Atmospheric aerosol as a climate-forming component of the atmosphere. Part 2. Remote sensing of the global spatiotemporal variability of aerosol and its climate impact

K.Ya. Kondratyev

Center for Ecological Safety RAS / Nansen Environmental and Remote Sensing Center, St. Petersburg

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A review is presented of recently developed techniques to remotely sounding aerosol as well as of some results of the sensing are considered with a particular attention paid to the data on aerosol optical thickness. The most recent assessments of aerosol radiative forcing are being discussed in the context of "aerosol and climate" problem.

Introduction

Recent studies connected with numerical simulation of the global climate change has led to understanding of the great importance of atmospheric aerosol as a climate-forming component of the atmosphere.[2–8,8a,b,13,20–23] The paucity of observations of various aerosol characteristics stimulated development of methods for remote sensing of aerosol (especially, lidar and satellite ones) as the most promising way of obtaining adequate information on the spatiotemporal variability of the aerosol content and properties on the global scale. The first results obtained in this field allowed more reliable estimation of the aerosol radiative forcing. In addition to recent reviews,[6,7] this paper discusses the results of most recent investigations.

1. Remote sensing of aerosol

The multiwavelength MODerate resolution Imaging Spectroradiometer (MODIS) on the Terra satellite has opened new wide possibilities of obtaining information about aerosol properties on the global scale. The complexity of the problem on retrieving the aerosol characteristics from the MODIS data (especially, in the atmosphere over land) requires serious efforts on estimating the retrieval errors caused by various factors. For this purpose, ground-support observations of such characteristics as the aerosol optical depth (AOD), aerosol microstructure, and others are being conducted at more than 100 sites of the AERONET network of automated observations located in different regions of the globe. Ichoku et al.,[17] compared the synchronous MODIS and AERONET data, and their comparison showed that the mean values of the aerosol characteristics are in quite a good agreement, although the data on their variance reflect the interfering effect of land inhomogeneities, especially, at low aerosol content in the atmosphere.

Chu et al.,[12] have processed MODIS data and retrieved, for the first time, the aerosol optical depth (τ_a) of the atmosphere over vegetated and partially vegetated land from measurements of the outgoing short-wave radiation at the wavelengths of 470 and 660 nm. The extensive validation made during July–September, 2000 encompassed 315 co-located τ_a in space and time derived by MODIS and AERONET (Aerosol Robotic Network) from more than 30 AERONET sites in Europe, Africa, North and South America. However, the lack of AERONET measurements in East Asia, India, and Australia makes this validation not globally representative.

In summary, the MODIS aerosol retrievals, except for those in coastal zones, were found within the retrieval errors $\Delta\tau_a = \pm 0.05$ or $\pm 0.2\tau_a$. The root mean square (RMS) errors are from ≤ 0.1 in the continental inland regions and up to 0.3 in the coastal regions (attributed mainly to water contaminated signal). The results obtained indicate that the MODIS AOD data can be used for solution of various applied problems, but with caution for possible presence of residual clouds, snow/ice, and water contamination. In this context, the planned analysis of data for the Asian region with its characteristic dust storms becomes of great importance.

The MODIS spectroradiometer aboard NASA's Terra spacecraft began collecting data in February 2000. Remer et al.,[34] described the results of testing the algorithm for determining aerosol characteristics over ocean from the spectral radiation measured using six channels of the MODIS in the spectral range from 550 to 2100 nm, at a spatial resolution of 500 km. The parameters retrieved with the spatial resolution of 100 km are the spectral aerosol optical depth, effective particle radius, and the AOD fraction due to fine (submicron) aerosol. The adequacy of the algorithm was validated through comparison with the co-located observations from the AErosol RObotic NETwork (AERONET) ground-based sunphotometer network. MODIS-retrieved

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AOD at 660 and 870 nm fall within the expected uncertainty interval, with the ensemble mean at 660 nm different from the AERONET observations by only 2% and having virtually no offset. Roughly 70% of MODIS retrievals of the aerosol effective radius for the optical thickness larger than 0.15 agree with the AERONET retrievals to within ±0.10 μm.

The problem of cloud screening is a major challenge for the remote sensing of aerosols from space. To solve this problem, Martins[28] has proposed a new cloud masking algorithm based on the spatial variability of reflectance at the top of the atmosphere in the visible range (the spatial pattern of the cloud reflectance as observed from space is usually strongly different from that of aerosols) for the retrieval of aerosol properties by MODIS. Clouds show a very high spatial variability on the scales of hundreds meters to few kilometers, whereas aerosols in general are very homogeneous. This masking algorithm, however, is applicable only to a sufficiently homogeneous surface (largely over ocean).

Martins et al.,[28] have also performed detailed study on the effect of cloud contamination on the aerosol retrievals with a justified selection of the cloud mask parameters. Application of this new technique for separation of the contributions from clouds and aerosol to formation of the field of outgoing short-wave radiation turned out quite successful for almost all types of aerosol and clouds.

Because of a wide spatiotemporal variability of the concentration and properties and a relatively short lifetime of the atmospheric aerosol, its regional impacts on the radiative balance and climate differ significantly from the globally mean estimates. It is known, for example, that in the regions with high aerosol content its direct forcing manifesting itself as cooling may exceed the effect of warming due to greenhouse gases. In this context, Menzies and co-authors[32] discussed the results of studying the scattering properties of the tropospheric aerosol using airborne lidar data at 532, 1064, 1540, and 9250 nm wavelengths obtained during the Global Backscatter Experiment (GLOBE) airborne campaigns covering a wide range of latitude and longitude over the Pacific Ocean. A particular attention was paid to the vertical profiles of aerosol properties in the marine boundary layer (MBL), as well as elevated dust layers due to transport of material from Asia.

The lidar data were used, in particular, to determine the aerosol microstructure and the dust and sea salt aerosol refractive index. The published observation data on aerosol characteristics from onboard *in situ* sensors were used to verify locations where mineral dust aerosol dominated the aerosol optical properties in the free troposphere. In such cases, the aerosol had a unimodal lognormal size distribution with the median radius for the volume microstructure

$$r_{\text{vol}} = r_{\text{g}} \exp\,(3(\log s)^2)$$

(r_{g} is the median radius) ranging from 0.5 to 0.7 μm and $s = 1.6$–1.7. These parameters agree with the data for the dust aerosol transported from Asia in the middle and upper troposphere. The complex refractive index of aerosol particles at the 9250 nm wavelength strongly depends on the aerosol mineral composition and varies quite widely.

The tropical Indian Ocean undergoes the effect of two air masses (marine and continental) with the significantly different properties connected with the Indian monsoon system. The summer monsoon, usually called southwest, begins usually in late May and lasts till November. During this period southwestern or western winds dominate. The winter (northeastern) monsoon is formed by late November and lasts till April with the dominant northeast wind. Li and Ramanathan[26c] have analyzed the specific variation of atmospheric aerosol attributed to these two contrast air masses using the satellite-derived aerosol optical depth data.

Earlier similar studies showed that the northern Indian Ocean is characterized by a major transition from anthropogenic aerosols during the northeasterly winter monsoon season to mineral dust and sea salt during the southwest summer monsoon. The former is dominated by low-level transport from south and southeast Asia, while the latter results from the low to mid-tropospheric transport from the African continent and the Arabian Peninsula.

According to the results obtained, the monthly mean AOD over the Arabian Sea displays a clear annual cycle with a maximum (0.60 ± 0.10) in July and a minimum (0.20 ± 0.05) in January. The summer monsoon maximum is due to both the southwest winds bringing dust from the Horn of Africa over the Arabian Sea and mid-tropospheric transport of dust from the Arabian Peninsula. Such long-range transports also lead to aerosol loading south of the equator. The Indonesia forest fires during the 1997 El Niño also led to a large increase in AOD over most of the equatorial Indian Ocean.

Ground-based measurements of AOD have been carried out at the Koldewey station in Ny-Elesund, Spitzbergen (Norway, 78.95°N, 11.93°E) since 1991. The basic instrumentation was a 17-channel solar photometer operated in the wavelength region from 361 to 1062 nm. To extend observations to the polar night period, these measurements were then complemented with observations using a Sun and Moon photometer (1995) and a star photometer (1996). The year-round measurements made it possible to study in detail the interannual and seasonal variations of total AOD in the Arctic. As additional information, the stratospheric AOD measured with a Stratospheric Aerosol and Gas Experiment (SAGE II) was used.

The processing of the experimental findings made by Herber et al.,[16] showed that the lowest tropospheric AOD values occur in late summer and fall. Each year, strong Arctic haze events were recorded not only during spring (when the haze frequency of occurrence is 40%) but also in late winter (frequency of occurrence of 17%) as was shown by the first star photometer measurements.

Five-day backward trajectories were used to analyze possible sources for high tropospheric AOD. Elevated tropospheric AOD appeared for northeasterly, easterly, or westerly winds. Analysis of the long-term changes of tropospheric AOD revealed a small positive trend in AOD near Spitzbergen during the nine-year measurement period.

During the Second Aerosol Characterization Experiment (ACE 2) in summer of 1997 Ansmann et al.,[8b] observed several European aerosol pollution outbreaks based on multiwavelength backscatter and 532-nm extinction profiling with a lidar at Sagres (37°N, 9°W), southern Portugal, and optical depth observations with a star photometer at the lidar site and a Sun photometer atop a nearby mountain.

The observations are mainly presented in terms of profiles of the 532-nm extinction-to-backscatter ratio (lidar ratio) and of Angström exponents calculated for the wavelength ranges 400–532 nm and 532–800 nm. The lidar ratio indicates the aerosol type (marine, soil, pollution) whereas the Angström exponents are sensitive to changes in the particle size distribution (accumulation mode, coarse mode).

Reference 8b discusses the results of an extensive correlation analysis considering all determined optical parameters, relative humidity, and measurement height. The data for three of four outbreaks were considered, and they showed good mixing of outbreak plumes, which determined the weak height dependence of both the lidar ratio and the column Angström exponents. According to the results of correlation analysis, the Angström exponents in the wavelength range of 532–800 nm increased with the measurement height, which reflected the weaker effect of the coarse aerosol fraction on the optical properties. The extinction coefficient was found to correlate with the relative humidity: as the latter increased from 30 to 80%, the volume extinction coefficient increased 2.2 times. No correlation was detected between the Angström exponent and the extinction-to-backscatter ratio. This can be explained by the fact that if the Angström exponent is largely sensitive to the aerosol microstructure, then the lidar ratio mostly depends on the chemical composition of aerosol particles. During the outbreaks, AOD varied within 0.04–0.21, and the Angström exponent changed from 1 to 1.5 (as estimated from observations in the 400–900 nm wavelength range).

Kirchhoff et al.,[19a] deduced AOD from measurements of narrowband direct solar UV-B (280–320 nm) radiation at S. Paulo (23.55°S, 46.73°W) made with a Brewer spectrophotometer. These measurements were compared with the results of similar observations at C. Grande (Brasil). Analysis of the spectral dependence of AOD showed that it usually increases with the increasing wavelength. This means, in particular, that in the most cases the difference between the absolute AOD values at 320.1 and 306.3 nm (at the edges of the spectral range under study) is positive. However, this difference is not always larger than the measurement uncertainty/error. This intriguing result opposes the Angström law, applicable to a much larger dynamic range in wavelength. For the C. Grande data (with generally larger values of optical thickness), the difference is positive in 16 out of 19 cases, but there are only 4 cases when the difference exceeds the measurement error. For the S. Paulo morning data, however, the opposite is true: the difference is positive with only one exception, and there are 13 cases out of 17 where the difference is exceeds the error.

Ichoku et al.,[17] described the results of testing five Microtops II Sun photometers used for measuring the aerosol optical depth $\tau_{a\lambda}$ and precipitable column water vapor (W). Each photometer derived $\tau_{a\lambda}$ from measured signals at four wavelengths (340, 440, 675, and 870 nm) and W from the 936 nm signal measurements. The accuracy of $\tau_{a\lambda}$ and W determination depended on the reliability of the relevant channel calibration coefficient (V_0). It was found that the manufacturer-determined value of the instrument constant for the 936 nm filter ($k = 0.7847$) used in the Microtops' internal algorithm is unrealistic, causing large errors in $V_{0(936)}$, τ_{a936}, and W. Therefore, it was recommended to determine τ_{a936} by logarithmic extrapolation from τ_{a675} and τ_{a870}. The main sources of error are inaccurate pointing to the Sun, neglect of cleaning the input quartz window, and the neglect of correct calibration. If these three issues are adequately taken care of, the Microtops can be quite accurate and stable, with root-mean-square (rms) differences between corresponding retrievals from clean calibrated Microtops and the AERONET Sun photometer being about ±0.02 at 340 nm, decreasing down to about ±0.01 at 870 nm.

Mattis et al.,[29] for the first time, simultaneously measured the height profiles of the extinction-to-backscatter ratio (lidar ratio) of desert dust particles at 355 and 532 nm. The observations were performed with an advanced Raman lidar during two long-range Saharan dust outbreaks in Leipzig, Germany (51.3°N, 12.4°E), in August and October 2001. Unexpectedly large lidar ratios, mainly between 50 and 80 sr, were found in the Saharan dust plumes. The lidar ratios at 355 nm were, on the average, higher by 10–30% than the ones at 532 nm, are such probably due to the enhanced light absorption in the UV. The large lidar ratios can be explained by model calculations, available in the literature for 532 nm, which focus on the deviations between the scattering characteristics of spheres and spheroids.

Shimota et al.,[34a] discussed the technique and results of retrieving the spectral dependence of the aerosol extinction coefficient (AEC) in the lowest atmospheric layer (0–2 km) from the downwelling atmospheric infrared radiation measured by a Fourier Transform spectrometer near Tokyo. Then the retrieved AEC spectra were used for the retrieval of the volume density of each aerosol component, in

which aerosols in the lowest layer of the urban area were assumed to consist of three components, namely, water-soluble, soot, and dust-like components. The results compared with backscattering intensity profiles and the size-selective particle number density, which were measured from synchronized operations of a lidar and a laser particle counter, respectively. The retrieved values of water-soluble and dust-like aerosol components were found to agree with the results of lidar and laser particle counter. Regarding the soot component, it was impossible to make a direct comparison using data acquired with the lidar and the laser particle counter, because they bear no information about this component. However, the spectrum calculated (FASCODE3 technique) applying the retrieved aerosol parameters closely corresponded to the spectrum measured. Therefore, the retrieved soot aerosol parameters were considered to be quite reliable.

POLDER-ADEOS-1 polarized measurements made possible the retrieval of aerosol content in the atmosphere over land, whose widely varying albedo hampered solution of the inverse problem of that kind. Deuzé et al.,[15] developed a technique for retrieval of the aerosol optical depth τ_0 and the Angström exponent α from the POLDER data. This technique was used to draw maps for revealing large-scale features in the spatiotemporal variability of τ_0. Comparison with the data of synchronous ground-based observations showed quite good agreement between τ_0 and α values in the case of fine aerosol, but in the presence of coarse aerosol some discrepancy arose that could be explained by the fact that radiation polarization is largely determined by fine aerosol. To overcome the relevant retrieval difficulties, the concept of aerosol index $AI = \tau_0\alpha$ was introduced; this index turned out practically identical for the data of ground-based observations and remote polarization sensing. This invariance is confirmed by analysis of observations over land and ocean in the coastal zone.

2. Aerosol radiative forcing

The key role of atmospheric aerosol as a climate-forming factor causes the need in obtaining adequate global information characterizing the aerosol properties. However, the solution of such a problem is strongly complicated by a wide spatiotemporal variability and diversity of aerosol characteristics. The latter is especially true for the aerosol optical properties varying from pure scattering (sulfate aerosol) to strong absorption (soot aerosol), which is of extreme importance for reliable determination of the aerosol radiative forcing. The most reliable way to solve the problem discussed is a combination of satellite and ground-based sensing facilities along with *in situ* measurements of microphysical and optical properties using ground-based, balloon-borne, and airborne instrumentation. A particular part belongs to combined field experiments conducted in order to carry out thorough analysis of the climate-forming role of aerosol under various conditions. Russian investigators followed just this way in the last decades,[2–8,21–23] and then their results were supplemented with similar developments made in the USA, Western Europe, and Japan. Kaufman with co-authors[19] gave some examples of AOD data obtained from satellite and ground-based observations for four types of aerosol: anthropogenic aerosol (due to regional pollution of the atmosphere), black carbon – soot and dark organic material produced from incomplete combustion of fossil fuel and from vegetation fires, mineral dust, and oceanic aerosol. The possible aerosol effect on clouds and precipitation was briefly discussed as well.

Affecting the processes of sunlight scattering and absorption in the atmospheric boundary layer (ABL), the atmospheric aerosol thus impacts the vertical distribution of the radiative heat influx, which results in a complex and interactive set of the following processes determining the ABL dynamics and thermal conditions. The tabulated data borrowed from Ref. 35 demonstrate possible impacts of aerosol and their consequences.

Major ABL perturbations induced by purely scattering and strongly absorbing aerosol

ABL parameters	Aerosol impacts	
	Purely scattering aerosol	Strongly absorbing aerosol
Solar flux at the surface	reduction	large reduction
ABL solar heating	negligible change	large increase
Sensible heat flux	reduction	large reduction
Evaporation	decrease	decrease
$T_x - T_{2m}$*	small decrease	large decrease
ABL temperature	decrease	increase
ABL humidity	a) small increase (dry-soil) b) decrease (wet-soil)	decrease
Inversion strength	increase	decrease
ABL growth/collapse	negligible change	later growth/earlier collapse
ABL height	decrease	a) large increase (dry-soil) b) small increase (wet-soil)
Entrainment	decrease	increase
Probability of cloud/fog formation	a) increase (dry-soil) b) decrease (wet-soil)	decrease

* Soil–air temperature difference.

Through the scattering and absorption of light, aerosols reduce the solar radiation reaching the surface, and the sum of the sensible and latent heat fluxes also decreases and induces important feedbacks. The ABL can either be moistened, in case of purely scattering aerosol, or be dried, for the case of absorbing aerosols, hence either suppressing or promoting evaporation. The absorption of radiation by aerosol reduces the surface–air temperature difference and hence sensible heat fluxes. The partitioning of energy between sensible heat fluxes and evaporation depends not only on this absorption but also on the soil moisture. If sensible heat fluxes are reduced, so is the heating of the overlying atmosphere. However, aerosols directly heat the atmosphere due to absorption. Thus, the purely scattering aerosols reduce the temperature of the ABL, but with strong absorption, increase it.

The average heat gain or loss induced by these perturbations is balanced mostly by changes of the adiabatic heating. In addition, entrainment heating also contributes during daytime, either positively for strongly absorbing aerosols or negatively for purely scattering aerosols. Aerosols alter the growth of ABL by changing both the surface buoyancy flux and capping inversion. A reduction in surface buoyancy flux delays growth and promotes collapse of the ABL. The reduced ABL temperature along with the reduced buoyancy flux in the purely scattering case increases the strength of capping inversion, lowering the top of ABL and hence reducing the entrainment heating and drying.

On the other hand, the increased ABL temperature in the case of absorbing aerosol raises the top of ABL in spite of the reduction in the buoyancy flux. For strongly absorbing aerosols, this elevation of the ABL can be substantial and the entrainment heating and drying increase.

The combination of the increased entrainment drying and the reduced surface evaporation decreases the ABL moisture. The decrease of the *RH* with absorbing aerosol decreases the probability of formation of clouds in the boundary layer, thus causing an additional warming through cloud-feedback effects. Yu et al.,[35] have illustrated the above qualitative characterization by quantitative estimates obtained with the CAPS 1D interactive model of the atmosphere–vegetation–land system for the conditions of fine weather on July 15 at 40°N latitude. The results of numerical simulation, naturally, depend significantly on the aerosol microstructure and optical properties. This determines the urgency of the corresponding combined ground-based, airborne, balloon-borne, and satellite observations along with modernization of numerical models.

The optical properties and hence the radiative forcing of atmospheric aerosols are determined, in part, by the way in which various constituents are externally or internally mixed. The mixing must be known to compute the effective refractive index, water activity, and size distribution of the aerosols. Lesins et al.,[26b] found that the percentage difference in the optical properties, including extinction, single scattering albedo, and asymmetry parameter, between an internal mixture and external mixture of black carbon and ammonium sulfate can be over 25% for the dry case and over 50% for the wet case at typical mass mixing ratios. The differences are the result of a complicated combination of nonlinear Mie theory on the refractive index, assumptions about the coagulated particle sizes for internal mixtures, and the role of water uptake and deliquescence as a function of relative humidity.

The computed optical properties were then used in Ref. 26b to estimate the globally average clear-sky direct (due to aerosol backscattering and absorption of sunlight) radiative forcing under different assumptions on mixing. The results were displayed as a function of relative humidity to conveniently see the mixing effects for dry aerosols at temperatures below the crystallization point, for dry internal and wet external mixtures between the crystallization and deliquescence points, and for fully wet mixtures above the deliquescence point. For a 9:1 ammonium sulfate to black carbon mass ratio, nearly all the cooling effect predicted for an external mixture is lost for the internally mixed assumption, especially for relative humidity less than the deliquescence point. The corresponding absolute values of radiative forcing (at the total aerosol content equal to 10 mg/m^2) can achieve 0.5 W/m^2. This means that the results obtained are very important from the viewpoint of estimating the anthropogenic impact on climate, since this level of changes in the radiative forcing is comparable with the direct anthropogenic forcing values. This reflects the need in taking the considered effect into account in climate models.

As Lohmann and Lesins[27] noted, the increasing contribution of anthropogenic components to the sulfate and carbon aerosol in the atmosphere has led to a significant growth of the globally mean aerosol loading as compared with the pre-industrial epoch, which must, undoubtedly, affect climate formation. The direct aerosol impact on climate is caused by aerosol absorption and scattering of short-wave radiation. Besides, aerosol can also produce the indirect effect, functioning as cloud condensation nuclei (and thus affecting the number density of cloud droplets), modifying cloud albedo and processes of precipitation formation, as well as lifetime of warm clouds. At a constant total cloud water content, an increase in the concentration of fine droplets can lead to the growth of cloud albedo, but slows down cloud formation and increase the cloud lifetime.

The global mean total anthropogenic aerosol effect was estimated to be from −1 to −4.4 W/m^2, but these values should be considered only as the tentative ones. According to the numerical simulations and satellite observations, for two weeks the cloud albedo may increase due to the aerosol

effect from 0.02 to 0.15 (at the unchanged total water content). Analysis of POLDER data revealed a decrease in the size of cloud droplets at the increase of the aerosol index *AI* characterizing the total content of aerosol particles in the atmospheric depth. This means that aerosol significantly affects the microphysical properties of clouds on the global scale.

Recent revision of estimates of the indirect anthropogenic aerosol effect on radiative forcing has narrowed the range of possible variations of the radiative forcing values to 0 to -1.2 W/m^2, which reflects the need in further refinement of the estimates. In this connection, the radiative forcing was assessed in Ref. 27 through numerical simulation of climate using the ECHAM-4 model. Analysis of the results obtained from the climate model with and without indirect aerosol effects suggested that the model agrees with satellite (POLDER) observations better if the indirect aerosol effects on climate are included. However, the simulated clouds were found more susceptible to aerosols than the observed clouds. By taking the difference in susceptibilities into account, the global mean total anthropogenic aerosol effect was reduced from -1.4 to -0.85 W/m^2.

Every year in winter and spring the aerosol haze of anthropogenic origin covers the most part of the Northern Indian Ocean, South and Southeast Asia, which determines the uniqueness of this region from the viewpoint of the possibilities of analyzing the climate impact of the radiative forcing of the absorbing aerosol. This analysis is especially important, because anthropogenic emissions in the considered regions have significantly increased in recent decades. Thus, for example, in South Asian region the atmospheric emissions of sulfur dioxide from fossil fuel burning increased by more than two times for the last 20 years.

Aerosol measurements during the Indian Ocean Experiment (INDOEX) revealed several inorganic (including sulfate) and carbon (including black carbon) components in the aerosol composition. Such aerosols in the tropical atmosphere are rather strongly absorbing (single scattering albedo in the mid-visible spectrum achieves 0.87–0.90), and the contribution of anthropogenic components to formation of the aerosol optical depth and radiative forcing is up to 70% and higher.

A distinctive feature of the South-Asian aerosol haze is its pronounced annual trend. Haze formation begins in winter during the pre-monsoon dry season at the presence of stable inversion in the atmospheric boundary layer and transport of atmospheric pollutants by the northeast wind in the lower troposphere. However, the aerosol loading of the atmosphere turns out to be low in June–September, when atmospheric aerosol is scavenged by precipitation during the summer monsoon.

The INDOEX findings showed that the effect of absorbing and scattering aerosol on the short-wave radiation transfer causes about a threefold decrease in the diurnal sums of radiative heating at the surface level under clear-sky conditions as compared with the decrease of radiative forcing at the atmospheric top level due to atmospheric absorption by black carbon, dust, and ash. In the 0–25°N belt the aerosol-induced decrease in solar radiation income varies from -10 to -30 W/m^2, and aerosol-induced heating of the 0–3 km layer consumes 10 to 30 W/m^2.

In connection with these circumstances, Krishnan and Ramanathan[24] have performed analysis of the observed surface-temperature variations over the Indian subcontinent, which filtered out effects of greenhouse gases and natural variability. This analysis revealed that the absorbing aerosols have led to a statistically significant cooling of about 0.3°C since the 1970s, which is characterized by the annual trend consistent with variations of the aerosol loading of the atmosphere. The surface cooling underneath the polluted regions is likely balanced by warming elsewhere through transfer of heat released in the troposphere at absorption of short-wave radiation enhanced by aerosol. If radiative forcing in the lower troposphere is +14 W/m^2 on the average for 1996–1999, then at the surface level it is -14 W/m^2, and at the atmospheric top it is close to zero: 0 (± 2 W/m^2). To confirm these results, further field observations and numerical simulations are needed.

Krüger and Grassl[26] have assessed the influence of anthropogenic emissions on cloud albedo over Europe using a set of satellite measurements from 1985 to 1999. Special emphasis was given to the Central European main emission area, including the so-called "Black Triangle," which covered parts of Germany, the Czech Republic, and Poland. Due to the decrease of aerosol precursor gases in the period under consideration this analysis revealed a pronounced decrease of cloud albedo by about 2% from the late 80s to the late 90s. During winter in source regions of anthropogenic particulate matter emissions the cloud reflectance was more than 5% lower referring in addition to an absorption effect caused by black carbon in clouds. The radiative transfer calculations indicated for the classical Twomey effect a change in radiative forcing of about 1.5 W/m^2 from the late 80s to the late 90s. In addition during winter a radiative forcing of about 3 W/m^2 due to the absorption effect was estimated.

Chameides et al.,[11] considered two independently derived data sets characterizing atmospheric aerosol properties. The one represented annually mean distribution of anthropogenic aerosols over East Asia derived using a coupled regional climate/chemical transport model. The other was the annually mean distributions of cloud optical depths and cloud amount over East Asia derived by the International Satellite Cloud Climatology Project (ISCCP) for 1990, 1991, 1992, and 1993.

The comparison of the two fields of atmospheric aerosol characteristics revealed a remarkable similarity in the distributions of model-calculated

anthropogenic aerosols and ISCCP-reported cloud optical depths, with both exhibiting a region of enhanced values extending over the east central portion of China, between the Sichuan Basin and the Yangtze Delta, and then in an easterly direction over the East China Sea, Japan, and South Korea, and the western edge of the Pacific Ocean. Linear regression between the estimated aerosol column burdens and the cloud optical depths yielded an $r^2 > 0.6$, indicating that the correlations are statistically significant at a confidence level that is > 99.9% and that more than 60% of the variation in the cloud optical depths is related to variations in the anthropogenic aerosol abundances. Multivariate analysis involving the distributions of boundary layer relative humidity and precipitation over East Asia, as well as that of the model-calculated anthropogenic aerosols and the ISCCP-reported cloud properties, indicated that the relationship between anthropogenic aerosols and cloud optical depth is unique to these two variables and not symptomatic of a more general mechanism involving the hydrologic cycle. Trend analysis of the ISCCP data suggested that there was an upward trend in cloud optical depths over some areas in the East Asia impacted by air pollution during the early 1990s that would have corresponded to the likely increase in anthropogenic aerosol concentrations over this period in East Asia in response to growing anthropogenic emissions. It is a likely explanation for these findings that there is a mechanistic coupling between anthropogenic aerosol concentrations and cloud optical properties; one such mechanism is the so-called first and second indirect effect by which aerosols enhance the optical depths and albedos of clouds by increasing the number of droplets within clouds and suppressing precipitation from clouds, respectively.

The regressions further suggested that the cloud optical depths increased, on the average, by 0.16 for each 1 mg/m^2 increase in the column-integrated anthropogenic aerosol burden. Simple box-model calculations showed that this is equivalent to a cooling over the model domain from anthropogenic aerosols via the indirect effect that is a factor of about 1.5 times that from the direct effect. Having in mind possible underestimate in the model-simulated aerosol concentrations over the model domain of as much as 40%, the estimated cooling from the indirect effect may prove roughly equal to that from the direct effect. In contrast to the results obtained using ISCCP-derived cloud optical depths, the correlation between the model-calculated anthropogenic aerosols and average cloud amount over the model domain was relatively weak and inconsistent. Possibly this result could be caused by the opposite effects on the cloud from their lifetime and frequency of occurrence and, on the other hand, by an indirect effect of the second kind (as well as by the so-called semi direct effect, i.e., the suppression of clouds by absorbing aerosols).

A particular attention should be paid to the carbon aerosol (usually called black carbon BC), which contains soot and dark organic material, being the products of incomplete combustion at biomass burning (including forest fires) and from internal combustion engines. Unlike the purely scattering aerosol that causes climate cooling, the BC impact leads to warming, which achieves globally 0.5–0.8 W/m^2, that is, roughly one third of the greenhouse-gas warming. Just this determines the recent understanding of the role of BC as one of the key components of the climate forcing. Particularly high BC concentration was found in the tropical biomass burning regions, as well as in the polluted atmosphere of the East and South Asia. Observations over the Indian Ocean showed that the short-wave radiation absorbed by BC is on the average about 7% of the net radiation, that is, roughly 17 W/m^2, which can slow down the hydrological cycle and decrease the cloud amount. It is an important circumstance that BC in East Asia can undergo long-range transport.

Since the existing estimates of aerosol-induced radiative forcing are quite approximate, Kaufman et al.,[19] proposed a technique using the oceanic sun glint used as a bright background against which aerosol absorption is measured. This technique enables one to determine the distribution of aerosol absorption optical thickness with an error of ±25% for aerosol optical thickness of 0.2 to 0.4 at 500 nm wavelength (18% of the ocean observations are for this range of optical thickness). The monthly mean aerosol absorption was estimated to have an error of ±12% and uncertainty in the single scattering albedo of ±0.02. A spaceborne technique to perform the measurements in order to retrieve the aerosol-induced sunlight absorption was justified in Ref. 19. The technique discussed can provide obtaining the most reliable results at a moderate wind (~7 m/s) and small solar zenith angles (< 45°), but fails to be reliable enough at a high (≥ 10 m/s) or low (≤ 3 m/s) wind, when the sun glint is too narrow and insufficiently bright.

The numerical simulations with an interactive account for radiative forcing are, naturally, important for reliable estimation of aerosol radiative forcing. Using the global climate model National Center for Atmospheric Research Community Climate Model Version 3 (NCAR CCM3), Kristjánsson[25] obtained new estimates of the indirect climate effect of the sulfate aerosol and black carbon caused by the aerosol effect on the cloud cover dynamics. Two components of the indirect aerosol effect on clouds, i.e., the radius and lifetime effects, were considered. The radius effect is connected with the fact that appearance of additional aerosol particles as cloud condensation nuclei leads to a decrease in the size of cloud droplets. The lifetime effect manifests itself as suppression of droplet coagulation due to a decrease of the particle size and, as a consequence, the

increase of the cloud lifetime. Both these effects lead to an increase in the cloud albedo.

The global concentrations and horizontal distributions of aerosol were obtained in Ref. 25 from simulations with a life-cycle model incorporated into the global climate model. Then they were combined with size-segregated background aerosol. The dynamics of the aerosol size distributions was described taking into account condensation, coagulation, and moisture variation. By making assumptions on supersaturation, number concentration of cloud droplets in water clouds was determined. Cloud droplet sizes and radiative fluxes at top of atmosphere (TOA) obtained from this numerical simulation were in a good agreement with the observations from satellite.

Using aerosol data for 2000 from the Intergovernmental Panel on Climate Change (IPCC),[13] it was found that in case of averaging over the globe, a 5.3% decrease in cloud droplet radius (by 0.58 μm at mean droplet radius of 10.31 μm) and a 4.9% increase in cloud water content due to anthropogenic aerosols occur. The largest changes were found over SE Asia, followed by the North Atlantic, Europe, and the east of the United States. This is also the case for the radiative forcing ("indirect effect"), which has a global mean of -1.8 W/m^2, and the contributions from droplet radius and lifetime changes, respectively, -1.3 and -0.46 W/m^2. When the experiment was repeated using data for 2001 from the IPCC A2 scenario, an unchanged globally mean radiative forcing is found, but the horizontal distribution has been shifted toward the tropics.

Sensitivity experiments showed that the radius effects ~3 times stronger than the lifetime and that black carbon contributes only marginally to the overall indirect effect. These estimates are approximate and this is largely determined by the neglect of some aerosol types, for example, organic carbon aerosol (because the needed information about the aerosol is lacking), as well as ice clouds and the effect of the considered processes on the long-wave radiation transfer. It is planned in the future to complement this climate model with considerations of the atmosphere–ocean interaction and to pay more attention to regional effects.

Lelieveld and Mihalopoulos[26a] discussed the results of six-week-long (July–August 2001) investigations of chemical processes in the Mediterranean atmosphere performed by specialists from eight countries within the framework of the MINOS (Mediterranean Intensive Oxidant Study) project. The MINOS observation campaign (conducted from Crete) included ground-based and airborne measurements of the chemical composition of the troposphere and the radiative conditions. The major tasks were to study the long-range transport of natural and anthropogenic minor gaseous and aerosol constituents in order to analyze their effect on the air quality and climate.

The results obtained indicate that anthropogenic emissions can give rise to large-scale changes of the air quality and precipitation reduction. Analysis of observations revealed the high pollution level all over the tropospheric depth up to 15 km at the maximum anthropogenic contribution near 4 km (this is caused by long-range transport from Western and Eastern Europe). Pollution sources were the industries, traffic, forest fires, agrowaste burning.

During the considered period, especially significant was the income of biomass burning products from the region to the north of Black Sea to the eastern Mediterranean. Because of high insolation (under conditions of almost clear sky) the phenomenon of photochemical smog was often observed. The high oxidation efficiency of air masses was determined by the high concentration of the OH hydroxyl radical ($4.5 \cdot 10^6$ 1/cm^3 with the maximum values of $(1.5–2) \cdot 10^7$ 1/cm^3). At the altitudes above 4 km, the pronounced long-range transport of pollution from North America and Asia was observed. About a half of the CO content in the middle troposphere was due to transport from Asia, and about 25–30% from North America. The ozone income from the stratosphere responsible for 25–30% of tropospheric ozone was also pronounced. In the upper troposphere (above 8 km), the pollution layer characterized by high concentration of such pollutants as formaldehyde attributed to anthropogenic emissions in South Asia was clearly seen (especially, in the eastern Mediterranean). During the summer the atmosphere of the whole Mediterranean region was characterized by the ozone concentration exceeding the acceptable standard for 8 hours (53 nmol/mol). Typical values of the concentration were 55–70 nmol/mol at a weak (~10%) diurnal trend.

The aerosol concentration was also high; the fine fraction (<1 μm) consisted mostly of sulfates (35–40%), organic substance (30–35%), ammonium (10–15%), and black carbon (5–10%). The main source of aerosol was burning of fossil fuel and biomass. The aerosol-induced sunlight extinction caused suppression of evaporation and humidity transport, and the aerosol effect on the cloud microstructure could induce precipitation reduction. Since the sulfate income from Western Europe decreased for the last two decades, one can guess that the aerosol impact on climate in the past was more significant.

The estimates of the direct radiative forcing at the top of the atmosphere gave the values of 3.0 W/m^2 (greenhouse gases) and -6.6 W/m^2 (aerosol), while at the surface the radiative forcing due to aerosol achieved -17.9 W/m^2. Lelieveld and Mihalopoulos[26a] emphasized the need in the future combined investigations including the following basic topics: 1) surface processes; 2) atmospheric processes and air quality; 3) atmosphere–ocean interaction; 4) climate and hydrologic cycle. Only such investigations will allow one to form the basis for recommendations

concerning the adequate environmental policy in the Mediterranean region.

Conclusion

The basic conclusion is that the problem of taking into account the climate impact of the atmospheric aerosol is still far from solution for two main reasons (see also Ref. 8a): (1) the paucity of observations on the global aerosol composition and properties and (2) imperfection of the schemes of taking aerosol into account in numerical climate models (only first steps are now taken toward interactive parameterization of the global aerosol dynamics in numerical climate models). The most urgent task is adequate planning of such a block of the Global Climate Observation System (GCOS), which reflects the needs in observations corresponding to the current understanding of the problem. As the current ideas will extend and become deeper, the observation system will be corrected stage-by-stage in parallel with modernization of the climate simulation technique.

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