over East Asia. The correlation between observed content of anthropogenic volatile organic compounds and submicron particle concentration was used to estimate the pollution fraction in the submicron particles, and the results were consistent with the model calculations of dust aerosol fraction. Model estimates suggest that the dust fraction in submicron aerosol of ABL generally was less than 28%. During dust storm the dust fraction was greater than 40% but could decrease to 24% when quite large amount of pollutants was present.

Tang et al.[43] performed numerical simulation of long-range transport and chemical conversion of atmospheric aerosol in Eastern Asian region for conditions of TRACE-P and ACE-Asia field observational experiments, with the purpose of analysis of observation data obtained during these experiments. The dynamics of chemical processes is reproduced using regional chemical transport model STEM-2K3, which includes the on-line gas–aerosol thermodynamic module SCAPE-II; this model describes the interaction of trace gases with aerosol and explicitly considers chemical aging of dust aerosol.

Analysis of calculated results demonstrated that numerical modeling quite adequately reproduces many features of the observed three-dimensional spatiotemporal variations of the aerosol properties. Powerful outflow of dust aerosol in March–April 2001, due to dust storms in China, has led to strong increase of atmospheric dust loading. In contrast, under conditions of low atmospheric dust loading, the interaction between trace gases and aerosol during long-range transport was determined by SO_2 condensation and gas-phase ammonium distribution. This situation was illustrated by detailed discussion of two series of airborne observations during TRACE-P.

The numerical results suggest that the presence of DA alters partitioning of partially volatile components between gas and aerosol phases, as well as the microstructure of the secondary aerosol. Calcium in DA affects the gas–aerosol equilibrium, manifesting itself in the shift of equilibrium balance toward anion-limited state, favoring uptake of sulfate and nitrate by aerosol particles, but reduces the amount of aerosol ammonium. Chemical reactions on the surface of DA particles provide an additional mechanism of formation of aerosol nitrate and sulfate. The DA microstructure plays a decisive role in the formation of the secondary aerosol microstructure. Since most of the mass (70–90%) is concentrated in coarse aerosol fraction, considerable part of sulfate and nitrate is found precisely in the composition of this fraction. The observation data show that during dust storm events, about 10–30% of sulfate is in the coarse fraction, while for nitrate this fraction reaches 80%.

Using data of numerical simulations, supported by airborne observations (from a C-130 research aircraft), during ACE-Asia period, on April 4–14, 2001, Tang et al.[42] studied the spatial distribution and chemical composition of dust aerosol, as well its effect on gas-phase chemical processes in the troposphere. The calculations were made using complex STEM-2K1 model enabling one to reproduce long-range aerosol transport with consideration of aerosol chemical conversion during the transportation.

Overall, the numerical results reasonably well agree with the available data of ground-based, airborne, and satellite observations. In this period, dust storms were initialized in the Takla Makan and Gobi deserts, and the resulting dust aerosol underwent long-range transport during which it suffered chemical conversions. Also of importance were additional aerosol supplies (along long-range transport pathways) due to dust emissions from Loess Plateau and in agricultural regions. Data of airborne observations of aerosol chemical composition are

analyzed for the cases of high- and low-DA content in the atmosphere.

Submicron fraction of dust-rich air masses was characterized by elevated concentration ratios $\Delta Ca/\Delta Mn$, $\Delta NH_4^+/\Delta SO_4^{2-}$, and $\Delta NO_3^-/\Delta CO$ (Δ is the difference between observed and background concentrations). Authors of Ref. 42 studied DA sensitivity to heterogeneous reactions involving O_3, NO_2, SO_2, and HNO_3. These reactions have a considerable effect on regional-scale chemical processes. For instance, low tropospheric ozone concentration observed during one of the flight can be explained by the influence of heterogeneous reactions. Calculated data averaged over entire observation period showed that in the near-surface atmospheric layer heterogeneous reactions have led to a decrease of O_3, SO_2, NO_2, and HNO_3 concentrations by 20, 55, 20, and 95%, respectively. In addition, in the regions of atmospheric pollution there could take place increases of NO and HONO concentrations and daytime abundance of OH by 20, 30, and 4%, respectively.

Under conditions, when DA mixed with new pollutants, the heterogeneous reactions could lead to complex and diverse responses of the photochemical system. In addition, these reactions alter chemical composition of aerosol of different size fractions. Under heavy DA loadings, the heterogeneous reactions may lead to increase of sulfate (> 20%) and nitrate (> 70%) contents in coarse aerosol fraction. The photochemical processes can also be influenced by DA-caused changes of the radiative fluxes. For instance, near the underlying surface the OH concentration may decrease by 20%. However, the radiative effect appears to be weaker than the effect of heterogeneous reactions in most cases.

Conclusion

Implementation of ACE-Asia program is an illustration of considerable progress reached during further development of complex observational experiments in context of aerosol–clouds–climate problem. However, the completeness and systematic character of these field experiments are still insufficient. First, the experimental completeness in terms of incorporation of different aerosol types and properties are of a primary concern, as well as those trace gas and cloud characteristics which are important for adequate simulation of aerosol–trace gases–clouds interactions. Most pressing, probably, is fragmentary character of observations of radiative characteristics of aerosol and atmosphere as a whole. This issue will the subject of the third part of the overview.

References

1. A. de Almeida Castanho, J.V. Martins, P.V. Hobbs, P. Artaho, L. Remer, M. Yamasoe, and P.R. Colarco, J. Atmos. Sci. **62**, 934–946 (2005).
1a. R. Arimoto, X.Y. Zhang, B.J. Huebert, C.H. Kang, D.L. Savoie, J.M. Prospero, S.K. Sage, C.A. Schloesslin, H.M. Khaing, and S.N. Oh, J. Geophys. Res. **109**, No. 19, D19504/1–D19504/15 (2004).
2. T.S. Bates, P.K. Quinn, D.J. Coffman, D.S. Covert, T.L. Miller, J.E. Johnson, G.R. Carmichel, I. Uno, S.A. Guazzotti, D.A. Sodeman, K.A. Prather, M. Rivera, L.M. Russel, and J.T. Merrill, J. Geophys. Res. **109**, No. 19, D19519/1–D19519/18 (2004).
2a. R.W. Bergstrom, P. Pilewskie, J. Pommier, M. Rabette, P.B. Russel, B. Schmidt, J. Redemann, A. Higurashi, T. Nakajima, and P.K. Quinn, J. Geophys. Res. **109**, No. 19, D19515/1–D19515/13 (2004).
2b. J. Chowdhary, B. Cairns, M.I. Mishchenko, P.V. Hobbs, G.F. Cote, J. Redemann, K. Rutledge, B.N. Holben, and E. Russel, J. Atmos. Sci. **62**, 1093–1117 (2005).
3. A.D. Clarke, Y. Shinozuka, V.N. Kapustin, S. Howell, B. Huebert, S. Doherty, T. Anderson, D. Covert, J. Anderson, X. Hua, G. Moore, II, C. McNaughton, C. Carmichel, and R. Weber, J. Geophys. Res. **109**, No. 15, D15509/1–D15509/20 (2004).
4. D.D. Cohen, D. Garton, E. Stelcer, O. Hawas, T. Wang, S. Poon, J. Kim, B.C. Chui, S.N. Oh, H.-J. Shin, M.Y. Ko, and M. Uematsu, J. Geophys. Res. **109**, No. 19, D19512/1–D19512/18 (2004).
5. D. Copsey, Weather **60**, No. 7, 273–274 (2005).
6. L.J. DeBell, R.W. Talbot, J.E. Dibb, J.W. Munger, E.V. Fischer, and S.E. Frolking, J. Geophys. Res. **109**, No. 19, D19305/1–D19305/16 (2004).
7. S. Deepshikla, S.K. Satheesh, and J. Srinivasan, Geophys. Res. Lett. **32**, L03811, doi: 10.1029/2004 GL022091 (2005).
8. A. Eldering, B.H. Kahn, F.P. Mills, F.W. Irion, H.M. Steele, and M.R. Gunsor, J. Geophys. Res. **109**, No. 20, D20201/1–D20201/11 (2004).
9. S. Glagolenko and D.J. Phares, J. Geophys. Res. **109**, No. 18, D18205/1–D18205/12 (2004).
10. Z. Han, H. Ueda, K. Matsuda, R. Zhang, K. Arao, Y. Kanai, and H. Hasome, J. Geophys. Res. **109**, No. 19, D19205/1– D19205/22 (2004).
11. B. Huebert, T. Bertram, J. Kline, S. Howell, D. Eatough, and B. Blomquist, J. Geophys. Res. **109**, No. 19, D19511/1–D19511/16 (2004).
12. L. Husain, P.P. Parekh, V.A. Dutkiewicz, A.R. Khan, K. Yang, and K. Swami, J. Geophys. Res. **109**, No. 18, D18305/1–D18305/12 (2004).
12a. A. Ignatov, P. Minnis, N. Loeb, B. Wielicki, W. Miller, S. Sun-Mack, D. Tanre, L. Remer, I. Laszlo, and E. Geier, J. Atmos. Sci. **62**, 1008–1031 (2005).
13. R. Kahn, J. Anderson, T.L. Anderson, T. Bates, F. Brechtel, C.M. Carrico, A. Clarke, S.J. Doherty, E. Dutton, R. Flagan, R. Frouin, H. Fukushima, B. Holben, S. Howell, B. Huebert, A. Jefferson, H. Jonsson, O. Kalashnikova, J. Kim, S.-W. Kim, P. Kus, W.-H. Li, J.M. Livingston, C. McNaughton, J. Merrill, S. Mukai, T. Murayama, T. Nakajima, P. Quinn, J. Redemann, M. Rood, P. Russel, I. Sano, B. Schinid, J. Seinfeld, N. Sugimoto, I. Wang, E.J. Welton, J.-G. Won, and S.-C. Yoon, J. Geophys. Res. **109**, No. 19, D19514/1–D19514/30 (2004).
14. D.-H. Kim, Sohn B.-J., T. Nakajima, Ta. Takamura, To. Takemura, B.-C. Choi, and S.-C Yoon, J. Geophys. Res. **109**, No. 2, D02209/1–D02209/18 (2004).
15. Y.S. Kim, Iwasaka Y., G.-Y. Shi, T. Nagatani, T. Shibata, D. Trochkin, A. Matsuki, M. Yamada, B. Chen, D. Zhang, M. Nagatani, and H Nakata, J. Geophys. Res. **109**, No. 19, D19526/1–D19526/9 (2004).
15a. K.W. Kim, Z. He, and Y.J. Kim, J. Geophys. Res. **109**, No. 19, D19502/1–D19502/15 (2004).
16. C. Kittaka, R.B. Pierce, J.H. Crawford, M.H. Hitchman, D.R. Johnson, G.J. Tripoli, M. Chin, A.R. Bandy,

R.J. Weler, R.W. Talbot, and B.E Anderson, J. Geophys. Res. **109**, No. 15, D15511/1–D15511/14 (2004).
17. J. Kline, B. Huebert, S. Howell, B. Blomquist, J. Zhuang, D. Bertram, and J Carrillo, J. Geophys. Res. **109**, No. 19, D19508/1–D19508/22 (2004).
18. K.Ya. Kondratyev, L.S. Ivlev, V.F. Krapivin, and C.A Varotsos, *Atmospheric Aerosol Properties, Formation Processes, and Impacts: From Nano- to Global Scales* (Springer/Praxis, Chichester, U.K., 2005), 382 pp.
19. K.H. Lee, Y.J. Kim, and W. von Hoyningen-Huene, J. Geophys. Res. **109**, No. 9, D19516/1–D19516/14 (2004).
19a. R.C. Levy, L.A. Remer, J.V. Martins, Y.J. Kaufman, A. Plana-Fattori, J. Redemann, and B. Wenny, J. Atmos. Sci. **62**, 974–992 (2005).
20. Y. Ma, R.J. Weber, K. Maxwell-Meier, D.A. Orsini, Y.-N. Lee, B.J. Huebert, S.G. Howell, T. Bertram, R.W. Talbot, J.E. Dibb, and E. Scheuer, J. Geophys. Res. **109**, No. 15, D15506/1–D15506/13 (2004).
20a. B.I. Magi, P.V. Hobbs, T.W. Kirchstetter, T. Novakov, D.A. Hegg, S. Gao, J. Redemann, and B. Schmidt, J. Atmos. Sci. **62**, 919–933 (2005).
21. C. Mari, M.J. Evans, P.I. Palmer, D.J. Jacob, and G.W. Sachse, J. Geophys. Res. **109**, No. 15, D15517/1–D15517/12 (2004).
21a. J.V. Martonchik, D.J. Diner, R. Kahn, B. Gaitley, and B.N Holben, Geophys. Res. Lett. **31**, L1602, doi:10.1029/2004GL019807 (2004).
22. K. Maxwell-Meier, R. Weber, C. Song, D. Orsini, Y. Ma, G.R. Carmichel, and D.G Streets, J. Geophys. Res. **109**, No. 19, D19507/1–D19507/20 (2004).
23. C.S. McNaughton, A.D. Clarke, S.G. Howell, K.G. Moore (II), V. Brekhovskikh, R.J. Weber, D.A. Orsini, D.S. Covert, G. Buzorius, F.J. Brechtel, C.R. Carmichael, Y. Tang, F.L. Eisele, R.L. Mauldin, A.R. Bandy, D.C. Thornton, and B. Blomquist, J. Geophys. Res. **109**, No. 19, D19506/1–D19506/19 (2004).
24. J.T. Merrill and J. Kim, J. Geophys. Res. **109**, No. 19, D19518/1–D19518/13 (2004).
25. K.G. Moore, II, A.D. Clarke, V.N. Kapustin, C. McNaughton, B.E. Anderson, E.L. Winstead, R. Weber, Y. Ma, Y.N. Lee, R. Talbot, J. Dibb, T. Anderson, S. Doherty, D. Covert, and D. Rogers, J. Geophys. Res. **109**, No. 15, D15515/1–D15515/35 (2004).
26. K.K. Moorthy and S.K. Satheesh, J. Roy. Meteorol. Soc. 2005 (in press).
27. M. Mukai, T. Nakajima, and T. Takemura, J. Geophys. Res. **109**, No. 19, D19204/1–D19204/19 (2004).
28. M.H. Park, Y.R. Kim, C.-H. Kang, and S.-G. Shim, J. Geophys. Res. **109**, No. 19, D19513/1–D19513/7 (2004).
29. P.K. Quinn, O.J. Coffman, T.S. Bates, E.J. Welton, D.S. Covert, T.L. Miller, D.E. Johnson, S. Maria, L. Russel, R. Arimoto, C.M. Carrico, M.J. Rood, and J. Anderson, J. Geophys. Res. **109**, No. 19, D19501/1–D19501/28 (2004).
30. S. Ramachandran, J. Geophys. Res. **109**, No. 19, D19207/1–D19207/11 (2004).
31. S. Ramachandran, J. Geophys. Res. **109**, No. 19, D19208/1–D19208/12 (2004).
31a. J. Redemann, B. Schmid, J.A. Eilers, R. Kahn, R.C. Levy, P.B. Russel, J.M. Livingston, P.V. Hobbs, W.L Smith. (Jr), and B.N. Holben, J. Atmos. Sci. **62**, 993–1007 (2005).
32. J.L. Santarpia, R. Li, and D.R. Collins, J. Geophys. Res. **109**, No. 18, D18209/1–D18209/16 (2004).
33. S. Satake, I. Uno, T. Takemura, G.R. Carmichel, Y. Tang, D. Streets, N. Sugimoto, A. Shimizu, M. Hematsu, J.-S. Han, and S. Ohta, J. Geophys. Res. **109**, No. 19, D19522/1– D19522/11 (2004).
34. S.K. Satheesh and J. Srinivasan, Geophys. Res. Lett. **29**, No. 18, 1874, doi: 10/1029/2002 GL015687 (2002).
35. H. Satsumabayashi, M. Kawamura, T. Katsuno, K. Futaki, K. Murano, G Carmichel. R., M. Kajino, M. Horiguchi, and H. Ueda, J. Geophys. Res. **109**, No. 19, D19202/1–D19202/17 (2004).
36. M. Schaap, H.A.C. Van Der Gon, F.J. Dentener, A.J.H. Visschedijk, M. Van Loon, H.M. ten Brink , J.-P. Putaud, B. Guillaume, C. Lioucse, and P.J.H. Builtjes, J. Geophys. Res. **109**, No. 18, D18207/1–D18207/16 (2004).
37. S. Sharma, D. Lovoué, H. Cachier, L.A. Barrie, and S.L Gong, J. Geophys. Res. **109**, No. 15, D15203/1–D15203/10 (2004).
38. A. Shimizu, N. Sugimoto, I. Matsui, K. Arao, I. Uno, T. Murayama, N. Kagawa, K. Aoki, A. Uchiyama, and A Yamazaki, J. Geophys. Res. **109**, No. 19, D19517/1–D19517/14 (2004).
39. H. Sievering, J. Cainey, M. Harvey, J. McGregor, S. Nichol, and P. Quinn, J. Geophys. Res. **109**, No. 19, D19317/1– D19317/12 (2004).
40. B.R.T. Simoneit, M. Kobayashi, M. Mochida, K. Kawamura, and B.J. Huebert, J. Geophys. Res. **109**, No. 19, D19509/1–D19509/13 (2004).
41. B.R.T. Simoneit, M. Kobayashi, M. Mochida, K. Kawamura, M. Lee, H.-J. Lim, B.J. Turpin, and Y. Komazaki, J. Geophys. Res. **109**, No. 19, D19510/1–D19510/22 (2004).
41a. W.L. Smith (Jr.), T.P. Charlock, R. Kahn, J.V. Martins, L.A. Remer, P.V. Hobbs, J. Redemann, and C.K Rutledge, J. Atmos. Sci. **62**, 903–918 (2005).
42. Y. Tang, G.R. Carmichel, G. Kurata, I. Uno, R.J. Weber, C.-H. Song, S.K. Guttikunda, J.-H. Woo, D.G. Streets, C. Wei, A.D. Clarke, B. Huebert, and T.L. Andersen, J. Geophys. Res. **109**, No. 19, D19521/1–D19521/21 (2004).
43. Y. Tang, G.R. Carmichel, J.H. Seinfeld, D. Dabdub, R.J. Weber, B. Huebert, A.D. Clarke, S.A. Guazzotti, D.A. Sodeman, K.A. Pratter, I. Uno, J.-H. Woo, J.J. Yienger, D.G. Streets, P.K. Quinn, J.E. Johnson, C.-H. Song, V.H. Grassian, A. Sandu, R.W. Talbot, and J.E. Dibb, J. Geophys. Res. **109**, No. 19, D19523/1–D19523/32 (2004).
44. F. Tsai, T.-H. Liu, S.C. Liu, T.-Y. Chen, T.L. Anderson, and S.J. Masonis, J. Geophys. Res. **109**, No. 19, D19520/1–D19520/17 (2004).
45. F.H. Tu, O.C. Thornton, A.R. Bandy, G.R. Carmichael, Y. Tang, K.L. Thornhill, G.W. Sachse, and D.R. Blake, J. Geophys. Res. **109**, No. 15, D15518/1–D15518/17 (2004).
46. I. Uno, S. Satake, G.R. Carmichel, Y. Tang, Z. Wang, T. Takemura, N. Sugimoto, A. Shimizu, T. Murayama, T.A. Cahill, S. Cliff, M. Uematsu, S. Ohta, P.K. Quinn, and T.S. Bates, J. Geophys. Res. **109**, No. 19, D19524/1–D19524/20 (2004).
47. T. Wang, C.H. Wong, T.F. Cheung, D.R. Blake, R. Arimoto, K. Baumann, J. Tang, G.A. Ding, X.M. Yu, Y.S. Li, D.G. Streets, and I.J. Simpson, J. Geophys. Res. **109**, No. 19, D19505/1–D19505/16 (2004).
48. J. Xu, M.H. Bergin, R. Greenwald, J.J. Schauer, M.M. Shafer, J.L. Jaffrezo, and G. Aymoz, J. Geophys. Res. **109**, No. 19, D19303/1–D19303/14 (2004).
49. Y. Yin, K.S. Carlslaw, and G. Feingold, Quart. J. Roy. Meteorol. Soc. **131**, 221–245 (2005).
50. D. Zhang and Y. Iwasaka, J. Geophys. Res. **31**, No. 15, L15101/1–L15102/4 (2004).