From nano- to global scales: properties, processes of formation, and aftereffects of atmospheric aerosol impacts. 1. Field observational experiments. Africa and Asia

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Results of complex investigations of the atmospheric aerosol properties, obtained during observational experiments in Africa and Asia, are analyzed.

Introduction

The growing interest to the atmospheric aerosol from the point of view of its effect on climate, chemical heterogeneous reactions in the atmosphere affecting the quality of the environment, atmospheric visibility, and human health manifests itself in unusually extent investigations. Therefore, following the recent thorough reviews on the problem,[2–8] a demand arose to analyze the latest results, concerned, first of all, with three aspects: 1) aerosol properties on evidence derived from complex field observational experiments; 2) processes of aerosol formation and interaction with clouds; 3) aerosol as a climate-forming component of the atmosphere (including the problem of long-range transport). In the wake of the topical issue of *Atmospheric and Oceanic Optics* (**17**, Nos. 5–6, 2004) and the previous publication,[9] which discussed the results of investigation of Siberian aerosols, other various results are considered in this paper.

1. Saharan Dust Experiment (SHADE)

The first combined observational experiments aimed at aerosol-radiation studies of the desert dust aerosol were started in October 1970 in the Ashkhabad region (Turkmenia) within the framework of the CAENEX Project and then have received a further wide development.[2,26–28] One of the newest similar investigations is connected with the SHADE field experiment due to the key role of the Sahara Desert as a most powerful source of natural dust aerosol (DA).[43,48]

Haywood with co-workers[22] have analyzed the physical and optical properties of the Saharan dust aerosol measured aboard the Met Office C-130 aircraft-laboratory during the SHADE. Four special-purpose flights were carried out from Sal, Cape Verde Islands on September 21, 24, 25, and 28 of 2000. The main sources of information were the observations with the airborne instruments: 1) PSAP photometer for measuring short-wave (567 nm) radiation (SWR) absorption; 2) TSI-3563 nephelometer (aerosol scattering at three wavelengths: 450, 550, and 700 nm); 3) spectrometer for measurement of aerosol and cloud particle radii in the range 1–23.5 μm; 4) broadband Eppley pyranometers for wavelength ranges 300–3000 and 700–3000 nm; 5) 16-channel Scanning Airborne Filter Radiometer (SAFIRE) for spectral measurements of shortwave radiation; 6) short-wave spectrometer (SWS) directed in nadir for measurements in ranges 300–950 and 950–1700 nm (spectral resolution of 10 and 18 nm, respectively).

The processing of the measurements enabled determination of the aerosol optical depth $\tau_{\mathrm{aer}\,\lambda}$ and the direct radiative effect (DRE) of the mineral dust. The results showed a significant overestimation of the SWR absorption by dust aerosol when setting standard refractive indices. The new measurements suggested that an imaginary part of the refractive index of $0.0015i$ is appropriate at $\lambda = 550$ nm.

Different methods for determining $\tau_{\mathrm{aer}\,\lambda}$ and corresponding errors were discussed in Ref. 22. For maximal dust loading of the atmosphere, $\tau_{\mathrm{aer}\,\lambda = 550} = 1.48 \pm 0.05$. Under these conditions, an instantaneous value of DRE was $\approx (-129 \pm 5)$ W/m^2, which corresponded to (2.7 ± 0.1) times enhancement of albedo of the system “underlying surface – atmosphere”. A comparison of the obtained DRE with the airborne data obtained in the framework of the CERES program demonstrated an agreement to a factor of 1.2. The results obtained suggested that the DRE due to the Saharan dust aerosol should be taken into account in studies of the global radiation budget.

The aircraft measurements during the SHADE were also used to quantify the effect of the Saharan dust aerosol on the long-wave radiation (LWR) transport in the atmosphere.[23] Comparing data for dusty and clear (free of clouds) atmospheres revealed a distinct signature of the dust aerosol in the upwelling and downwelling LWR radiation spectra. The comparison of the measured values of spectral LWR fluxes with simulated based on the simultaneous measurements, has allowed a retrieval of the DA refractive index in the IR spectral region, because the degree of agreement between observed and simulated data depends on the choice of the refractive index, the mass loading, and the altitude of the dust layer. The aerosol microstructure has a less effect when considering large particles (radius greater than 1 μm).

The effect of the dust aerosol (DA) layer showed itself in the form of relative warming with a rate of 0.5 K/day below the dust layer and relative cooling up to 0.5 K/day above it. These radiation changes of the temperature are by about an order of magnitude lower than the corresponding changes due to the short-wave absorption. In the field of long-wave radiation (LWR) fluxes, the effect of the dust outbreak on irradiance was a decrease (6.5 W/m^2) in upwelling LWR at the top of the atmosphere and an increase (11.5 W/m^2) in downwelling LWR at the surface. The aircraft measurements have shown that the dust causes a 2–4 K decrease in the brightness temperature in the window region, that agrees well with corresponding space-borne measurements.

Estimates of the globally averaged data on direct radiation impact of the dust aerosol gave a wide spread of values from +0.09 to –0.46 W/m^2 because of uncertainty of data on the DA chemical composition, microstructure, and content. An important factor is that the DA particle diameters can achieve several micrometers, and the imaginary part of the refractive index is nonzero, that is, DA is the atmospheric constituent, which not only absorbs, but also scatters the shortwave atmospheric radiation. Another, no less important function of DA is that it significantly affects the properties of clouds and the gas composition of the atmosphere. Being a part of the sea-salt aerosol, DA is simultaneously the most important component of the atmospheric aerosol as well. The estimates of the DA content in the atmosphere gave the values of 14–41 Tg with their pronounced spatiotemporal variability.

During the Saharan Dust Experiment in September 2000, Formenti with co-workers[14] collected filter samples aboard the C-130 aircraft over the Atlantic Ocean between Sal Island and Senegal in order to study the chemical composition of aerosol. Dust was found in the altitude range from 0.5–1 to 4–4.5 km, and within the main dust plume, sublayers with different particle size distributions and different originations were observed. Inverse correlation between O_3 and dust in the troposphere suggests a removal of O_3 by DA. Dust loadings containing up to 54 $\mu g/m^3$ (submicron plus supermicron fractions) were measured during particularly intense dust events. Mixing of dust with anthropogenic aerosols, mainly NH_4HSO_4, was observed in the fine fraction and attributed to air masses originated from Europe and then traveled over Northern Africa.

The ratio of nitrate to non-sea-salt (nss) sulfate in DA was around 0.3. This argues against any significant influence of biomass burning emissions, which have much higher nitrate concentration. However, in some cases, traces of fossil fuel pollution plumes, likely from North America, were observed. The geochemical signature of DA was consistent with previous results in the area. Si, Fe, and Ti were not enriched with respect to the soil composition, while other elements, such as Ca and S, were. Although Ca is prevalently present in the form of calcite in African soils, it is also found as calcium sulfate in the atmosphere.

Summarizing the results, Tanré with co-authors[45] have briefly characterized the program, observational instruments, and the results of the SHADE *in situ* observational experiment, whose main goal was to study the properties of Saharan dust aerosol transported to the Atlantic Ocean from the Northern Africa in the periods of dust storms in the Sahara Desert. The main observational facilities were various airborne instruments of two aircraft-laboratories flying in the Ñàðe Verde area in September 19–29 of 2000.

In addition to already mentioned devices, the onboard instrumentation included: standard sensors for measuring meteorological parameters, Passive Cavity Aerosol Spectrometer Probe 100X (PCASP), Fast Forward Scattering Spectrometer Probe (FFSSP) (however, FFSSP did not operate reliably during the campaign), Small Ice Detector (SID), aerosol filter sampling facilities, and Fourier Transform spectrometer for obtaining high-resolution (about 0.5 cm^{-1}) spectral data on SWR.

The French Mystere-20 (M-20) aircraft was equipped with Eppley broadband radiometers with standard *in situ* sensors of pressure, temperature, and humidity; two-wave (532 nm and 1064 nm) lidar LEANDRE-1, and the airborne version of POLDER for polarization and brightness measurements between 440 and 865 nm. Two sun photometers of the Aerosol Robotic Network located in Sal Island (14°43′N, 25°56′W) and in M'Bour (16°43′N, 22°56′W) operated during the SHADE experiment. The M-20 observations were coordinated with data obtained from the MODIS instrument set on the TERRA platform.

As was already mentioned, the chemical analysis has shown that aerosol under study was not contaminated by biomass burning products, but in a minor extent by anthropogenic pollution. The typical microstructure of aerosol in the offshore Senegal had a double-peaked size distribution centered at roughly 0.40 μm and 1.5–2.0 μm. The nonsphericity of dust particles in the inversion scheme (in calculations of the SWR field) was taken into account by assuming a spheroidal particle shape. The measurements suggested much less absorption than the values derived earlier, i.e., the single scattering albedo turned to be 0.90 ± 0.02 in the blue region of spectrum, 0.97 ± 0.02 at 670 nm, and almost zero near infrared.

The maximal direct radiation effect was shown to reach instantaneous values of approximately –130 W/m^2 over ocean in the solar spectrum on the 25th of September. Given the measured aerosol optical thickness of approximately 1.5 at 550 μm, the normalized direct radiative effect is approximately – 90 W/m^2 per unit aerosol optical thickness. At the surface level, the radiative impact is around 1.6 times as higher as that at the top of the atmosphere, which is smaller than recent results derived over the Indian Ocean. The dust radiative impact in the IR is of the order of 10% of that in the short waves and always is of positive sign. According to the results of numerical simulation, in the area around the Sahara (0°N–30°N and 60°W–40°E) the aerosol radiative impact varies around –8.0 to –10.0 W/m^2 for clear skies and around –6.0 W/m^2 when clouds are considered with a corresponding LWR impact of +1.0–1.2 W/m^2 and +0.7–0.9 W/m^2, respectively. The global radiative impact of the Saharan DA was estimated as approximately –0.4 W/m^2.

In connection with DA investigations, a particular attention was paid to improvement of measurement techniques for aerosol profiling. In particular, Léon with co-authors[30] have discussed the improvement in retrieval capabilities of profiles of aerosol optical properties using a synergy between passive and active (lidar) remote sensing techniques, keeping in mind the near-term outlooks of satellite lidar sensing with such perfect instruments as Moderate resolution Imaging Spectrometer (MODIS) and the Polarization and Directionality of Earth's Reflectance (POLDER). The lidar data on the spectral backscattering coefficient between 532 and 1064 nm were used to determine the profile of backscatter-to-extinction ratio, and then the effective radius of an assumed bimodal aerosol size distribution was retrieved as a function of the altitude.

The analysis of the algorithm sensitivity to the input parameters has demonstrated the inversion procedure to be robust relative to noise detection and calibration errors. The algorithm has been tested during the Saharan dust experiment, which took place in the northeastern tropical Atlantic in September 2000 as a part of SHADE. The vertical profile of extinction agreed well with *in situ* measurements of the aerosol extinction. Profiles derived from lidar measurements on the 25th of September highlighted a presence of the aerosol layer located between 2.2 and 4.5 km with particle effective radii of 1.19 ± 0.6 μm. Another dust layer within the sub-Saharan transition layer over the marine boundary layer was also observed, with particle radii significantly smaller than those within the Saharan aerosol layer.

Starting from the mid-1980s, a series of airborne and ground-based experiments was carried out with support of the European Space Agency (ESA) in order to analyze the potentiality of spaceborne imaging spectroscopy for different scientific applications. In particular, the Digital Airborne Imaging Spectrometer Experiment (DAISEX) involved simultaneous data acquisitions using different airborne imaging spectrometers over test sites in the southeast Spain (Barrax) and the Upper Rhine valley (Colmar, France, and Hartheim, Germany). The results obtained were quite important for provision of successful functioning of the MERIS imaging spectrometer aboard the Envisat satellite.

Pedrós with co-authors[40] have discussed the results of ground-based spectroradiometer (two Licor 1800 and one Optronic OL-754) measurements over the Barrax area during the DAISEX campaign days in 1998, 1999, and 2000 in order to retrieve the aerosol optical thickness (AOT). The analysis of the visible data has shown in all cases a predominant contribution of the coarse-particle mode. The analysis of the back trajectories of the air masses has indicated a predominance of marine-type aerosols with smaller continental aerosols in the lower atmospheric layers in all cases.

To characterize chemically end products of transformation of the Saharan dust (end-member), Guieu with co-workers[21] used two types of samples: 1) fine fractions of surface soil from Northern Africa; 2) aerosol phases of typical Saharan rains. In the latter case, to estimate the initial composition of particles entering the Saharan rains, they were corrected from the dissolution losses in the cloud water.

The analysis of particles for the content of Al, Fe, Đ, and Đb suggested that (except for lead) the chemical composition of the transported Saharan dust was more homogeneous than the composition of individual soils. As confirmed by the air mass back trajectories, the higher level of homogeneity of the aerosol was partly due to the fact that dust formation events covered large areas of the Saharan desert, and the composition of the particles reflected the average composition of the eroded areas. The Pb concentration in the transported dust reflected clearly an anthropogenic fraction. By using Pb/Al measurements from the soils it was shown that typical Saharan rain composition is not free of mixing with air masses from Europe.

The following values were proposed to characterize the Saharan dust end-member: Al (%) = 7.09 ± 0.79; Fe (%) = 4.45 ± 0.49; P (%) = = 0.082 ± 0.011; Pb (ppm) = 24 ± 9. As the most typical characteristics of the DA composition, the ratios of trace gas components (TGC) to Al or Fe should be treated. The Saharan dust represents a potential source of nutrients (P, Fe) for the Mediterranean water. Indeed, it accounts for about 30–40% of the total atmospheric flux of phosphorus and about 90% of iron in the western Mediterranean, and it governs the biogeochemical cycle of iron being the main source of dissolved iron in the western Mediterranean waters.

Observations in regions neighboring Northern Africa have become the sources of various information about the properties of Saharan dust aerosol. Thus, for example, observations over Leipzig (51.3°N, 12.4°E), Germany, revealed extremely high values of the extinction-to-backscatter (lidar) ratio with the income of Saharan aerosol. The initial observations of the dust plume on October 13 to 15, 2001, suggested that the high values of the lidar ratio were caused by the presence of nonspherical particles of atmospheric aerosol. For more reliable interpretation of the data, Müller and co-authors[37] have considered the information about the geometric and optical characteristics of the dust aerosol plume based on combined observations with a dual-wave Raman lidar (Nd:YAG laser with the pulse repetition period of 30 s) and a sun photometer (λ = 340, 380, 440, 500, 670, 870, 940, and 1020 nm) located in Leipzig.

This observation site is a part of the European Aerosol Research Lidar Network (EARLINET). Automated sun photometer observations of aerosol optical depth and sky brightness were made in the framework of the worldwide operating Aerosol Robotic Network (AERONET). The analysis of the data obtained has shown that the dust plume reached a top height of 6 km, and the columnar aerosol optical depth at 532 nm increased from 0.25 on 13 October 2001 to a maximum of ~ 0.63 on 14 October 2001. According to lidar observations, up to 90% of the optical depth at λ = 532 nm was contributed by the dust layer located above heights of 1000 m.

It follows from sun photometer observations that the Angström exponents between 380 and 1020 nm were about 0.45 at the beginning of the dust period, and dropped to a minimal value of 0.14 during the peak of the dust outbreak. Vertically resolved Angström exponents derived from lidar profiles of the extinction coefficients at 355 and 532 nm showed a strong variability with values as low as –0.2 in the dust plume center. Below 1000-m height, the column-averaged Angström exponents strongly varied between 1.0 in the beginning of the dust period and 0.39 on 14 October 2001 when the dust penetrated into the boundary layer.

Comparison of the column-averaged optical depth and Angström exponents derived from lidar and Sun photometer observations showed their well agreement. Particle depolarization ratios of up to 25% were derived from lidar observations at 532 nm. Scattering phase functions retrieved from Sun photometer observations at λ = 440, 670, 870, and 1020 nm indicated particles of nonspherical shape. Just this shape caused unusually large particle extinction-to-backscatter (lidar) ratios at 532 nm in the range from 50 to 80 sr.

Using the scanning electron microscopy method, Li and co-workers[32] studied aerosol samples collected from Punta del Hidalgo in the Canary Islands and Sagres in Portugal during June–July 1997 in the framework of the Aerosol Characterization Experiment-2 (ACE-2). The analysis of the chemical composition of individual aerosol particles has shown that major aerosol types consisted of fresh and partly or completely reacted sea salts including NaCl, mixed-cation (Na, Mg, K, and Ca) sulfate, Na_2SO_4, and $NaNO_3$; particles of industrial origin that included $(NH_4)_2SO_4$, soot, ashes, silica, Fe oxide, and $CaSO_4$; and minor terrestrial mineral dust.

Because of their different geographic locations, samples from the two sites showed different degrees of contribution from European anthropogenic emissions. At Sagres, depending on the sample, between 0 and 30% of sea salt particles remained unreacted, while 0 to 50% were partly reacted, and 20 to 100% were completely converted to sulfate and nitrate through reactions with pollutants from the European continent. In contrast, at Punta del Hidalgo, the sea salt particles were much less affected by industrial pollution. Even during polluted periods, only less than 5% were completely reacted. The dramatic difference between aerosols from Punta del Hidalgo and Sagres indicates different processes of pollution dilution, reaction rates, and exhaustion of reactive pollutants during their transport over the open ocean.

Israelevich with co-authors[25] proposed to identify the sources of desert dust aerosols with local maxima of the TOMS aerosol index distribution averaged for the long period. Being simpler than the approach based on dusty days occurrence, this method gave the same results.

It was first shown[25] that in spring–summer, the flux of dust from the sources located at ~16°N and ~16°E, as well as around ~19°N and ~6°W exceeded the sinks due to settling and transport. As a result, the atmosphere over Northern Africa is almost permanently loaded with a significant amount of mineral desert dust in spring and summer periods.

It was also shown that the Chad basin source (16°N and 16°E) was most stable with maximal activity in April. The region around 19°N and 6°W turned to be a more variable source with a maximum in July. Low pressure systems, called Sharav cyclones, transport mineral dust eastward and northward along the Mediterranean basin. Observation of the dust plume propagation dynamics has shown that dust plumes appear first in the western sector of the Mediterranean Sea and then move eastward with a speed of about 7 to 8 degrees per day. In spring, this motion continues at least up to the eastern coast of the Mediterranean. In summer the dust plume is prevented from penetrating further east of about 15°E.

In June–July 1997, Hoornaert with co-authors[24] have studied the microstructure and chemical composition of aerosol at the island of Tenerife to analyze the dependence of the North Atlantic aerosol characteristics on the atmospheric circulation in both the marine boundary layer (MBL) and the free troposphere (FT). The data processing has shown that in the FT, the air masses can be classified as clean sea (Atlantic), containing Saharan dust, and polluted (European). In the MBL, air masses were classified as clean, polluted or perturbed by emissions from Europe, however, the Sahara DA contribution is insignificant.

For both the FT and MBL samples, the main changes in chemical composition were observed between fine and coarse aerosol modes. The FT fine mode aerosol is dominated by S-poor aluminosilicates (62%) in dust or sulphate samples; carbonaceous particles (20%) and S-rich aluminosilicates (46%) in the polluted samples. For larger fractions, a strong decreasing trend was observed for the sulphates (less than 20%) and carbonaceous particles (10%) in the polluted samples. The MBL fine mode was completely dominated by S-rich particles, and to a lesser extent, carbonaceous and sea salt particles. In the coarse mode, the polluted air mass is dominated by sea salt particles (62%) and contains only 5.3% of S-rich particles.

Vukmirović et al.[46] have analyzed the event of Saharan dust transport to Belgrade in the period 14–17 April 1994 through the numerical simulation of evolution of the aerosol chemical composition under the effect of dry and wet deposition.

2. Asian Pacific Regional Aerosol Characterization Experiments: ACE-Asia

Investigations of dust aerosol coming to the atmosphere in the deserts of China and then transported to a long range towards the East Chinese Sea and the Pacific Ocean have attracted a significant attention.[51]

Mass loading and chemical composition of aerosol particles were investigated over the China Dust Storm Research (ChinaDSR) observational network stations from March to May 2001 during the intensive field campaign period of ACE-Asia.[51] Four extensive dust storm (DS) events were observed in that period. Mass balance calculations showed that 45–82% of the observed aerosol mass was attributable to Asian soil dust particles, in which Ca and Fe contents were enriched to 12% and 6%, respectively, in the Western and Northern High-Dust source regions. For the latter areas, elemental contents exhibited high Si (30%) and low Fe (4%). For all major source areas, Al comprised 7% of Asian dust.

Analysis of air mass back-trajectories showed that five major transport pathways of Asian dust storms dominated in the dust transport in China during spring 2001, all of which passed over Beijing. Measurements also suggested that the sand land in Northeastern China was a potential source for Asian dust. The microstructure of aerosol with diameters between 0.25 and 16 μm was characterized by a lognormal distribution with a mass mean diameter of 4.5 μm and a standard deviation of 1.5. This range of soil dust constituted about 69% of the total dust loading. The fractions of particles in the size ranges <2.5 μm and >16 μm made around 1.7% and 30%, respectively.

Gong with colleagues[17] have simulated numerically the production and transport of Asian soil dust using the Northern Aerosol Regional Climate Model (NARCM), based on the DA mass budget equation determined by relations between the DA sources and sinks. The calculations referred to the period from March to May 2001 of the Aerosol Characterization Experiment-Asia (ACE-Asia), whose goals included the study of the DA dynamics in the Southeastern Asia. Rather complete meteorological information set *a priori* has allowed thorough reconstruction of processes of soil dust production, transport, growth, coagulation, and dry and wet deposition in China with allowance for 12 categories of soil aerosol sources. The size distribution of vertical dust flux was derived from the observed surface dust-size distribution in the desert regions.

Comparison between model simulations and ground-based and satellite measurements in East Asia and North America for the same period gave a good agreement. It was estimated that about 252.8 Mt of soil dust ($d < 40$ μm) was emitted in the East Asian deserts between 1 March and 31 May 2001, and about 60% was attributed to four major dust storms. The vertical dust loadings above 700 hPa correlate reasonably well with Total Ozone Mapping Spectrometer aerosol index (TOMS AI) observations. The sensitivity analysis of model performance to soil size distribution, water moisture, and meteorological conditions allowed one to find the most appropriate parameters and conditions being typical for the Chinese soil dust production and transport.

Mader with co-workers[33] studied the atmospheric aerosol in the Pacific region from the Twin Otter aircraft during the Aerosol Characterization Experiment (ACE)-Asia (March 31–May 1, 2001).

Particles were collected using a newly developed honeycomb denuder sampler and analyzed for concentration of organic carbon (OC), elemental carbon (EC), and carbonate (CC) carbon using a thermal–optical carbon analyzer. During some flights, increased concentrations of pollution and (or) mineral dust aerosol were observed.

Angström exponent (α) values, calculated based on data from the onboard three-wavelength nephelometer, were used to discern the nature of some individual layers. Values of α for individual layers ranged from 0.2 to 2, corresponding to dust- and pollution-dominated layers, respectively. OC and EC concentrations below 3 km ranged from 0.58 to 29 μg C/m^3 and from 0.20 to 1.8 μg C/m^3, respectively. In general, for a given type of atmospheric layer, higher levels of total carbon (TC) were observed during ACE-Asia comparative to observations during ACE-2, Tropospheric Aerosol Radiative Forcing Observational Experiment (TARFOX), and Indian Ocean Experiment (INDOEX).

Mixed layers of dust and pollution were found in some cases. A presence of CC was detected in samples taken from layers with $\alpha = 1.6$, indicating that significant amounts of dust can be present even though $\alpha > 0.2$. The linear regression of the light absorption coefficient σ_{ap} (Mm^{-1}), depending on EC concentration, was characterized by r^2 of only 0.50, indicating that parameters other than the mass of EC significantly affected the value of σ_{ap}. The mass absorption coefficient E_{abs} (m^2/g) varied by 8 times between sampling events at an average value of (11 ± 50) m^2/g, that agrees well with earlier data.

The key problems in interpretation of observations are associated with the need to answer the following questions: 1) whether the observed levels of OC and EC concentrations in the marine boundary layer influence the physical-chemical properties of cloud condensation nuclei (CCN) in MBL; 2) whether this influence affects the cloud formation and lifetime in MBL; 3) to what extent the OC chemical composition affects the CCN level and physical-chemical properties. In the last case, the important problem to be solved is to determine relative contributions of carbonaceous aerosol produced from burning of the biomass or fossil fuel.

The dust aerosol emitted into the atmosphere in spring in the desert regions of Northwestern Asia, and experiencing then the wind-driven long-range transport to the territory of the North Pacific attracts a significant attention in the context of its climatic effect and solving the problem of atmospheric correction. To solve the last problem, Li et al.[31] have analyzed the results of simultaneous observations of the outgoing SWR, based on the data of a spaceborne Sea-viewing Wide Field-of-view Sensor (SeaWiFS), and the sky irradiance measured *in situ* onboard the research vessel *Ronald H. Brown* in March–April 2001. The measurements were obtained within the framework of the ÀÑÅ-Asia Program. The sky irradiance data were used to retrieve aerosol optical thickness (AOT) using the optimization techniques. The measurements of outgoing SWR were reconstructed through iterative calculations with different aerosol characteristics specified.

The results obtained by Li et al.[31] agreed well with AOT data and direct measurements of water reflectance (albedo). According to observations at three stations, the error in the retrieved water reflectance was much decreased due to atmospheric correction, with average values of about 6% at 412 and 443 nm. The effect of the Asian dust was also simulated in comparison with that of fine absorptive aerosols. One of the observations showed that simultaneous accounting of dust and absorbing (soot) aerosols improved the accuracy of the AOT retrieving (from 44% to 13% at 865 nm). This underlines a necessity of accounting for the absorbing aerosol, when solving the problems of atmospheric correction for retrieving optical properties of sea water.

Increasing anthropogenic emissions from Asia, especially from regions undergoing rapid industrial development, have raised interest to chemically and radiatively important gases and aerosols. Earlier studies have shown that high concentrations of Asian pollution spread over a broad region of the North Pacific every spring. Prospero et al.[41] discussed the obtained data on aerosol concentrations at Midway Island (28°13′N, 177°22′W) in the center of North Pacific over the period 1981–2000. Using a relatively simple separation procedure, they estimated the natural and anthropogenic fractions of sulfate and nitrate aerosols.

They have shown that the estimated anthropogenic component almost doubled from 1981 to the mid-1990s, which closely correlates with the estimates of increased emissions of SO_2 from China. This conclusion is based on the assumption that the excess of SO_4^{2-} in spring with respect to the dimethyl sulfide (DMS) concentration estimated from the methyl sulfonate (ÌSÀ) data is determined by the contribution of anthropogenic emissions. This assumption is based on the consideration of episodic volcanic eruptions as a main natural source of SO_2. There are natural continental sources of NÎ$_õ$ emissions (for example, lightning discharges, soil emissions, biomass burning), but their contribution (as compared to anthropogenic sources) can be considered as small, especially in spring (this is true for biomass burning). The contribution from non-Asian sources of these MGCs, whose influence is marked mostly in winter, is likely low as well.

Measurements in the late 1990s suggest that sulfate SO_4^{2-} and nitrate NO_3^- concentrations have stabilized and perhaps decreased (starting from the mid-1990s). However, these data are less credible, because of the lower (by various organizational reasons) observation quality, because many aerosol samples were rejected. Nevertheless, the conclusion about the trend to SO_4^{2-} stabilization or decrease can be considered reliable, because in this period the scales of using fossil fuels in China stabilized and began to decrease. In the late 1990s, the emissions of active nitrogen compounds decreased significantly as well. The experimentally observed stabilization or decrease of the SO_4^{2-} and NO_3^- concentrations mean that the earlier predictions overestimate the concentration of these components in the troposphere, especially, because the available data suggest possible decrease of NO_x emissions in China. From the viewpoint of refining the information about the aerosol radiative forcing, of great importance is the consideration of the carbonaceous aerosol, whose content is rather high in the emissions coming from China to the Atlantic Ocean.

Since biomass is commonly used as a fuel source for cooking and heating homes throughout South Asia, the smoke from domestic use of these fuels is expected to be a major source of the atmospheric particulate matter in the region. To study such aerosol products, Sheesley et al.[44] carried out laboratory experiments on combustion of various bio fuels used in Bangladesh. The chemical composition of burned samples including total organic and elemental carbon, sulfate, nitrate, ammonium and chloride ions, and elements such as potassium and sodium did not show conclusive differences among the biomass samples tested. Unique features, however, exist in the detailed organic characterization of the combustion smoke from different sources. The organic compound fingerprints of the particulate matter were shown to be distinct from North American wood fuels. Fecal stanols including 5β-stigmastanol, coprostanol, and cholestanol were found to be good molecular markers for the combustion of cowdung. Additionally, the patterns of methoxyphenols and plant sterols provide a unique information on biomass.

The drying up of the Aral Sea with following desert expansion belongs to the most staggering environmental disasters of the twentieth century. Over the last 40 years over 36 000 km^2 of the former seabed have been exposed, creating a potentially significant aeolian dust source. It is widely believed, that increased dust storm activity in the region has a major impact on human health. In this context, Wiggs with co-workers[50] studied a link between dust exposure and respiratory health amongst children in the Autonomous Republic of Karakalpakstan. The findings indicated that the northerly regions of the former bed of the Aral Sea are the most intensive sources of dust for the atmosphere. In these regions it appears that local sources and more distant sources to the south and south-west represent significant sediment providers, particularly in the early summer. Provisional analysis of the respiratory health data suggests that children living in the north of the country, where aeolian dust deposition rates are greater, show a lower frequency of respiratory problems. This inverse correlation requires further investigation, but underlines the complexities of environmental and human health inter-relationships.

Certainly, dust aerosol is very important, but only one of the components of atmospheric aerosol in Asia. Volcanic eruptions also contribute significantly, but episodically to the atmospheric aerosol.

The Japanese Archipelago is one of the most volcanically active regions in the world. It incorporates 83 active volcanoes, including 12 especially high-active ones, such as Sakurajima, Asosan, Unzendake, and Miyakejima. The analysis of observations performed by Fujita et al.[15] suggested that volcanic emissions of sulfur dioxide make up about 20% of total SÎ_2 emissions and contribute about 20% to the total (dry and wet) sulfur deposition under the conditions of Japan. In this context, the eruption (in July 2000) of the Miyakejima stratovolcano located 200 km south of Tokyo (34.68°N; 139.53°E) was unprecedented in the scales and, in particular, affected the air quality in the Tokyo megapolis, whose population is about 30 million.

The long activity of the volcano caused a significant increase of the SO_2 concentration in the atmosphere. The daily average emission of SO_2 during the period from August 2000 to March 2001 achieved $15 \cdot 10^9$ gS/day, that is, it was the same level as the recent emission of SO_2 originating from fossil fuel combustion in East Asia. The estimated total (dry and wet) deposition of sulfur after the eruption amounted to about 4.26 mgS/(m$^2 \cdot$ day), which was more than three times higher than the pre-eruption level [1.41 mgS/(m$^2 \cdot$ day)]. The contribution of wet deposition increased with increasing daily precipitation amount, which indicates that the precipitation had a significant effect in the removal of the volcanic sulfur from the atmosphere.

In the warm season, when the subtropical high-pressure system over the Pacific is dominant, SO_2 high-concentration episodes took place sometimes with a time lag due to north transport of volcanic plumes. In the cold season, however, the northwesterly wind from the Asian continent is predominant in this region, therefore, most of the volcanic sulfur is transported over the Pacific Ocean. Although no direct correlation was found between the SO_2 dry deposition and the volcanic SO_2 emission, there was a close parallel in temporal variation between the nss-SO_4^{2-} wet deposition and the SO_2 emission. It is believed that the interaction between the precipitation cloud and the volcanic cloud took place just after the eruption of Miyakejima and that wet deposition played an important role in the removal of the volcanic sulfur inflow outside the region.

Gao et al.[16] analyzed the chemical composition of 41 aerosol filter samples obtained by airborne sampling over the ocean to the north, south, and west of Japan. These samples were analyzed for their water-soluble chemical components, particularly, organic species. Inorganic anions, carbohydrates, organic acids, and metallic elements were identified and quantified (21 distinct species). The chemical composition of the aerosol suggested that the aerosol could be largely attributed to three factors: anthropogenic emissions, biomass burning, and soil dust.

As was already mentioned, the dust aerosol, emitted into the atmosphere during dust storms in the Sahel region of Northern Africa and subject to the long-range transport, often reaches Western Europe and even Southern Great Britain. For example, the traces of such aerosol deposited onto the snow cover surface were found in the Alps. Potential sources of different Saharan dust events can be identified by combining reconstruction of air mass back-trajectories from dust deposition sites in Europe and measurements of the Nd isotopic composition of the deposited dust particles. Thus, for example, the study of “red dust” events collected in France allowed identification of a distinct North African source area.

Unexpectedly, the back-trajectory analysis performed by Grousset with co-workers[19] showed that the air mass trajectory of one dust event (March 6, 1990) was of Chinese origin. The Nd isotopic composition of the dust was consistent with the range of isotopic compositions of Chinese loess. Moreover, the atmospheric global model simulation revealed that a dust plume left China before February 25, 1990, crossed North America around the February–March transition and reached the French Alps by March 6, 1990, revealing that intercontinental west–east transport of dust and pollution may occur across the Pacific Ocean and the North Atlantic.

Within the framework of the ÀÑÅ-Asia program aimed at investigation of the atmospheric aerosol in East Asia, Wang with co-workers[47] measured the aerosol size distributions and chemical compositions, using differential mobility analyzers (DMA), an aerodynamic particle sizer (APS), Micro-Orifice Uniform Deposit Impactors (MOUDI), and denuder samplers onboard the Twin Otter aircraft. Measurements of four flights were analyzed in detail, and aerosol extinctions predicted using *in situ* aerosol size distribution and chemical composition measurements were compared to those derived from the 14-wavelength NASA Ames Airborne Tracking Sun photometer (AATS-14) under conditions of the cloudless atmosphere.

The spatial distribution of aerosol was characterized by the presence of pollution in the atmospheric boundary layer (ABL) and the virtual absence of mineral dust in the free troposphere. For all the four flights, the best fit lines yielded predicted/observed extinction ratios in boundary and pollution layers of 0.97 ± 0.24 and 1.07 ± 0.08 at $\lambda = 525$ nm and 0.96 ± 0.21 and 1.08 ± 0.08 at $\lambda = 1059$ nm, respectively. In dust layers located in the free troposphere, aerosol extinctions predicted from the measured size distributions were generally smaller than those derived from the AATS-14 data, with predicted/observed ratios of 0.65 ± 0.06 and 0.66 ± 0.05 at 525 and 1059 nm, respectively. The discrepancy was likely a result of the lack of the knowledge of mineral dust shape in the modeling, as well as variations in aerosol extinction derived from AATS-14 data when viewing through horizontally inhomogeneous layers.

3. Indian Ocean Experiment INDOEX

The Indian Ocean Experiment (INDOEX) measurements on land, sea, and in air on 11–25 of February, 1999, were designed to obtain information on chemical, physical, and optical properties of the atmospheric aerosol with the use of ground-based, aircraft, and ship-borne observation technique.[12,13,38,49] In order to best interpret the properties of the INDOEX aerosol, comparisons of data by platform, air mass origin, and light scattering intensity were undertaken. They revealed a significant variability in platform averages of aerosol extensive properties (e.g., mass, light scattering, and absorption) but less variability in intensive properties (e.g., mass scattering efficiency, single scattering albedo, backscatter fraction, and Angström exponent) and the ratios of constituents. In general, ratios of chemical species were found to show greater variability than aerosol microstructure parameters or optical properties.

The variability of the aerosol characteristics revealed from the comparison of data of instruments installed on different carriers can be interpreted both as really existing and as caused by specific features of measurement techniques and nonidentical observational conditions. At higher haze concentrations with higher scattering values, various determinations of the mass scattering efficiency (MSE) at 33% relative humidity converged on values of about $3.8 \pm 0.3\ \mathrm{m^2 \cdot g^{-1}}$, providing a firm constraint upon the description and modeling of haze optical properties. MSE values became more variable in clean air or regions of low aerosol concentrations. The results obtained by Clarke et al.[12] suggest that “closure” between chemical, microphysical, and optical properties across platforms to better than about 20% will require significant improvements in techniques, calibration procedures, and comparison efforts.

The important objective of INDOEX was to study the evolution and transport of aerosols and trace components emitted in the atmosphere in the region of the Indian subcontinent, and aerosol interactions with clouds, radiation, and climate. The observational facilities involved in the INDOEX included a system of balloons drifting horizontally at different altitudes. During the INDOEX Intensive Field Phase (January 15 – February 27), 17 superpressure balloons drifting at lower tropospheric levels were launched from Goa.

Satellite tracking of the balloon drift showed that the drift was consistent with the Madden–Julian oscillation, but balloons also showed evidence of coastal anticyclonic circulations off the western coast of India, linked to both the orography and mesoscale circulation systems. The latter may be important in the far transport of anthropogenic or mineral pollutants across the coast.

Comparison of the observed and pre-calculated data was generally good in terms of wind velocities, temperature, and humidity. However, above the marine boundary layer, some discrepancy was revealed, including a cold bias of analyzed temperatures, which can be partly explained by spatial smoothing in calculations.

As a part of the INDOEX 1999 Intensive Field Phase, measurements of aerosol properties were made aboard the R/V *Ronald H. Brown* in the Indian Ocean north and south of the Intertropical Convergence Zone (ITCZ) in the Arabian Sea and in the Bay of Bengal. On the basis of air mass trajectories, Quinn with co-authors[42] identified eight air mass source regions including the southern hemisphere Atlantic; southern hemisphere Indian Ocean; northern hemisphere Indian Ocean; east Indian subcontinent, where trajectories came from near Calcutta through the southern portion of India; Indian subcontinent where trajectories came from across central India; Arabia; Arabia/Indian subcontinent, a mixed region where lower-level trajectories came from Arabia and upper-level trajectories came from India; and Arabian Sea / coastal India where trajectories came from along the coast of India.

Properties of the aerosol, measured in the marine boundary layer, included chemical composition, number size distribution, and scattering and absorption coefficients. In addition, vertical profiles of aerosol backscatter and optical depth were measured. Quinn with co-authors[42] analyzed the concentrations and mass fractions of the dominant aerosol chemical components, extinction, aerosol scattering and absorption coefficients, single scattering albedo, Angström exponent, and optical depth at the 500 nm wavelength as a function of air mass source region. All results except the aerosol optical depth were obtained at relative humidity of 55 ± 3%. For air masses that originated from the southern hemisphere, sea salt dominated the extinction by sub-1 μm and sub-10 μm aerosol particles. The ratios of the above extinctions were the lowest measured of all air mass source regions (mean values of 28 and 40%) due to the dominance of the aerosol mass by supermicron sea salt. In addition, aerosol optical depths were the lowest measured averaging 0.06 ± 0.03.

Non-sea-salt (nss) sulfate aerosol concentrations in air masses from the northern hemisphere (Indian Ocean region) were a factor of 6 higher than those in southern hemisphere air masses, while submicron sea-salt concentrations were comparable. Sulfate aerosol made up 40% of the sub-1 μm extinction, while sea salt dominated the sub-10 μm extinction. Aerosol optical depths for this source region averaged 0.10 ± 0.03. Mean single scattering albedo near 0.89 and detectable black carbon (BC) concentrations (0.14 ± 0.05 μg / m^3) indicated the transport of continentally derived aerosol to the ITCZ. The two regions influenced by low-level (500 m) airflow from Arabia had higher concentrations of submicron nss sulfate, particulate organic matter (POM), and inorganic oxidized material (IOM) than were observed in the marine regions. Concentrations of supermicron IOM were comparable to supermicron sea-salt concentrations. Nss sulfate aerosol dominated the sub-1 μm extinction and made significant contributions to the sub-10 μm extinction, when the salt marine aerosol dominated.

Mean BC contributions to submicron extinction were 8 and 12% for marine and continental regions, respectively. Single scattering albedo values were 0.93 ± 0.02 and 0.89 ± 0.02 for these two source regions. Aerosol optical depths averaged 0.19 ± 0.12 and 0.38 ± 0.07 with the higher value due to upper-level (2500 m) flow from India. Regions influenced by low-level airflow from the Indian subcontinent had the highest submicron nss sulfate, POM, BC, and IOM concentrations. Supermicron sea-salt concentrations were lower than or comparable to supermicron nitrate concentrations. Sub-1 μm and sub-10 μm extinction were dominated by nss sulfate aerosol although a burning component consisting of BC, HNO_3, and H_2SO_4 made a nearly equivalent contribution. These regions had a mean single scattering albedo of 0.85 ± 0.11, the lowest measured for any region. Mean aerosol optical depths were highest (0.3 to 0.4) for regions of far transport of airflows from the Indian subcontinent.

Since the carbonaceous aerosol is one of the main fractions of the atmospheric aerosol, of great interest is information about the content of elemental carbon, responsible for absorptive properties of aerosol, and organic carbon, which contributes significantly to the aerosol scattering of radiation. This is especially true for almost lacking data about the OC / EC ratio for different particle size fractions. If EC originates mainly from biomass burning, then the sources of OC are more diverse, including direct atmospheric emissions upon incomplete burning of fossil fuels or biomass, gas-phase reactions with participation of volatile organic compounds, and others. Neusüß with co-authors[39] discussed the concentrations and mass fractions of organic carbon, elemental carbon, and numerous organic species measured for submicrometer and supermicrometer particles in the Indian Ocean on the cruise of the Research vessel *Ronald H. Brown* during INDOEX in February – March 1999.

The total carbon (TC) concentration was low for air masses originating from the southern hemisphere of the Indian Ocean, where only small amounts of organic carbon (OC) could be detected in supermicrometer (sea salt) particles. The OC concentration was typically more than one order of magnitude higher in the northern Indian Ocean, where continental air masses were encountered, both for submicrometer and supermicrometer particles. Elemental carbon (EC) was mostly found in submicrometer particles, increasing up to more than two orders of magnitude in the northern part compared to the southern part of the Indian Ocean. The mass fraction of carbonaceous material was in the ranges 6 – 15% and 2 – 12% for submicrometer and supermicrometer particles, respectively, with the higher values for more polluted air masses.

The measured high absorption coefficients were partly attributed to a high degree of mixing with light scattering material. The absorption efficiency turned out to be higher for lower mass fractions of EC. Various short-chain dicarboxylic and hydroxylated dicarboxylic acids have been determined as absorbing components. Dicarboxylic acids were mostly found in supermicrometer particles, whereas hydroxylated dicarboxylic acids showed up mainly in submicrometer particles. Alkanes and polycyclic aromatic hydrocarbons have been found in small amounts, leading to the conclusion that an important part of the organic material is of secondary origin. This was confirmed by good correlation of the sum concentration of carboxylic acids with OC concentration, nitrate, and a less pronounced correlation with sulfate. However, low OC / EC ratios indicated the existence of primary OC. Observations of OC and EC sources in Southeastern Asia seem to be extremely important.

During the Indian Ocean Experiment, low-level flow from the continent transported pollutants over the Indian Ocean toward the Intertropical Convergence Zone (ITCZ) took place. The back-trajectory analysis of air masses revealed the transformation of the chemical composition of aerosol and trace gases depending on their geographic origin. In this connection, Guazzîtti et al.[20] compared temporal evolutions of acetonitrile, a long-lived specific tracer for biomass/biofuel burning, number concentration of submicrometer carbon-containing particles with potassium (indicative of combustion sources), and mass concentration of submicrometer non-sea-salt (nss) potassium. High correlation coefficients ($0.84 < r^2 < 0.92$) were determined for these comparisons indicating that most likely the majority of the species evolve from the same, related, or proximate sources.

Aerosol and trace gas measurements provide evidence that emissions from fossil fuel and biomass /biofuel burning are subject to long-range transport. High concentrations of submicrometer nss potassium, carbon-containing particles with potassium, and acetonitrile are observed in air masses advected from the Indian subcontinent, indicating a strong impact of biomass/biofuel burning in India during the INDOEX period ((74 ± 9)% contribution to submicrometer carbonaceous aerosol). In contrast, lower values for these species were measured in air masses from the Arabian Peninsula, where dominance of fossil fuel combustion made (63 ± 9)% contribution to submicrometer carbonaceous aerosol. The results obtained demonstrate the importance of complex study of regularities of variations of aerosol chemical composition and trace gas components in different regions of the world.

Micropulse lidar (MPL) systems were used to measure aerosol properties during the INDOEX 1999 field phase. Measurements were made from two platforms: the NOAA ship R/V *Ronald H. Brown*, and the Kaashidhoo Climate Observatory (KCO) in the Maldives. Multichannel sun photometers (wavelengths of 380, 440, 500, 675, and 870 nm) were used in measurements of aerosol optical depths (AOD) needed to calibrate the MPL. Welton et al.[49] discussed the results of observation of the vertical profiles of aerosol optical properties at 523 nm. The height of the highest aerosols (top height) was below 4 km for most of the cruise. At that time, the marine boundary layer (MBL) top was less than 1 km.

The lidar sensing results were combined with computed air mass trajectories and radiosonde measurements. Humidity varied from approximately 80% near the surface to 50% near the top height during the entire cruise. Marine aerosols in the absence of any continental influence were found to have an AOD of 0.05 ± 0.03, an extinction-to-backscatter ratio (*S* ratio) of 33 ± 6 sr, and peak extinction values around 0.05 km^{-1} (near the MBL top). The marine results are in agreement with previously measured values. Polluted marine areas over the Indian Ocean, influenced by continental aerosols, had AOD values in excess of 0.2, *S* ratios well above 40 sr, and peak extinction values approximately 0.20 km^{-1} (near the MBL top). All these results are typical for continental aerosols.

Welton et al.[49] compared the MPL derived extinction near the ship (a height of 75 m) and extinction calculated at the ship level using data on the scattering measured with a nephelometer and absorption measured with a particle soot absorption photometer. The comparisons indicated that the MPL processing algorithm (using a constant *S* ratio throughout the lower troposphere) calculates extinction near the surface in agreement with the ship-level measurements only when the MBL aerosols are well mixed with aerosols above. Analysis of the MPL extinction profiles showed that the model of aerosol vertical extinction developed during an earlier INDOEX field campaign (at the Maldives in 1996) did not correctly describe the true vertical distribution over the Indian Ocean region. A new model of aerosol vertical extinction was determined for marine atmospheres over the Indian Ocean, as well as a new model for polluted marine atmosphere influenced by continental aerosols.

Mayol-Bracero with co-workers[35,36] measured carbonaceous material and water-soluble ionic species in the fine fraction ($D_p < 1.3\ \mu$m) of aerosol samples collected on NCAR's C-130 aircraft during the intensive field phase (February–March 1999) of the Indian Ocean Experiment in order to analyze the chemical composition of aerosol as a function of the altitude. Polluted layers were present north of the equator at altitudes up to 3.2 km. The estimated mass concentration (sum of carbonaceous and soluble ionic aerosol components) of fine-mode particles in these layers was 15.3 ± 7.9 $\mu\text{g/m}^3$. The major components were particulate organic matter (POM, 35%), SO_4^{2-} (34%), black carbon (BC, 14%), and NH_4^+ (11%).

The main difference between the composition of the marine boundary layer (0 to 1.2 km), and the overlying residual continental boundary layer (1.2 to 3.2 km) was a higher abundance of SO_4^{2-} relative to POM within the MBL, probably due to a faster conversion of SO_2 into SO_4^{2-} in the MBL. The results obtained have shown that carbon is a major, and sometimes dominant contributor to the aerosol mass and that its contribution increases with altitude. Low variability was observed in the optical properties of the aerosol in two layers. Regression analysis of the absorption coefficient at 565 nm on BC mass ($\text{BC} < 4.0\ \mu\text{gC/m}^3$) yielded a specific absorption cross section of 8.1 ± 0.7 m^2/g for the whole observation period. The unusually high fraction of BC and the good correlation between the absorption coefficient and BC suggest that BC was responsible for the strong light absorption observed for the polluted layers during INDOEX. High correlation between BC and total carbon (TC) ($r^2 = 0.86$) suggest that TC is predominantly of primary origin. Good correlations were also found between the scattering coefficient at 550 nm and the estimated aerosol mass for the fine fraction. A specific scattering cross section made 4.9 ± 0.4 m^2/g. The observed BC/TC, BC/OC, SO_4^{2-}/BC, and K^+/BC ratios were fairly constant throughout the period. These ratios suggest that between 60 and 80% of the aerosol in the polluted layers during INDOEX originated from fossil fuel combustion and between 20 and 40% from biofuel combustion.

The aerosol extinction properties in the framework of INDOEX were measured over the Goa area (western coast of India, 15.45°N, 73.08°E) between 1 and 15 March 1999.[11] The vertical profiles of the backscatter-to-extinction ratio (Ô$_\text{ð}$) were retrieved in order to study the temporal evolution and the vertical distribution of the aerosol trapped inside the winter monsoon plume (micropulse lidar emitting at 523 nm). Sun photometer-retrieved optical thickness at 523 nm was used to constrain the inversion in daytime and led to Ô$_\text{ð}$ = (0.03 ± ± 0.010) sr^{-1}. Black carbon concentration (BC) was shown to be a representative tracer of the surface scattering coefficient of aerosols (α_s); relative humidity (*RH*) effect was of secondary importance.

A statistical linear relationship between BC and α_s has been found in Ref. 11 and then used to constrain the lidar inversion during nighttime, when values of Ô$_\text{ð}$ are practically the same as in daytime. The lidar-derived optical thickness was shown to be higher during nighttime over the entire measurement period (mean value of 0.76 ± 0.15 for nighttime instead of 0.55 ± 0.09 for daytime). A significant contribution to the optical thickness was due to the aerosol layer within 0.7 km above sea level (asl). The second aerosol layer was found between 0.7 and 3.5 km asl with an important diurnal variability both in altitude extension and optical thickness. Such variability seems to be due to the sea breeze cycle. In the upper aerosol layer, optical thickness varied from 0.49 ± 0.14 during nighttime to 0.25 ± 0.07 during daytime.

Masonis with co-workers[34] compared aircraft *in situ* and Raman lidar profiles of aerosol light extinction (σ_{ep}) and 180° backscattering (β_p) for 6 days during the INDOEX campaign at Maldives (4.18°N, 73.53°E). The measurements of σ_{ep} and β_p were made onboard the C-130 aircraft using two integrating nephelometers and one photometer to measure light absorption by soot particles. Aerosol optical depth was also measured using a sun photometer.

On average, the lidar-derived values of σ_{ep} and β_p were ~30% larger than the *in situ*-derived values at a 95% confidence interval. Possible reasons for the discrepancy are (1) a low bias in the *in situ* measurements because of losses in the aircraft inlet; (2) underestimation of the humidification effect on light extinction in the *in situ* measurements; (3) overestimation of σ_{ep} and β_p in the lidar measurements because of subvisible cloud contamination; (4) errors in data processing (especially, lidar data). Spatial-temporal variability also appears to be a source of at least some of the discrepancy in the *in situ* and lidar data.

Rainwater samples for chemical analysis were collected over the Indian Ocean during the Indian Ocean Experiment (INDOEX) campaign in January–March 1999 on board the research vessels *Ronald H. Brown* and *Sagar Kanya*.[18] Samples were analyzed for major ions and some trace metals. The rainwater data were interpreted in terms of transport from potential source regions in Asia using air mass trajectories covering 10 days. A comparison was also made between the rainwater data and the concentration of aerosol components measured simultaneously onboard the ships.

Though the concentrations of nonsea-salt (nss)-SO_4^{2-}, NO_3^-, NH_4^+, nss-K^+, and nss-Ca^{2+} in rainwater over the Indian Ocean were a factor of 2 to 3 lower than over the Indian continent, they were still clearly influenced by pollution and soil sources in Asia. The concentration of nss-Ca^{2+} decreased more rapidly as the air moved southward from the continent over the ocean, whereas the concentration of nss-SO_4^{2-} became relatively more abundant. This was consistent with the observed higher acidity of the rainwater over the ocean (pH in the range 4.8 to 5.4) than over the Indian subcontinent, with NH_4^+ as the main cation (rather than Ca^{2+}, as over land). Variations in the concentration of Al and Fe correlated well with those of nss-Ca^{2+}, indicating a continental (rock) source for these elements.

The relation between Na^+, Cl^-, and Br^- in the rainwater was close to that of seawater, implying no excess or deficit of the two halogen ions. The ratio between the concentrations of ions of metals in rainwater and in surface air was systematically larger for aerosol components that exist in the coarse mode of sea-salt origin (Na^+, Mg^{2+}, and Cl^-) comparative to the fine mode (NH_4^+, nss-K^+, and nss-SO_4^{2-}). This indicates that fine-mode particles are scavenged mainly by in-cloud processes whereas coarse-mode sea-salt particles are scavenged also by falling raindrops under the clouds. Nss-Ca^{2+} and NO_3^- fell in a category in between, indicating that these compounds are not as effectively removed by below-cloud scavenging as sea-salt aerosols.

The Asian–African winter monsoon is characterized by the stable northwestern air mass transport from the Indian subcontinent to the Arabian Sea and equatorial belt of the Indian Ocean. This situation considerably affects the development of chemical processes on the global scale due to the transport of large amounts of the anthropogenic atmospheric aerosol. To analyze the regularities of the air mass transport, Krichak et al.[29] computed forward trajectories of the air masses from the Arabian Sea area for each day from February 1 to April 10, 1999, during the Indian Ocean Experiment (INDOEX). Numerical simulation of one of the transport episodes in the presence of intense cyclone and dust plume development allowed the evolution of dust plume formation to be followed. Potential role of the weather-forming systems in the transport to Europe of the polluted air masses from the INDOEX area was discussed in Ref. 29. This transport can significantly affect the precipitations and climate in Southern Europe and the Mediterranean region.

In Part 2 of this review, we will discuss the results of most important combined field observational experiments in America and Western Europe.

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