

Aerosol and climate studies: current state and prospects.

3. Aerosol radiative forcing

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Results of recent estimates of direct and indirect (via changes of cloud albedo and precipitation rate due to aerosol effect on cloud microstructure) aerosol radiative forcing (ARF) are discussed. It is demonstrated that these estimates are difficult to make because of the diversity of aerosol types and its strong spatiotemporal variations. The ARF predictability within global climate models is analyzed, and prospects of further studies are considered.

Introduction

The problem of aerosol effect on climate is by no means new.⁵ For instance, we long know of an important role of stratospheric aerosol as a climate forcing agent of the atmosphere that manifests itself on a global scale in the periods of large volcanic eruptions.²⁷ The mechanisms of influence of the tropospheric aerosol on climate are far more complicated.^{6,7,29,30} Recently, some attempts have been made to incorporate aerosol effects into the global three-dimensional (3D) climate models. Examples of this efforts can be the studies by Quaas et al.⁴² and Watterson and Dix.⁶¹ In the former, an ensemble-type numerical simulation of climate has been performed for the period since 1930 until 1989 taking into account the effect of five greenhouse gases (GG) (CO₂, CH₄, N₂O, CFC-11, and CFC-12) and sulfate aerosol on climate. Consideration of only purely scattering sulfate aerosol (for the period from 1881 to 2100, i.e., with the elements of climate prediction) is also characteristic of the study by Watterson and Dix.⁶¹ Naturally, the inclusion of only purely scattering aerosol leads to the effect of climate cooling. Nonetheless, Watterson and Dix⁶¹ considered not only aerosol direct effect (ADE) but also indirect effect (AIE) on climate (through aerosol-induced change of microphysical and optical cloud properties) that can be not only negative but positive as well.

Recall that the radiative forcing (RF) ΔF is defined as the difference between the effective fluxes of short-wave (long-wave) radiation at the top of the atmosphere and the underlying surface. In the case of globally mean near-surface air temperature,

$$\Delta T_s = \lambda \Delta F,$$

where λ is the sensitivity of climate system to RF.

Quaas et al.⁴² showed that annually and globally average radiative forcing at the top of the atmosphere,

calculated taking into account only GG, had increased from 0.74 W/m² (1930) to 2.07 W/m² (1989). Aerosol direct radiative forcing (ADRF) increased, due to the sulfate aerosol content growth, from -0.2 to -0.5 W/m² over the considered period, while the first aerosol indirect radiative forcing (AIRF), determined by changes of cloud albedo, ranged from -0.6 to -1.3 W/m². If ARF components are considered additive, the net ARF would be weakly negative in the period from 1930 to 1980 (with minimum of 0.25 W/m² in 1956), and then it became positive (up to 0.27 W/m² in 1989). It is important that this evolution has been mainly determined by variations of the first IRF, while the second IRF (being estimated quite unreliably) was completely disregarded. The "greenhouse" RF under clear-sky conditions is approximately 0.1–0.2 W/m² stronger than it is under mean cloud cover conditions (this difference is determined by the influence of upper-level clouds that strengthen the greenhouse effect). The contribution of aerosol to long-wave ARF is relatively small (though significant).

The net greenhouse effect (GHE) of the atmosphere can be defined as

$$\text{GHE} = [\varepsilon \sigma T_s^4 + (1 - \varepsilon) F_{s\downarrow}] - F_{\text{TOA}\uparrow},$$

where ε is the surface emissivity; σ is the Stephan–Boltzmann constant; and $F_{s\downarrow}$ and $F_{\text{TOA}\uparrow}$ are the fluxes of long-wave radiation at the surface level (sky counterradiation) and at the top of the atmosphere (outgoing radiation). With f denoting the cloud amount, under conditions of partial cloudiness,

$$\text{GHE}^a = f \text{GHE}^{cc} + (1 - f) \text{GHE}^{cf}.$$

Here the subscripts a, cc, and cf denote actual cloud-cover, overcast, clear-sky conditions, respectively. As mentioned above, generally we have that $\text{GHE}^{cc} > \text{GHE}^{cf}$. It is noteworthy that the RF estimates obtained in the framework of climate model

and from radiative flux data differ. The RF differences for clear and cloudy skies are, respectively, 0.23 to 0.49 and 0.13 to 0.37 W/m², which is due to the inclusion of cloud feedback effect in the climate model.

Pirjola et al.⁴¹ have addressed the question on how important is the inclusion of nucleation process into the numerical simulation on regional/global scales. Awareness of significant role of fine-mode aerosol in climate processes and its effect on human health stimulated the interest in the study of this aerosol, mainly being a secondary aerosol as a product of gas-phase conversion: nucleation and subsequent particle growth. This study has been facilitated by perfection of instrumentation presently capable of measuring characteristics of particles larger than 3 nm in size. In the free troposphere (near evaporating clouds, in atmospheric marine boundary layer, and in boreal forests) such an interesting phenomenon as “explosive” particle formation was discovered.

As noted by Pirjola et al.,⁴¹ different nucleation mechanisms have been proposed in this regard to explain the new particle formation. The possible mechanisms include (under conditions of real atmosphere): classical theory of binary nucleation H₂SO₄–H₂O with subsequent formation of hydrates; later hypothesis of ternary nucleation (H₂SO₄–H₂O–NH₃), dimer formation, and ion-induced nucleation. The particles involved in the nucleation process, with characteristic sizes of about 1 nm, however do not present interest until they grow to a detectable size, with particle growth depending on the presence of condensable vapor. On the other hand, particles decrease in number concentration because of their coagulation.

Numerical modeling of nucleation and subsequent particle growth in the context of their parameterization within the frameworks of regional and global models is a difficult task. Therefore, it is important to clarify, under which conditions the fresh nucleated particles are able to survive the coagulation barrier and grow to the Aitken mode and further to cloud condensation nuclei thus affecting the climate. Using sectional aerosol dynamics model AEROFOR, Pirjola et al.⁴¹ analyzed the roles of nucleation processes in aerosol formation. It is assumed that, in addition to other condensable vapor, the atmosphere contains vapor of sulfuric acid participating both in the processes of nucleation and condensation.

Results obtained by Pirjola et al.⁴¹ indicate that the nucleation is a significant process in the atmosphere almost always, excluding cases when the concentration of condensable vapor is insufficient for nucleating particles to be able to reach sizes of Aitken mode and ensure the growth of concentration of particles larger than 20 nm in size by at least 10%. Thus, the parameterization of nucleation process must become an indispensable component of climate models. Development of this process depends strongly on the total number of particles participating in nucleation, total concentration of vapor (H₂SO₄ and vapor of organic compounds), as well as on the number concentration and microstructure of preexisting particles (i.e., on the condensation sink). The numerical

simulation showed that if the total concentration of vapor is higher than $(1-5) \cdot 10^7$ cm⁻³, and the number concentration of particles that undergo nucleation exceeds 100–3000 cm⁻³, but is lower than 10^7-10^8 cm⁻³, then, depending on the intensity of condensation sink (under conditions of clean marine, rural, and urban air masses), the particles can quite rapidly reach the size of 20 nm.

As noted by Andreae et al.,¹⁰ in the past and presently the contribution of aerosol as a climate cooling agent was considerable, however, in the future it may be much weaker as a consequence of measures undertaken to reduce atmospheric pollution. Naturally, this situation must favor intensification of the process of global warming.

An important progress in numerical climate modeling, aimed at estimating aerosol effect under conditions of Arctic, was achieved by Hu et al.²³ As known, large attention during last decades was paid to the effect of enhancement of the process of greenhouse climate warming at high latitudes, determined from calculated data.³⁰ On the other hand, analysis of observation data did not reveal the presence of this effect over Arctic ocean.^{12,25} Therefore, it is of doubtless importance the results by Hu et al.²³ who, using Northern Aerosol Regional Climate Model (NARCM), obtained estimates of quantitative aerosol contribution to Arctic climate change taking into account the direct and indirect effect of aerosol of different types including sulfates, Arctic haze, black carbon (BC), sea salts, organic compounds, and dust. Important findings in this regard include the dependence of climatic aerosol effect on aerosol microstructure and chemical composition, as well as on the high level of annual mean surface-level ARF due to sulfate aerosol, reaching -7.2 W/m².

Although independent (“isolated”) ARF estimates are quite conditional, their abundance and certain information content stimulate replenishment of earlier overviews concerning this problem.^{2,6,7,29,38}

1. Aerosol radiative forcing

As the ARF problem has been discussed in detail earlier,^{3,4,6,7} here we will consider only latest results. The ARF estimates can be calculated using preselected atmospheric models. However, insufficient adequacy of these models stimulated Costa et al.^{16,17} to resort to retrieval of required aerosol parameters from both low earth orbit (LEO) and geostationary earth orbit (GEO) satellite remote sensing data (in the case of LEO, the aerosol optical depth (AOD) is obtained through inversion of high-spectral-resolution measurements from the Global Ozone Monitoring Experiment (GOME)). Application of this approach was illustrated by Costa et al.,¹⁷ who determined ARF at the top of the atmosphere for three situations: 1) strong Saharan dust outbreaks to the Atlantic Ocean in June 1997; 2) long-range transport of biomass burning products across Atlantic Ocean in 2000; and 3) desert dust aerosol events in the region of Indian

Ocean (2000). The AOD (τ_a) retrievals were checked against the Aerosol Robotic Network (AERONET) sun photometer measurements. For AOD > 0.4 the errors of AOD determination were $0.02 \pm 0.16\tau_a$. The calculations of outgoing short-wave radiative fluxes (serving the source of information for ARF estimates) gave accuracy about $\pm 15\%$.

Moorthy et al.³⁹ and Satheesh et al.^{44–51} obtained ARF estimates at the level of underlying surface and top of the atmosphere (TOA); they used as the input information the results of observations of spectral AOD, aerosol mass concentration and microstructure (including some measurements of black carbon (BC) mass concentration), performed in the intermonsoon period during second phase of the Arabian Sea Monsoon Experiment (ARMEX-II).³⁹ The average AOD value at the wavelength of 500 nm over ocean (from data of shipborne observations) was 0.44, and on the territory adjoining Indian Peninsula they were approximately 0.47 (over the plateau regions of central Indian subcontinent the AOD reached ~ 0.61). The aerosol properties were characterized by specific features determined by the trajectory of aerosol long-range transport. Higher AOD values and more smooth AOD spectra corresponded to the cases of advection of air masses from west Asia, as well as from northwest and west-coastal India. The BC abundance was only $\sim 2.2\%$ of the total aerosol mass (compared to climatological value of approximately 6% in coastal Indian regions in this same season).

The BC aerosol is mainly of anthropogenic origin, as a product of internal combustion, and causes considerable absorption of shortwave radiation. Naturally, the BC content over ocean is smaller than over land. However, even in the atmosphere of remote regions of Pacific and Atlantic Oceans, as well as in Antarctic, considerable BC amounts were detected, showing the influence of long-range transport from other continents. For instance, from data of observations in tropical Indian Ocean in 1998–1999 it follows that BC mass concentration varied in the range 1.5–2.8 $\mu\text{g}/\text{m}^3$ (which is equivalent to 6–14% of the total aerosol mass).

Within the framework of ARMEX field observational experiment, which included the study of composite and black carbon aerosol in 2003 during intermonsoon and summer monsoon periods, Babu et al.¹¹ performed shipborne measurements of aerosol mass concentration (AMC) over coastal region of Arabian sea. Analysis of observations showed that the diurnal AMC variations were weak in March and vanished completely in May–June. The AMC level continuously decreased from $\sim 700 \text{ ng}/\text{m}^3$ (which corresponds to the 2.5% fraction of the total aerosol mass) in March to $\sim 104 \text{ ng}/\text{m}^3$ (0.5% fraction) in June. The corresponding aerosol radiative forcing decreased from $\sim 70 \text{ W}/\text{m}^2$ in winter to $\sim 30 \text{ W}/\text{m}^2$ in intermonsoon and to $45 \text{ W}/\text{m}^2$ in summer monsoon seasons.

The estimates by Moorthy et al.³⁹ of the average ARF gave $-27 \text{ W}/\text{m}^2$ (surface level) and $-12 \text{ W}/\text{m}^2$ (TOA), i.e., the radiative forcing for the atmosphere

was $+15 \text{ W}/\text{m}^2$ (if the greenhouse RF is on the order of $2.5 \text{ W}/\text{m}^2$, it becomes clear that adequate treatment of aerosol effect on climate is very important). The ARF level depends substantially on specific features of long-range aerosol transport. Under conditions of advection from the west Asian and western coastal India, the ARF increased to $-(40–57) \text{ W}/\text{m}^2$ (surface level) and reached $27–39 \text{ W}/\text{m}^2$ for the atmosphere. On the other hand, if the advection was mainly from the region of the Bay of Bengal and from central India, the corresponding ARF values decreased to -19 and $+10 \text{ W}/\text{m}^2$. Recent publications by Satheesh et al.^{44–50} contain detailed information on the results of ARF studies.

In particular, the results by Satheesh et al.⁴⁹ suggest that the average ARF (TOA) over northern Arabian sea (up to 12°N) in periods of winter (summer) monsoons were -6.1 (-14.3) W/m^2 and decreased in southward direction to the values -3.8 (-3.4) W/m^2 at the equator. As to the ARF values (surface level), they decreased from -16.2 (-15.2) W/m^2 for the northern Arabian Sea to -5.5 (-3.5) W/m^2 on equator. In the Northern Arabian Sea, the instant ARF values (surface level) could reach $-50 \text{ W}/\text{m}^2$, decreasing with latitude at a rate of about $3 \text{ W}/(\text{m}^2 \cdot \text{deg lat.})$.

Data of Vinoj et al.^{57,58} suggest that the regionally averaged ARF values (surface level) over Bay of Bengal in February 2003 ranged from -15 to $-24 \text{ W}/\text{m}^2$, while ARF (TOA) varied from -2 to $-4 \text{ W}/\text{m}^2$. These ARF values were less than those observed in the same region in March 2001, but larger than the values recorded over Arabian Sea and Indian Ocean. The aerosol-induced radiative heating was about $0.5 \text{ K}/\text{day}$. Vinoj and Satheesh⁵⁷ showed that the presence of aerosols in the atmosphere over Arabian Sea during summer monsoon season causes decrease of total radiation by as much as $21 \text{ W}/\text{m}^2$, and increase of top of the atmosphere reflected radiation by $18 \text{ W}/\text{m}^2$.

Lubin et al.³⁴ calculated the change of the spectral distribution of the sky counter-radiation and outgoing long-wave radiation (OLR) in the wavelength range $3.9–500 \mu\text{m}$ in order to analyze the aerosol effect in the atmosphere over Indian Ocean on long-wave radiative transfer, and they showed the significance of this effect. For instance, with AOD at the wavelength of 500 nm being 0.3, the increase of sky counter-radiation by $7.7 \text{ W}/\text{m}^2$ occurs, compared with that under conditions of clean atmosphere and decrease of OLR by $1.3 \text{ W}/\text{m}^2$. With AOD increasing to 0.7, the sky counter-radiation increases by $11.2 \text{ W}/\text{m}^2$, and OLR decreases by $2.7 \text{ W}/\text{m}^2$ (compared with that under conditions of clean atmosphere). Approximately 30% of the increase of the corresponding ARF values (TOA) is due to sea salt aerosol, and about 60% of the increase is due to combined effect of sea salt and dust aerosol (the remaining ARF variation is due to anthropogenic aerosol). All these estimates are obtained under assumption that aerosol particles are external mixture. If model of internal mixture is assumed, the estimates of long-wave ARF (TOA) approximately

double, indicating the importance of reliable information on aerosol mixing state. It is also doubtless that the inclusion of long-wave ARF component leads to substantial reduction of short-wave ARF, and so it should be considered by numerical modelers.

Satheesh and Srinivasan⁵⁰ analyzed the relationship between contributions of natural and anthropogenic aerosol to ARF over ocean regions adjoining Indian subcontinent. The presence of powerful sea salt aerosol sources in the ocean and long-range transport of dust aerosol from Arabian and Saharan deserts determines in this case important role of natural aerosol, whose contribution to ARF in April–May is comparable or even a factor of 1.5 larger than the contribution of anthropogenic aerosol, arriving primarily from South and Southeastern Asia in the January–March periods.

Lau et al.³¹ considered the processes responsible for determination of dynamics of Asian monsoon and noted that there occurs ARF-induced heating of atmosphere–land system by 8 W/m^2 , and cooling of atmosphere–ocean system by 78 W/m^2 . The atmosphere itself is heated by 97 W/m^2 over ocean and much stronger (by 115 W/m^2) over land.

In the context of Atmospheric Radiation Measurement (ARM) program, McFarlane and Evans³⁶ made observations of broadband and spectral fluxes of short-wave radiation on the islands of Nauru and Manus in the Western Pacific, which provided the longest observation series of the kind for the region under study. The observations on Nauru Island lasted from June 1999 until May 2000. The average short-wave cloud radiative forcing (CRF) was found to be 48.2 W/m^2 , much less than values obtained earlier during the Tropical Ocean Global Atmosphere Coupled Ocean–Atmosphere Response Experiment (TOGA COARE) field project. However, the cloud and above-mentioned aerosol radiative forcing have very close values.

Bellouin et al.¹³ estimated the biases of calculations of direct short-wave ARF due to the use of the following two assumptions: 1) orthotropic (Lambertian) underlying surface; and 2) sphericity of aerosol particles. These estimates indicate that the neglect of angular dependence of short-wave reflectivity of the surface leads to considerable errors in the instant ARF values in the case of water surface. However, after averaging over a day, the errors are cancelled out, and the error of the average values is reduced to acceptable level of less than 3% (with the exception of Northern Hemisphere winter season). In addition, the daily averaging ensures practical removal of errors arising due to assumption on aerosol particle sphericity, whose inadequacy, however, is manifested in AOD calculation, primarily because of the large difference between scattering phase functions of spherical and nonspherical particles.

To understand how the surface–atmosphere system responds to anthropogenically induced aerosol radiative forcing, it is important to know the relative impacts of different aerosol species on radiative budget. Generally, the estimates of direct ARF are based on

assumptions that different aerosol constituents are internally or externally mixed. However, it is possible, for situation when one component coats the other component and forms the core-shell particle structure, which ARF substantially differs from the corresponding ARF of the internally or externally mixed particles.

In this regard, Chandra et al.¹⁴ performed calculations, which showed that the state of mixing of sulfate or sea-salt and BC aerosols may be one of the causes for the observed “excess” atmospheric absorption (they considered four variants of particle structure: sulfate core coated with BC shell, and, inversely, BC core coated with sea salt shell; single scattering albedos corresponding to these models are 0.78, 0.86, 0.89, and 0.80, versus albedo of 0.90 in the case of external mixture of all components). Earlier estimates of anthropogenic BC aerosol effect on climate should be considered as the lower bound, whereas actual ARF values may be much larger. To obtain the corresponding quantitative estimates, further field observations and theoretical calculations are needed. A critically important problem is that there is almost no data on the state of mixing of aerosol particles on a global scale.

The calculations of aerosol direct effect on climate in the Arctic, based on the regional climate model, showed that the associated changes of near-surface temperature (NST) of air vary in the range $\pm 1 \text{ K}$. However, the aerosol effect strongly varies regionally depending on the surface albedo, atmospheric humidity, and cloud conditions in the region. The aerosol effect on climate has pronounced interannual variations (reaching 2 K in the NST), caused by strong variations of year-specific atmospheric conditions. Rinke et al.⁴³ numerically modeled the dynamics of Arctic climate (in the latitude region above 60°N for spatial resolution of 0.5°) using 19-level regional atmospheric climate model HIRHAM, which (by virtue of its high spatial resolution) can reproduce both large-scale and mesoscale atmospheric circulation. The calculations have shown that, through the aerosol–radiation–circulation feedback, the aerosol scattering and absorption can cause the changes of the spatial structure of atmospheric pressure field in the Arctic, which can substantially modify Arctic teleconnection patterns, including the so-called Barents Sea Oscillations.

In recent years there is observed growing interest in the organic aerosol,^{7,22,29} which can account for over 80% of the total aerosol, and whose formation is determined by complex interactions in the biosphere–aerosol–clouds–climate system. Very scarce studies of organic aerosol (OA) motivates the necessity of research into the following main directions²²: 1) OA sources; 2) OA transformation; 3) OA physical and chemical characteristics; and 4) numerical simulation of the processes of OA formation and transformation.

In this regard, the results obtained by Venkataraman et al.⁵⁶ are of great value who estimated the atmospheric emissions of carbonaceous aerosol (black carbon) from residential biological fuel combustion in South Asia. Venkataraman et al.⁵⁶ suggest that these

emissions can have a substantial climate effect. Estimates of direct and indirect ARF due to organic aerosol, obtained by Maria et al.,³⁵ indicate the comparability of this ARF to that produced by greenhouse gases.

In the context of ARF estimates, the problem is of importance of aerosol–cloud interaction. In the composition of atmospheric aerosol, there are both organic and inorganic species; however, the cloud condensation nuclei (CCN) are generally considered to be composed of ammonium sulfate or sea salts. Recent studies, however, have led to conclusion on large role of organic species capable of substantially influencing the process of drop activation. For example, it was shown that decrease of surface tension in the case of certain surface active organic aerosols can increase the number concentration of cloud droplets, increase the cloud albedo, and, as a result, produce globally mean negative radiative forcing up to -1 W/m^2 .

Influence of aerosol chemical composition, giving rise to a number of effects (dissolving gases and little soluble solid substances, reduction of surface tension due to organic species, as well as change of accommodation coefficient) is found to be comparable with the so-called first aerosol indirect effect on climate. Comparison of laboratory and field measurements of activation with the use of ammonium sulfate in the pure form and hypothetical ammonium sulfate, possessing the same solubility as adipic acid ($\text{C}_6\text{H}_{10}\text{O}_4$) (AA), delayed dissolution in the case of AA leads to a considerable decrease of cloud drop number concentration despite the higher (in the second case) level of maximum supersaturation (the supersaturation level increases because more water is required for dissolving AA particles). In the presence of AA, the number concentration of cloud droplets turns out to be lower because only droplets, on which water is condensed in the amount sufficient for AA dissolution, can be activated. However, the AA-induced activation can be more efficient than in the case of mineral aerosol. After first touch with the cloud, the mineral aerosol particles become sulfate coated after evaporation of clouds, which predetermines their activity as effective nuclei of droplets in the process of subsequent cloud formation.

In view of the above said, Lohmann et al.³² analyzed the nucleation efficiency of AA aerosol particles coated with ammonium sulfate (AS) in comparison with that of completely insoluble mineral particles, also AS coated. In addition, they studied how the process of cloud droplets nucleation with participation of AA changes in response to addition of such surface active species (SAS) as nonanoic acid ($\text{C}_9\text{H}_{18}\text{O}_2$) instead of AS. Calculations showed that addition of only 10% AS to AA dramatically increases its ability to become activated. The fraction of activated cloud droplets reaches 36–97% of the cloud droplets that would be obtained from pure AS, whereas the droplet concentration in the almost pure AA aerosol is only 11–47% of that in the case with AS. Addition of a surface active species, such as

nonanoic acid, instead of AS to AA reduces its activated fraction by 3–34% as compared to the AA/AS system.

The AS-induced decrease of the number concentration of cloud droplets (in comparison with pure-AS case) in the range from 8 to 69% results in a positive increment of RF. On the other hand, replacement of mineral aerosol particles, on which AS condensation takes place, with adipic acid particles produces increase of the number concentration of cloud droplets by 18 to 265% and the corresponding negative increment of RF.

A consequence of aerosol–cloud interaction is the increase of cloud particle number concentration; therefore, as noted by Lohmann,³³ under the influence of anthropogenic aerosol, decrease of the rate of snowfall from supercooled clouds may occur.

Early in July 2002, the midlatitude region of Atlantic Ocean and northeastern United States were overcast with a smoke sheet from forest fires in Quebec, Canada. The optically thick smoke plume was subcontinental in size and had considerable influence on air quality and radiation budget. Moreover, a large amount of trace gas constituents (TGCs), constituting typical biomass burning products such as CO, NO_x , and tropospheric ozone, were emitted into the atmosphere. The tropospheric ozone is formed in regions downwind of biomass burning areas in the photochemical reactions with participation of NO_x , non-methane hydrocarbon compounds, and CO during illumination by solar UV radiation. Generally, the formed TGC gases contain, in addition, sulfur and only minor amounts of SO_2 .

The dominating species in the composition of biomass burning aerosol particles are organic carbon (OC) and black carbon, causing not only scattering (as in the case of sulfate aerosol), but also absorption of solar radiation. It is noteworthy that the aerosol emissions from boreal forest fires in North America ensure weaker absorption (as from forest fires in Amazon River basin) than in the cases of fires in African savannas and southern American cerrado, primarily due to longer snow melting phase.

On July 8, 2002 Taubman et al.⁵⁴ performed aircraft observations of characteristics of two smoke plumes over five locations in Virginia and Maryland, USA: 1) a smoke plume in the layer 2–3 km above the mean sea level, advected from Quebec forest fires; and 2) underlying plume from fossil fuel combustion. These plumes were characterized by strongly increased number concentration of submicron particles that caused enhanced light scattering and absorption, as well as O_3 and CO (but not SO_2) mixing ratios. The mean single scattering albedo at the wavelength 550 nm was 0.93 ± 0.02 in the layer 2–3 km, but in the underlying atmospheric layers it did not exceed 0.95 ± 0.01 . For each of the five locations surveyed, they determined aerosol optical depth at 550-nm wavelength for lower 3-km atmospheric layer that ranged from 0.42 ± 0.06 to 1.53 ± 0.21 .

Calculations of clear-sky aerosol direct radiative forcing by the smoke plume have shown that ARF at

the top of the atmosphere was small relative to the forcing at the surface level, indicating that attenuation of solar radiation due to absorption by the atmosphere was approximately equal to the total extinction by the atmosphere. Thus, a consequence of ARF was cooling at the surface level and atmospheric warming. The morning subsidence inversion positioned the smoke plume in a dense enough layer under the planetary boundary layer. The inversion layer lived until afternoon because of the atmospheric warming up. This created a positive feedback loop that prevented vertical mixing and dilution of the smoke plume, thereby increasing the regional radiative impact.

Medina and Nenes³⁷ analyzed the role of drizzle particles, residing inside stratocumulus clouds, as a collector of giant cloud condensation nuclei (GCCN) and suggested, in this regard, that the presence of film-forming compounds (FFCs) on the GCCN may decrease drizzle particle growth and even cease drizzle formation. It was found that even low mass fractions (as low as 0.2%) of FFCs can significantly reduce GCCN size and weaken their potential as collector drops. This conclusion applies to different conditions of cloud pollution by aerosol, implying that in the presence of FFCs, GCCN may influence the microphysical evolution of clouds less efficiently than it was thought previously.

Indirect effect of aerosol on climate, proceeding as modification of cloud properties under aerosol impact, has different manifestations in warm and ice clouds. The corresponding processes occurring in warm clouds (including the relationship between aerosol, cloud drop number concentration, and precipitation) are much better studied; whereas the mechanisms of aerosol indirect effect on clouds (and, hence, on climate), existing in cases of ice and mixed-phase clouds, remain unclear in many respects. In this connection, Cziczo et al.¹⁸ have performed two complementary studies on one important process, the homogeneous formation of ice by small particles. In the first study, the freezing of atmospheric aerosol was induced using controlled temperature and humidity conditions (in a cloud chamber). In the second study, the chemical composition of the residue of ice crystal samples collected in high-altitude clouds was analyzed. The results obtained show that organic compounds participate unequally in transformation of solid and liquid phases: organic-rich particles preferentially remain unfrozen. This paves grounds to believe that atmospheric emissions of organic species have the potential to influence the interaction of aerosol with cold clouds and, thereby, the climate.

2. Aerosol and climate

To estimate reliably ARF and its role in climate change, it is of key importance to perform joint treatment of both aerosol direct and indirect effects on climate, in numerically modeling the climate. Takemura et al.⁵³ contributed much to solution of this problem. They used global numerical model, representing interactive coupling of SPRINTARS

models of long-range transport of tropospheric aerosol and its radiative impact, as well the atmospheric general circulation model (GCM) reproducing variations of the global fields of cloudiness, precipitation, and temperature, caused by direct and indirect ARF. The microphysical parameterization scheme, used by GCM, precalculates the cloud drop number concentration (based on the Köhler theory) as a function of aerosol particle number concentration, updraft velocity, as well as microstructure characteristics and chemical properties of anthropogenic aerosol of different types (sulfate (SO₂), black carbon, and organic aerosol) using preset global distributions of atmospheric emissions of aerosol of these types in the years 1850 and 2000.

Comparison of the effective radius of cloud drops, CRF, and precipitation rate, calculated taking into account indirect ARF, with satellite observations showed quite satisfactory agreement. The calculations indicate that the effective droplet radius decreases on a global scale, while changes in the cloud water content and precipitation rate strongly depend on specific features of hydrological cycle, caused by temperature variations due to first and second indirect ARF (and not only the second indirect ARF), manifested as a decrease of precipitation with shift of cloud microstructure toward smaller droplets. However, there may occur simultaneous increase of the cloud water content and decrease of precipitation in regions where a large amount of anthropogenic aerosol is observed, which is a strong signal of the second indirect ARF. The globally mean direct and indirect ARF at the tropopause level due to anthropogenic aerosol are respectively -0.1 and -0.9 W/m². Hence, the anthropogenic aerosol may produce approximately 40% reduction of greenhouse increase of globally mean near-surface air temperature.

Table 1 presents annually mean values of direct ARF at the tropopause and surface levels under clear-sky conditions and conditions of observed cloudiness; data refer to the period from beginning of industrial revolution to the present time. Table 2 contains information (for the same time period) on the annually mean ARF at TOA level and on variations of different meteorological parameters.

Table 1. Annually mean direct radiative forcing (W/m²) due to anthropogenic aerosol at tropopause and surface levels under clear-sky and real cloud conditions

Aerosol types	Observed cloud conditions			Clear sky		
	Land	Ocean	Global	Land	Ocean	Global
<i>Tropopause</i>						
BC	0	+0.30	+0.42	+0.67	+0.10	+0.26
OC	-0.49	-0.19	-0.47	-0.84	-0.39	-0.51
Sulfate	-0.36	-0.15	-0.21	-0.90	-0.37	-0.52
Total	-0.09	-0.04	-0.06	-1.07	-0.66	-0.77
<i>Underlying surface</i>						
BC	-1.41	-0.51	-0.76	-1.64	-0.68	-0.94
OC	-0.65	-0.24	-0.36	-0.97	-0.42	-0.57
Sulfate	-0.27	-0.12	-0.16	-0.69	-0.31	-0.41
Total	-2.33	-0.87	-1.28	-3.30	-1.41	-1.92

Table 2. Annually mean ARF at TOA level and variations of the meteorological parameters under the impact of anthropogenic aerosol for the period from beginning of industrial revolution to the present time

Climate characteristics	Land	Ocean	Global
Direct ARF, W/m ²	-0.08	-0.02	-0.04
Indirect ARF, W/m ²	-1.72	-0.65	-0.94
Effective droplet radius, μm	-1.2	-1.1	-1.1
Cloud liquid water, g/m ³	-4.2	-1.8	-2.4
Precipitation rate, mm/day	-0.10	-0.11	-0.11
Near-surface air temperature, K	-1.5	-0.9	-1.0

Of course, the data in Tables 1 and 2 should be considered just as approximate estimates. Takemura et al.,⁵³ in particular, proposed that one of the key missing factors is the role of aerosol particles as sublimation nuclei in ice (cirrus) cloud formation. In comparing the average calculated and actual ARF (discussed above) values, the strong spatiotemporal variations (regionally specific features) of observed values are to be noted that, as a result, make the smoothed globally mean quantities quite conditional. Undoubtedly, from the viewpoint of realistic analysis of climate dynamics, it is highly important to take these variations into account.

Since climate changes in Arctic attract much attention, Hu et al.²³ have recently estimated aerosol effect on climate under conditions of high latitudes, using Northern Aerosol Regional Climate Model (NARCM). The model includes direct and indirect ARF due to aerosol of different types such as sulfate, Arctic haze, black carbon, sea-salt and organic aerosol, and mineral dust, with model precalculated aerosol component. The results obtained by numerical simulation were compared with the data of observations as a part of Surface Heat Budget of the Arctic Ocean (SHEBA) and the Atmospheric Radiation Measurement (ARM) programs in the period from October 1997 until September 1998.

The calculations show that the annual mean surface radiative forcing due to only sulfate aerosols, including both the direct and indirect components reaches -7.2 W/m^2 . The different aerosol types were found to produce quite specific impacts on climate exhibiting, in addition, strong annual variations. Comparisons with observations have shown that incorporation of five aerosol types substantially increases the reliability of numerical climate modeling, determined by variations of NST and other variables (Table 3).

Table 3. Comparison of monthly mean values of different parameters (May 1998)

Variable	Control	Sulfate aerosol	Five aerosol types	Observation data
NST, K	259.46	255.98	259.62	258.79
Atmospheric counterradiation, W/m ²	225.35	93.29	216.885	220.69
Total radiation, W/m ²	227.23	178.38	181.89	189.74
Cloud amount	0.979	0.888	0.890	0.895

The fact that, in contrast to numerical simulation, the observations have revealed no "greenhouse" intensification of climate warming in Arctic²⁵ can be explained, in particular, by a decrease in temperature near surface and its increase in the free atmosphere, caused by strong dehydration due to Arctic aerosol. An important factor may be the complexity of the processes of interactions in high-latitude aerosol-clouds-radiation system in the absence of solar radiation during long period of polar night.¹

Chung and Seinfeld¹⁵ examined the equilibrium climate effect of direct radiative forcing of anthropogenic black carbon (BC) via 100-year simulations at the Goddard Institute for Space Studies General Circulation Model II-prime coupled to a mixed-layer ocean model. Anthropogenic BC is predicted to raise the globally and annually averaged equilibrium surface air temperature by 0.20 K, if the BC is assumed to be externally mixed. If the BC is assumed to be internally mixed with the present-day level of sulfate aerosol, the predicted annual mean surface temperature increase rises to 0.37 K globally, 0.54 K for the Northern Hemisphere, and 0.20 K for the Southern Hemisphere. The climate sensitivity (average NST) to BC direct radiative forcing is calculated to be $0.6 \text{ K} \cdot \text{W}^{-1} \cdot \text{m}^2$, which is about 70% of the climate sensitivity to CO₂ doubling.

Analysis of global distribution of response of temperature field to BC ARF has shown that this response is most strong in the Northern Hemisphere at high latitudes in winter and early spring, when NST increase exceeds 1K.

This NST increase is due to high surface albedo and the presence of positive feedback that manifests itself in warming-induced reduction of snow- and ice-covered area, accompanied by a decrease of surface albedo and growth of absorbed shortwave radiation.

Most appreciable temperature increase in the tropics and midlatitudes (major part of Asia and Atlantic and Pacific Oceans) is concentrated in the upper troposphere, especially near 45°N at 450 hPa level. Direct RF of anthropogenic BC must also lead to a change of precipitation field in the tropics, with an increase of precipitation in the latitude belt 0–20°N and decrease in the 0–20°S zone, accompanied by a shift of intertropical convergence zone northward. The changes of precipitation pattern are enhanced, if BC is assumed to be internally, and not externally, mixed with sulfate. These variations of precipitation pattern must not, however, result in significant impact on the global BC burden in the atmosphere through aerosol scavenging because these changes take place predominately in the regions far from BC sources. This situation reflects the important features of aerosol effect on climate, in that this effect manifests itself in the regions far removed from the main aerosol sources.

Janisková and Morcrette²⁴ performed numerical climate modeling to estimate the sensitivity of parameterization of radiative fluxes to changes of the input parameters. In this regard, they estimated the aerosol effect on outgoing short-wave radiation under

different conditions (depending primarily on the surface albedo).

In a recent study, Hansen et al.^{22a} undertook numerical climate modeling with the use of new Goddard Institute for Space Studies (GISS) model to estimate distortion of the earth radiation budget (ERB), whose results illustrate the reality of the process of global warming. The conclusions of Hansen et al.,^{22a} however, apply to globally mean values, and so these may be as misleading as the hospital-average temperature. Data of Table 4 illustrate the estimates of globally mean ARF.

Table 4. Effective radiative forcing for the period from 1880 to 2003 taking into account greenhouse gases and atmospheric aerosol, as well as other factors

Impacting factor	RF, W/m ²	
Greenhouse gases (GG):		
well-mixed GG	2.75	—
ozone	0.24	—
stratospheric H ₂ O (due to CH ₄)	0.06	—
<i>Total</i>	3.05±0.4	
Extraterrestrial solar radiation	0.22	(×2)
Land use	−0.09	(×2)
Snow albedo	0.74	(×2)
Aerosol:		
volcanic	0.00	—
black	0.43	—
scattering tropospheric	−1.05	—
indirect aerosol effects	−0.77	—
<i>Total (ARF)</i>	—	−1.39±0.7
<i>Arithmetic sum of separate RF components</i>	—	1.93
<i>Interactive incorporation of all RF components</i>	—	1.8±0.85

It is sufficient to compare these results with actual ARF variations to see the approximate character of globally mean estimates.

Conclusion

Two principal unsolved problems in the adequate treatment of atmospheric aerosol as climate forcing agent are the following:

1. Deficiency of information on microstructure characteristics, chemical composition, and optical properties of aerosol of different types. This explains the high importance of complex field observational experiments in context of acquisition of quite full observation data on the processes in the aerosol–clouds–radiation system. On the other hand, it is also no less important to further develop the conventional and satellite systems for routine observations on a global scale.

2. Insufficient development of the global three-dimensional climate models, not in terms of inclusion of *a priori* prescribed aerosol characteristics, but rather in the sense of reproducing internal interactive aerosol dynamics, based on numerical simulation of

the corresponding biological, geological, and physical (carbon, sulfur, etc.) cycles in the framework of numerical climate modeling.

In view of the growing attention to the anthropogenic aerosol problem, Satheesh and Moorthy³¹ have quite timely stressed that it is necessary to continue and further develop the studies of natural aerosol, whose average contribution to the global aerosol content makes about 70%, primarily in the form of contributions from sea-salt and mineral dust aerosols.

The special aspect of the discussed problem is reliance on globally mean estimates of aerosol radiative forcing, which can be misleading in view of the real wide variety of aerosol types and properties, as well as the strong spatiotemporal aerosol variations. A clear illustration of this conclusion may be the study by Lau et al.³¹; it was devoted to analysis of aerosol effect on Asian monsoon and demonstrates the complex spatiotemporal variations of the corresponding processes.

The documents formulating the prospects of further progress of the World Climate Research Program still make no allowance for development of adequate (interactive) treatment of the processes in the aerosol–clouds–radiation system as key climate forcing agents.^{5,8,30} The contribution of atmospheric aerosol is still one of the main factors of uncertainty in estimates of different climate-forcing and, moreover, climate-prediction factors.

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