From nano- to global scales: properties, processes of formation, and aftereffects of atmospheric aerosol impacts. 7. Aerosol radiative forcing and climate

K.Ya. Kondratyev

Scientific Research Center for Ecological Safety RAS/ North-Western International Cleaner Production Centre, St. Petersburg

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Regularities in formation of the aerosol radiative forcing (ARF) and values of ARF as a climate-forming factor are overviewed.

Introduction

The continuing growth of population in coastal regions and the related increase of pollutant emissions into the atmosphere, as well as the extended land use determine the growing load on coastal ecosystems.[13b] In this situation, the anthropogenic impact on climate becomes heavier. Thus, for example, the air masses with the enhanced content of aerosol and minor gases of both natural and anthropogenic origin may come to the ocean from the continents due to the long-range transport. Air masses, containing different chemically active aerosol components and minor gases, over the coastal ocean create the conditions favorable for various heterochemical reactions. The deposition of the products of these reactions on the ocean surface leads to a significant change in the inflow of organics into the ocean, and this affects significantly the ocean biodynamics (coastal ecosystems). The quantitative description of such processes is seriously complicated by the large number of physical and chemical processes proceeding in the transient zone from the continental to marine atmosphere.

The investigations carried out in recent years have led to the commonly accepted conclusion that the atmospheric aerosol and its interaction with clouds are among the key climate-forming factors.[1–71] The aerosol effect on the climate is most often characterized by the aerosol radiative forcing (ARF), and only recently, attempts have been undertaken to interactively consider the aerosol in numerically simulating the climate. Let us discuss some new results obtained in this field.

Chung and Ramanathan[13a] have overviewed the studies on determination of the aerosol radiative forcing from the INDOEX data and on the estimation of possible aerosol effects on the local climate. A characteristic feature of the atmosphere over the most part of the Indian Ocean in the Northern Hemisphere, over the vast regions of Southern and Southeastern Asia is the presence of a thick layer of atmospheric haze in the troposphere every year during dry season (November–May). The presence of this Indian-Asian haze was clearly seen from the analysis of observations within the INDOEX Program in 1995–2001.

The chemical analysis of aerosol has revealed the presence of some inorganic and carbonaceous compounds, including black carbon (BC) clusters, which absorb the short-wave radiation, ash suspended in the atmosphere, and mineral dust. Since the relative BC content in the fine aerosol achieved 14%, the single scattering albedo was about 0.9 over both the land and ocean. The anthropogenic component of aerosol was the major contributor (up to (75 ± 10)%) to the total aerosol content and to the aerosol optical thickness (AOT).

The direct and indirect aerosol radiative forcing was estimated in Ref. 13a with the use of all the available data and satellite observations. The analysis of these data has shown that, at the presence of aerosol haze in the atmosphere, the monthly mean short-wave radiation balance at the surface level decreases by 20–40 W/m^2 and the lower 3-km atmospheric layer is heated by up to 0.4–0.8 K/day, which is equivalent to 50–100% intensification of heating (as compared to that under conditions of clean atmosphere). The monitoring of the haze level during INDOEX suggested the presence of its interannual variations. The southern boundary of this layer varies from 10°S to 5°N.

To analyze the aerosol impact on the climate during the dry season, two numerical experiments were carried out with the use of the CCM-3 model under conditions of two extreme situations: 1) extended (in the area) haze forcing (EHF); 2) shortened (in the area) haze forcing (SHF). Over the Indian territory, where the aerosol effect is most pronounced, the calculated climate changes under the EHF and SHF conditions are roughly identical, and the main change is in cooling near the surface and the enhancement of the inversion in the lower troposphere, what is confirmed by the data of observations.

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Because of stabilization of the atmospheric boundary layer, the evaporation and turbulent heat flux on the land become weaker. The spatial distribution of precipitations, which significantly depends on the location of the southern boundary of the aerosol haze, changes considerably. The forcing factors, manifesting themselves as changes in the evaporation and in the turbulent heat flux, affect the spatial distributions of the atmospheric circulation and precipitations, and the effect of EHF is more significant. One of the key manifestations of the reaction to the presence of the haze layer is the suppression of the convection in the western sector of tropics of the Pacific Ocean, which influences the variability of El Niño. Since in this case the trade winds weaken over the Pacific Ocean and warming is formed in the eastern part of the ocean, Chung and Ramanathan[13a] supposed that the Indo-Asian aerosol haze may play an important role in the formation of the amplitude and frequency of El Niño in the forthcoming decades. The presence of such a relation is also demonstrated by the results of numerical simulation with the use of the Cane–Zebiak model of the atmosphere–ocean system.

Determination of the long-wave radiative forcing due to the Saharan dust aerosol[71] was of great significance for this sort of studies. The monthly average RF value for September of 2000 was 7 W/m^2, which characterizes the marked aerosol-induced warming.

1. Some aspects of the ARF retrieval techniques

The reliability of ARF estimates depends on many factors, one of which is the reliability of the information about the aerosol optical thickness.

As was noted in the paper by Chylek et al.,[14] the maximum permissible error of the satellite-based estimates of the outgoing radiation flux of $\Delta F = 0.5$ W/m^2 requires the accuracy aerosol optical depth (AOD) τ retrievals to be about $\Delta\tau = 0.015$ over the land and to $\Delta\tau = 0.010$ over the ocean. None of the current operational satellite based instruments for AOD retrieval has been able to achieve this accuracy. The RMSE (Root Mean Square Error) of the AVHRR (Advanced Very High Resolution Radiometer) is typically between 0.06 and 0.15, while the RMSE of the MODIS (Moderate Resolution Imaging Spectroradiometer) over the land has been estimated to be $\Delta\tau = 0.05 + 0.2\tau$, which varies between $\Delta\tau = 0.07$ and $\Delta\tau = 0.21$ within the limit of usual aerosol optical depth between $\tau = 0.1$ and $\tau = 0.8$. The Department of Energy research satellite instrument, Multispectral Thermal Imager (MTI), is capable of retrieving aerosol optical depth with an accuracy of $\Delta\tau = 0.03$ using an off nadir viewing at medium scattering angles.

Theoretical analysis shows that the uncertainties in aerosol phase function (due to uncertainties in the aerosol particle shape, size distribution, and optical properties) are the major obstacles for achieving tolerable accuracy of the aerosol optical depth retrievals. These uncertainties lead to a much larger errors in the aerosol optical depth retrievals at large scattering angles (usually at close to nadir viewing) than at off nadir views at medium scattering angles. Based on the theoretical analysis and on MTI experience, it was suggested that in order to achieve the required accuracy in AOD retrieval, future satellite instruments using a single or dual-view AOD retrieval algorithm should use off-nadir views at medium scattering angles (between 50° and 100°).

Myhre et al.[52] have compared the aerosol optical depth (AOD) for an 8-month period (November 1996 – June 1997), derived over the oceans with five different retrieval algorithms applied to processing data of four satellite instruments: AVHRR, OCTS, POLDER, and TOMS. Large uncertainties in the global mean AOD were found. There was at least a factor of 2 differences between the AOD values retrieved. The largest uncertainties, found in the Southern Hemisphere, are probably caused by differences in the cloud screening procedure.

According to the IPCC (Intergovernmental Panel on Climate Change) Report published in 1995, the indirect (connected with the effect of aerosol on the optical properties of clouds) globally averaged aerosol climate forcing, characterized by the aerosol indirect effect (AIE), varies from 0 to -1.5 W/m^2. Six years later (in the IPCC Report–2001) this range was extended: from 0 to -4.8 W/m^2.

As was noted by Brenguier,[6] one of the uncertain factors is mizzle in clouds, which are formed in the atmospheric boundary layer (ABL). In particular, this circumstance illustrates the importance of adequate reconstruction of the dynamics of cloud cover in the ABL. Another problem is connected with the consideration (parameterization) of small-scale processes in the ABL and their nonlinearity. Thus, for example, activation of aerosol as cloud condensation nuclei (CCN) is determined by the upward motions at the cloud base level, which should be reconstructed with the spatial resolution (along the horizontal) about 100 m. Modern parameterization schemes are still far from meeting such requirements.

The needs for taking into account the interactions among various processes determining the cloud cover dynamics and its effect on the microphysical and optical properties of clouds creates serious difficulties. In this connection, the attention was first paid to the aerosol-induced increase in the number concentration of cloud droplets, leading to the corresponding changes in the cloud albedo and AIE. This effect was called the *first indirect effect* of clouds on climate. Then the need for considering the *second indirect effect* was understood. This effect manifests itself through the change in the intensity of precipitation formation under the effect of aerosol. The reconstruction of this effect requires a description of the interrelation between microphysical characteristics of clouds and ABL dynamics. The

significance of the "*semi-indirect*" *effect*, arising due to the aerosol absorption of the short-wave radiation, which prevents the development of cloud cover, was analyzed as well (Johnson et al., Ref. 35a).

It is an important problem to provide for the interactive consideration of all the three types of aerosol impact on the ABL, clouds, and AIE. It was just this problem, the solution of which was among the main objectives of the ACE-2 field observational experiment carried out in 1997 in the region of the Canary Islands. A part of the ACE-2 program (CLOUDCOLUMN) was devoted just to the study of the indirect effect of the anthropogenic aerosol on climate. In 1999 the European Commission supported further investigations in this field within the PACE (Parameterization of the Aerosol indirect Climatic Effects) Project.

As was noted by Mitra,[48a] the Indian Ocean Experiment (INDOEX) was the first integrated problem-oriented international observation program, whose main objective was to study the aerosol-induced radiative and climate forcing on the regional and global climate with the due regard for the corresponding feedbacks. The preliminary stage of INDOEX started in 1996–1997, and the major part of observations was carried out in 1988–1999 with participation of specialists from many institutions and agencies of India, the USA, Europe, and from Maldives and Mauritius. It was a multi-platform campaign combining ground-based, shipborne, spaceborne and airborne observations.

Although the campaign assumed obtaining information on the content and properties of aerosols and most significant optically active trace gases (O_3, CO, NO_x, SO_2, etc.), the subsequent analysis concentrated on aerosols targeted on obtaining data on the direct and indirect radiative forcing of aerosols. The most interesting and often surprising results of this study were the detection of an extensive tropospheric haze layer with an appreciable component of black carbon and clear evidences of long-range transport of aerosols and trace gases.

Obtaining of the variety of information about aerosol has opened the possibility of analyzing its effect on climate, health, and agriculture, while various data on trace gases allowed further investigations within the Chemical Weather Program. An important component of the INDOEX Program was the observation from onboard *Sagar Kanya* Research Vessel (India), which made a cruise in January–March 1999 before the start of the mission by *Ronald H. Brown* Research Vessel (USA), which conducted an east-west cruise along the 20°S parallel in the region of the clean air to the south of the Intertropical Convergence Zone (ITCZ), as well as in the region of the Arabian Sea (along 15°N, toward India). The comparison of ARF values in the regions with the clean atmosphere and in the presence of a thick aerosol layer has shown that in the later case the ARF was 6 to 10 times higher, achieving -35.3 W/m^2 (coastal zone) and -18.6 W/m^2 (dusted maritime atmosphere).

Feingold et al.[24] have checked the Twomey hypothesis on the indirect effect of atmospheric aerosol on climate using the data of ground-based remote observations under the ARM Program at a continental US site in the region of Great Plains (Oklahoma). According to this hypothesis, aerosol particles in clouds, functioning as cloud condensation nuclei, favor the increase of the number concentration of fine particles and thus cause the increase of the cloud albedo, what leads to climate cooling (an important aspect of this hypothesis is the assumption of constant liquid water content of clouds).

The solution of the problem of aerosol–cloud interaction is significantly complicated by the presence of numerous feedbacks, arising due to interactions among microphysical, dynamical, and chemical processes. In Ref. 24, the response of nonprecipitating clouds to changes in aerosol load was analyzed assuming constant liquid-water content of the clouds. This reaction was quantified as the relative change of the cloud-drop effective radius due to relative change in the aerosol extinction. This was done in a single column of air at a temporal resolution of 20 s (spatial resolution of 100 m). Cloud-drop effective radius was derived from data acquired with a cloud radar and microwave radiometer. Aerosol extinction was measured below the cloud base using a Raman lidar. Results suggested that aerosols associated with maritime or northerly air trajectories tend to have a stronger effect on clouds than aerosols associated with northwesterly trajectories that also have local influence. There was good correlation (0.67) between the cloud response and a measure of cloud turbulence.

The processes of interaction in the aerosol–cloud–radiation system, determining the indirect effect of aerosol on climate, are still poorly studied, though they are potentially important factors in the formation of the radiative forcing (the spread of the corresponding estimates is determined by variations from -0 to -4.8 W/m^2). Certainly, the contribution from lower-level stratus clouds to AIE is important, because: 1) their albedo is more sensitive to variations of cloud microphysical characteristics than the high albedo of the upper-level clouds (this is a manifestation of the first indirect effect of aerosol on clouds and climate); 2) moderate geometric thickness of clouds is often sufficient for droplets to achieve the size of precipitating droplets, and therefore even a small increase in the number concentration of cloud droplets N can prevent the precipitation, which affects the liquid water content and albedo of clouds (this is the cause of the second indirect effect of aerosol on climate).

To analyze the formation and variability of AIE and to justify the AIE parameterization, Menon with co-workers[48] have compared, while working in ACE-2 and PACE, six different single-column models (SCMs) of the processes in the aerosol–clouds–radiation system, determining the aerosol effect on climate

under the conditions of clean and polluted atmospheric boundary layer (ABL). This study has been performed in three stages. First, SCMs were configured with the same fine vertical resolution as the ACE-2 *in situ* database to evaluate the numerical schemes for prediction of aerosol activation, radiative transfer, and formation of precipitation. Second, the same test was performed at a poorer vertical resolution. Finally, SCMs were run for a 24 to 48 h period to examine predictions of the boundary layer clouds when initialized with large-scale meteorological fields. Several schemes were tested for the prediction of cloud droplet number concentration (N).

The results obtained in Ref. 48 showed noticeable discrepancies compared to empirical schemes due to biases in the diagnosed cloud base vertical velocity. Prognostic schemes exhibited a larger variability than the diagnostic ones, due to coupling between aerosol activation and drizzle scavenging in the calculation of N.

When SCMs were initialized at a fine vertical resolution with locally observed vertical profiles of liquid water content, predicted optical properties were comparable with the observed ones. However, the agreement becomes worse at a poor vertical resolution. Predicted precipitation fluxes were severely underestimated and improve when accounting the sub-grid variability of the liquid water content. Results from the 24–48 h runs suggested that most models have problems in simulating boundary layer cloud morphology. As a result, models significantly overestimated cloud optical properties. Improved cloud morphologies were obtained for models with subgrid inversions and subgrid cloud thickness schemes.

During the Lindenberg Aerosol Characterization Experiment (LACE 98), airborne measurements of aerosol size distribution, fine-particle concentration, particle absorption coefficient, backscatter coefficient, depolarization, and chemical composition, as well as ground-based measurements of spectral particle optical depth and spectral backscatter and extinction coefficients were performed. Fiebig with co-workers[25] compared the measured optical parameters with the numerically simulated ones; the simulations made assumed the aerosol to consist of sulfuric acid near the tropopause and mixtures of ammonium sulfate and soot in the remaining column. (see also Ref. 53b by Osborne et al.).

This comparison provided for the closure (agreement between the calculations and measurements) within 25% for the optical depth of the column, which included a biomass-burning aerosol of North American origin, and inferred a soot volume fraction of 35% for this aerosol. Assuming the biomass-burning aerosol to be an ensemble of spheroidal particles of prolate shapes with the average aspect ratio of 1.3, the calculated depolarization agreed with the lidar measurement data, whereas comparing the spectral backscatter coefficient showed the soot to be externally mixed with the nonabsorbing particles. With the two-stream approximation, the local, instantaneous, cloud-free radiative forcing of the biomass-burning aerosol at the tropopause was estimated to be -5.8 W/m^2 with a corresponding optical depth of 0.09 at 710 nm wavelength and solar zenith angle of 56°. The radiative forcing of aerosol of the biomass-burning origin is equally sensitive to the state of mixture of particles (internal or external) and the surface albedo.

In the context of the problem on indirect effect of atmospheric aerosol on climate through the aerosol-induced changes in the microphysical and optical characteristics of clouds, Peng and Lohmann[54] have discussed the results of two Canadian field studies of the influence of anthropogenic aerosols on the shape of the cloud droplet size spectra. The calculated cloud albedo scaled by the parameter β, which is a function of the relative dispersion of cloud droplet spectra ε, characterizing the mean droplet radius and the radius variance, showed better agreement with the independently measured cloud albedo values than the cloud albedo calculated without scaling. The scaling factor β $[\beta = (1 + 2\varepsilon^2)^{2/3}/(1 + \varepsilon^2)^{1/3}]$ positively correlated with the cloud droplet number concentration. A linear relationship between β and the cloud droplet number concentration obtained from different field studies was applied to the ECHAM4 general circulation model. The correction made with the account of the parameter β, reduced the global mean indirect aerosol effect at the top of the atmosphere including the dispersion effect by 0.2 W/m^2 as compared with the reference simulation.

The indirect effect of aerosol on climate, manifesting itself in the atmospheric boundary layer, is determined by a series of interactions of the aerosol with the dynamical, microphysical, and optical properties of clouds. The income of aerosol particles, functioning as cloud condensation nuclei, into the atmosphere favors an increase in the cloud droplet number concentration (CDNC). As was already mentioned, the related increase of the optical thickness and the albedo of clouds at the constant liquid water content was called *the first indirect effect*, characterizing the effect of aerosol on climate.

On the other hand, the changes in the cloud microstructure are also of great significance, as those affect the cloud dynamics (in the first turn, through the process of precipitation formation, changing the cloud lifetime and spatial extension, which determine the cloud albedo). This "microphysical" feedback, affecting the dynamics of the cloud cover, was called *the second indirect effect*, which determines the aerosol impact on climate. Although the increase of the cloud albedo related to the manifestations of the indirect effects is relatively small, it can be significant on the global scales as a factor, mitigating the warming due to the greenhouse effect in the atmosphere. That is why the study of AIE and its satellite monitoring are extremely urgent.

The objectives of the CLOUDCOLUMN (CC) Project, one of the five projects within ACE-2,

discussed by Brenguier et al. in Ref. 7 were to study AIE for marine stratocumulus clouds and to justify the strategy of "closed" aerosol–cloud radiative experiments. The observations within the CC Project were carried out in June–July 1997 in the region of the Canary Islands with the use of instrumentation installed onboard three instrumented airplanes and at a ground site.

Reference 7 discussed the results of eight series of airborne observations of the microphysical characteristics of marine stratocumulus clouds in a wide range of the observation conditions (different physical-chemical properties of aerosol, CDNC ranging within 50–25 cm^{-3}, etc.). The obtained unique set of observed microphysical and radiative characteristics of the cloud cover can be used in estimating the indirect effect of aerosol on clouds and climate based on the analysis of the relation between the optical thickness of clouds and the effective radius of cloud droplets. The correlation between these parameters is usually negative, but can become positive under conditions of the heavily polluted atmosphere. In fact, the most polluted cloud systems sampled during ACE-2 were slightly drier, hence thinner, than the marine and intermediate cases, hence producing a positive correlation between optical thickness and effective radius of droplets.

The so-called black carbon (BC), consisting of soot and smoke aerosols absorbing the short-wave radiation, is a product of incomplete combustion of various fuels (first of all, fossil fuels and biomass). The direct radiative forcing caused by BC and organic matter (OM) was estimated to be from +0.16 to +0.42 W/m^2, and the net absorbed radiation was estimated as 0.56–2 W/m^2 (it was proposed to call the sum BC + OM the soot component produced due to the fossil fuel combustion). Aerosol produced by biomass burning makes up about 10% (in mass) of BC, and the RF for it was estimated to be from −0.16 to −0.74 W/m^2, whereas the radiation absorbed by it varies within 0.75–2 W/m^2.

Components of the products of biomass and fossil fuel combustion responsible for light scattering mostly consist of the organic matter. These components (along with water-soluble organic and inorganic compounds, emitted into the atmosphere as components of smoke and soot compounds) can also be cloud condensation nuclei. This means that BC and OM emissions take part in the formation of the indirect RF due to the action of CCN on the formation of clouds and their properties. The available estimates show that their contribution may exceed 80% of the total indirect RF. The effect of BC can also cause local heating of the atmosphere and the accompanied reduction of the number of clouds and their liquid water content thus decreasing the albedo. With the earlier obtained estimates of the related semi-indirect RF, these estimates confirm the possibility of some additional climate warming caused by these effects.

In connection with the above-said, Penner et al.[56] have obtained new estimates of the effect of the soot and smoke aerosol on climate using the GRANTOUR/CCM global climate model that accounts for the influence of BC contained in cloud droplets on the cloud albedo. The data collected in Table 1 characterize the annual average total content of different types of aerosol, used in the model considered. Numerical simulation of the direct, semi-direct, and indirect RF with the allowance made for both short-wave and long-wave RF shows that the latter determines the reduction of the value or even alternation of the sign of the semi-direct RF, but not the enhancement of warming.

Table 1. Annual average total atmospheric content of aerosol of different types in the Northern Hemisphere (NH), Southern Hemisphere (SH), and the globe

Type of aerosol	NH	SH	Globe
Anthropogenic sulfates SO_4^{2-}	0.87	0.22	1.09
Natural sulfates SO_4^{2-}	0.45	0.42	0.86
Fossil-fuel OM	0.39	0.03	0.41
Fossil-fuel BC	0.08	0.01	0.09
Biomass-burning OM	1.28	1.24	2.52
Biomass-burning BC	0.13	0.13	0.26
OM emissions at the surface level	0.49	0.52	1.02
BC emissions at the surface level	0.05	0.06	0.11
Natural OM	0.13	0.10	0.23
Dust ($r < 1$ μm)	11.11	3.57	14.68
Sea-salt aerosol ($r < 1$ μm)	1.82	2.85	4.68

The total RF significantly depends on the altitude of aerosol emissions, because the high-altitude emissions favor the enhancement of negative long-wave RF. In addition, the emissions of absorbing aerosol at high altitudes may favor the enhancement of clouds at lower altitudes, where, as a rule, the temperature may decrease. The globally mean short-wave direct RF at the top of the atmosphere (TOA) was estimated to be 0.17 W/m^2. Within the framework of the "quasi-forcing," concept, which partly accounts the climatic feedbacks, the RF estimates are (0.28 ± 0.32) W/m^2.

If, according to the earlier results, the radiation absorption by aerosol causes a decrease of the cloud amount and compensates for the aerosol-induced cooling, then the discussed results of numerical simulation (based on the concept of quasi-forcing) suggest that the total RF (determined by both the short-wave and long-wave radiation) appears to be smaller (more negative) than earlier. Thus, the effect of smokes and soot aerosol manifests itself as cooling, if we take into account the changes in the cloudiness and temperature, occurring during the RF formation.

The calculations of short-wave RF with the allowance for "total" anthropogenic aerosol in regions, where the BC content exceeds 2 mg/m^2, gave the RF values of −3.0 and −3.1 W/m^2 at aerosol emissions caused by biomass burning near the surface and in the middle troposphere, respectively. If the emissions of soot aerosol occur near the surface, then they cause, on the average, the heating of the troposphere at all levels and the decrease of the cloud amount near the surface. The total quasi-forcing with the allowance for both short- and long-

wave radiation in the case of the soot aerosol appears to be close to zero.

The estimates obtained recently have indicated a very strong effect of the phase state of atmospheric ammonium sulfate aerosol (at the relative humidity about 80%) on the level of aerosol radiative forcing. It is also of great interest the effect of the phase state of aerosol on the heterogeneous chemical reactions. Although the processes of deliquescence (assimilation of water) and efflorescence (loss of water) by the pure ammonium sulfate are studied quite well, the main problem is the complex chemical composition of aerosol particles, including up to 50% and more (by mass) of organic compounds.

The analysis of aerosol samples collected at some sites of Western Europe has shown that about 60% of the organic carbon contained in the tropospheric aerosol are the water-soluble organic compounds. According to observations in a rural region of Austria, mono- and dicarboxylic acids make up about 11% (of the total content of organic carbon in cloud water). If the insoluble organic compounds prevent the assimilation of water by aerosol, then the soluble organics usually favors this process.

Because of the deficient information about the role of phase changes of aerosol, Brooks with co-workers[8] used a temperature controlled flow tube system equipped with Fourier transform infrared (FTIR) spectrometer for detection of particle phase and relative humidity and to measure the deliquescence and efflorescence of ammonium sulfate, maleic acid, and internally mixed maleic acid/ammonium sulfate particles. The results obtained indicated that maleic acid aerosols begin to take up water starting at a low relative humidity (*RH*) about 20%, and continue the uptake of water until the final deliquescence relative humidity of 89% is reached. Internally mixed particles containing maleic acid and ammonium sulfate were found to deliquesce at a lower relative humidity than either of the pure species. Efflorescence studies indicated that while pure maleic acid particles crystallize at about 18% *RH*, pure ammonium sulfate and all mixed aerosols effloresce at or just below 30% *RH*. Taken together, the results suggested that the presence of water-soluble organics internally mixed with ammonium sulfate aerosol could increase the range of conditions under which the aerosol is a solution.

Zender et al.[70] have developed a numerical model for predicting the size-resolved distribution of atmospheric dust for climate and chemistry-related studies. This model is to be used as a component of climate models and chemical processes in the atmosphere. The Mineral Dust Entrainment and Deposition (DEAD) model was applied to reconstruct the global dust distribution with the allowance for the transformation of aerosol properties caused by the entrainment, dry and wet deposition, and chemical reactions with the participation of dust aerosol (DA). It was assumed that the soil texture is globally uniform and is replete with substances favoring saltation.

The soil erosion was parameterized by a new physically justified geomorphic index, which is proportional to the runoff area upstream of each DA source region. Dry deposition processes included sedimentation and turbulent mixing. Nucleation scavenging and size-resolved washout in both stratiform and convective cloud types were represented. Simulations broadly agreed with station observations and satellite-inferred dust distributions.

Without invoking anthropogenic mechanisms, the DEAD model captured the seasonal migration of the transatlantic African dust plume and the spring maximum in Asian dust outflow and concentration over the Pacific. According to the estimates, the 1990s global annual mean and variability of $D < 10$ μm dust were the following: yearly emission 1490 ± 160 Tg/yr, interannual variability (burden) 17 ± 2 Tg; and optical depth at 0.63 μm, 0.030 ± 0.004. The following data characterized the contributions of different continents (Tg/yr): 980 (Africa), 415 (Asia), 37 (Australia), 35 (South America), and 8 (North America). The emission, burden, and optical depth were significantly lower than some recent estimates. The model underestimates transport and deposition of East Asian and Australian dust to some regions of the Pacific Ocean. The underestimation of long-range transport of particles larger than 3 μm contributes to this bias. The experiments supported the hypothesis that dust emission "hot spots" exist in regions where alluvial sediments have accumulated and may be disturbed.

The heavy anthropogenic emissions of large amounts of trace gases under conditions of big cities attract growing attention to this problem in the context of possible effects on the environment and on the human health (in particular, on the formation of RF). One of the most prominent examples is the Mexico City, situated at 2240 m above the sea level (~ 18°–20°N, 98°–100°W), which is surrounded by mountains, serving a barrier for the atmospheric circulation. The processes of formation of the city heat island are complicated in the Mexico City by the rugged relief. In the morning, when the cold air flows to the city down the mountain slopes, the situation of stagnant air and, correspondingly, concentrated pollution arises. After sunrise, the heating of the southwestern mountain slopes favors the income of the humid air from the Gulf of Mexico. The significant height of the Mexico City determines the decreased concentration of oxygen and favors the increase in the concentration of near-surface ozone.

Based on the Northern Aerosol Regional Climate Model (NARCM), Munoz-Alpizar with co-workers[49] calculated the transport, diffusion, deposition, and size distributions of sulfur aerosol particles in the region of Mexico City. The model assumed only simple sulfur oxidation, not taking explicitly into account the urban air chemistry. Rather, it focused on detailed aerosol microphysics and 3D optical properties. The simulation performance was compared with the upper air and ground-based observations for the following specific

days of intensive measurements: 2, 4, and 14 March, 1997. Time series at Mexico City airport showed lower values of the visibility in the morning due to a shallow boundary layer and higher values in the afternoon following the evolution of the boundary layer depth. Modeled visibility showed large dependence on the wind direction and size distribution of particles. It was found that better resolution of particle size led to better representation of coagulation processes and to more realistic size distributions, which showed a characteristic accumulation mode at 0.3 μm. As a result, the numerically simulated visibility was closer to the observed one.

In connection with the important role of sulfate aerosol (SA) in the formation of the direct and indirect effect of anthropogenic aerosol on the radiative climate forcing, particular interest has been shown in recent years to the numerical simulation of ARF. The calculated values of the globally mean direct and indirect RF vary, respectively, from −0.2 to −0.8 and from 0 to −1.5 W/m^2, that is, they are comparable with the positive RF due to the increase in the concentration of greenhouse gases. The wide uncertainty in the calculated ARF is mostly caused by the approximate character of the numerically simulated global spatiotemporal variability of the aerosol content in the atmosphere, as well as by the difficulties in the adequate consideration of the aerosol–cloud interaction.

Gong and Barrie[27] used the Canadian Aerosol Module coupled with the Canadian third generation Global Climate Model to simulate the global distributions of size-segregated sea salt and sulfate atmospheric aerosols of both anthropogenic and natural origin. A sectional model of 12 size bins was used to treat the size distribution of sea salt and sulfate, which were assumed to be internally mixed in each of the size bins. The spatial and temporal distributions predicted by the model compared reasonably well with observations, in particular, in the marine boundary layer (MBL). It was shown that sea salt particles redistribute the mass and number distributions of sulfate aerosols by serving as a quenching agent to nucleation and as an additional surface for condensation and by changing the cloud properties in the MBL. By differential simulations of the global sea salt and sulfate it was found that the presence of sea salt increases the mean diameter of sulfate aerosols by a factor of 2 over the MBL with high sea salt concentrations and reduces the global sulfate aerosol mass in the surface MBL layer by 5 to 75% depending on the sea salt distributions.

The high impacts are in the midlatitudes of both Northern and Southern Hemispheres with a minimum in the equatorial regions. In the polluted anthropogenic regions of North Pacific and Atlantic, sea salt reduces the sulfate concentration by 10 to 30%. The peak reduction of 50–75% occurs in the roaring 40s of the Southern Hemisphere in spring and fall. The impact of sea salt on the annual global mass and number loading was estimated to be 9.13% and 0.76%, respectively.

A reduction of 20–60% in the marine cloud droplet number concentrations (CDNC) was predicted because of the presence of sea salt, with greatest reduction in the roaring south 40s (40–70%) and in the midlatitude north (20–40%) where the sea salt concentrations were high. Along the equatorial regions some enhancement of total CDNC was simulated because of the presence of sea salt aerosols. It is planned for the future to estimate the effect of these changes in aerosol and cloud cover on the global climate.

The first indirect effect of aerosol on climate (Twomey effect) is explained based on the assumption that at the constant liquid water content (LWC) of clouds an increase in the number concentration of atmospheric aerosol (and, consequently, in the concentration of cloud condensation nuclei (CCN)) should lead to an increase in the concentration of cloud droplets and, thus, in the growth of cloud albedo. In this connection, Feingold[23] has performed the numerical simulation to analyze the possibility of using the extinction coefficient α, reconstructed from the data of ground-based remote sensing, for the low-cloud atmosphere as a suitable proxy for the size distribution of aerosol affecting drop formation. The adiabatic model of cloud droplets, restricted to the consideration of a thin layer of nonprecipitating stratocumulus clouds (just in this case, the assumption of adiabaticity can be considered acceptable), allows the hygroscopically caused growth of CCN to be reconstructed and the condensation of water vapor on droplets (neglecting the coagulation growth of droplets) to be taken into account. The model was used to calculate the cloud microstructure at a given microstructure of ammonium sulfate aerosol under conditions of updrafts with the rate from 20 to 300 cm/s. A unimodal aerosol microstructure N_a was approximated by the lognormal particle size distribution at $20 \le N_a \le 3000$ cm^{-3}, median radius of particles $0.03 \le r_g \le 0.1$ μm, and the parameter of the distribution width $1.3 \le \sigma \le 2.2$. Different values of the mass fraction of ammonium sulfate and the liquid water content of clouds were specified.

The investigations of the sensitivity of the cloud drop effective radius r_e to different input parameters had shown that r_e is most sensitive to LWC, a parameter often ignored in earlier indirect effect analyses. The relative importance of other parameters varies for different conditions but aerosol concentration N_a is consistently important. Updraft plays an increasingly important role under high aerosol loadings. The general analysis of the results showed that the use of aerosol extinction coefficient as a proxy for size distribution and composition tends to underestimate the magnitude of the first indirect effect. The possible systematic errors in the

extinction coefficient remain uncertain, because they depend on the varying characteristics of aerosol (thus, for example, the aerosol number concentration N_a cannot be reconstructed from remote sensing data), updrafts, and LWC.

Feczkó with co-workers[20,21] estimated the radiative forcing due to aerosol and greenhouse gases in Hungary in the early 1980s. The results obtained revealed significant changes in RF caused by ammonium sulfate and carbon dioxide during the past 20 years. If the contribution to the climate warming from the greenhouse effect of the growing CO_2 concentration increased by 60%, then the anthropogenic contribution to the climate cooling due to the sulfate aerosol decreased by 45%, that is, the joint effect of these two atmospheric constituents on the climate yielded its warming.

Different types of vegetation emit great amounts of non-methane hydrocarbon compounds (NMHC) into the atmosphere. The total level of emissions of NMHC, such as isoprene (C_5H_8), monoterpenes ($C_{10}H_{18}$), sesquiterpenes ($C_{15}H_{28}$), and oxygen-containing compounds ($C_nH_{2n-2}O$), in the growth period amounts to 825–1150 Tg C/yr, exceeding the level of the corresponding anthropogenic emissions (about 100 Tg C/yr).

As Barr with co-authors[4] noted, the importance of such emissions is mostly determined by their influence on three atmospheric processes. The first one is that such NMHC as isoprene are produced in the course of carboxylization in plants and thus contribute considerably to the biospheric carbon cycle. The second process is connected with the fact that NMHC are chemically highly active with respect to major atmospheric oxidants, such as hydroxyl radicals (OH), ozone (O_3), and nitrate radicals (NO_3). The reactions with the participation of these components yield dialkyl peroxide radicals (RO_2), which favor the efficient transformation of nitrogen monoxide (NO) into nitrogen dioxide (NO_2), which favors the increase of the ozone concentration in the atmospheric boundary layer. Finally, the oxidation of NMHC leads to the production of carbonyl compounds, such as formaldehyde (HCHO), which stimulates the processes of O_3 formation. The oxidation of monoterpenes and sesquiterpenes yields an intense formation of fine carbonaceous aerosol with the particle diameter < 0.4 μm (the production of such aerosol achieves 10–30%), which considerably affects various processes in the ABL.

Barr et al.[4] have analyzed the radiative influences produced by phytogenic aerosols over woodlands in the eastern Canada where measured aerosol concentrations resulted from α-pinene and β-pinene oxidation processes. The studied forest ecosystem produced moderate amounts of biogenic hydrocarbons, with isoprene and pinene mixing ratios reaching 1.6 ppb. Once oxidized, these pinene levels gave rise to maximum phytogenic aerosol concentrations of $5 \cdot 10^8$ cm^{-3}. The amount of short-wave solar irradiance resulting from the interaction of aerosols with incoming radiation was quantified using the measured aerosol size distributions and concentrations.

For the irradiance levels measured on clear sky days in July and with aerosol concentrations in the range from 2000 to 5000 cm^{-3}, average daytime attenuation of solar irradiance amounted to 0.04 W/m^2 with the diffuse component of 0.01 W/m^2. The maximum extinction of solar irradiance reached 0.2 W/m^2. This negative radiative influence could offset substantial fractions of the regional thermal forcing resulting from increased levels of greenhouse gases such as carbon dioxide. Phytogenic aerosol can also be of great importance as a factor affecting the optical properties of clouds serving the cloud condensation nuclei.

The global contribution of the plant cover to atmospheric emissions of volatile organic compounds (VOC) amounts to about 90%. Biogenic volatile organic compounds (BVOC) include isoprene (C_5H_8), monoterpenes ($C_{10}H_{16}$), and other chemically active carbon compounds. The total carbon content in the global BVOC emissions can exceed 1 Gg/yr. Many BVOC react with near-surface ozone and other atmospheric oxidants, affecting significantly the atmospheric chemical processes on local, regional, and global scales. The presence of BVOC, for example, increases the residence time of methane in the troposphere by about 15%. The transformation of BVOC in the global atmosphere yields 13–24 Tg/yr of secondary organic aerosol, which is comparable with the level of production of carbonaceous aerosol due to burning of biomass and fossil fuel. As was noted by Levis et al.,[40] the biogenic aerosol affects significantly the radiative budget and the formation of cloud condensation nuclei. This means that BVOC emissions have a considerable impact on the global climate through their effect on the chemical processes, concentration of atmospheric aerosol, and the global carbon cycle. It is for this reason that IPCC recommended accounting the VOC emissions in the emission scenarios used in climate models. According to the corresponding estimates, the level of anthropogenic emissions of VOC in the middle in 1999 amounted to about 140 Tg C/yr, while the predictions for 2100 vary from < 100 to > 550 Tg C/yr. The emissions of BVOC depend on the plant species and on the ambient conditions (air temperature, solar irradiance, plant water supply, concentration of O_3 and CO_2).

Levis and co-authors[40] described an algorithm developed based on field and laboratory observations, which allows one to calculate BVOC emissions. This algorithm is incorporated in CCSM (version 2.0; developed at the National Center for Atmospheric Research, Boulder, Colorado, USA), as a component of the atmosphere–ocean–sea ice and land model, aimed at integrating biogeochemical processes in this model. Two simulations were performed: 1) a land-only simulation driven with prescribed atmospheric data and satellite-derived data on vegetation and

2) a fully coupled processes in the climatic system including pre-calculated vegetation dynamics.

In both of these cases, warm and forested regions emitted more BVOC than other regions what well agrees with observations. At a preset vegetation cover, global terrestrial isoprene emissions of 507 Tg C per year compared well with other model simulations. With the dynamics of vegetation included, the BVOC emissions responded to climate changes. The interannual variability of the simulated biogenic emissions can exceed 10% of the estimated annual anthropogenic emissions provided in the IPCC emission scenarios. This determines the necessity to include BVOC emissions in the interactive global climate models.

2. IGAC, ACE-Asia, and INDOEX projects

The International Global Atmospheric Chemistry Program (IGAC) has conducted a series of Aerosol Characterization Experiments (ACE) that integrate *in situ* measurements, satellite observations, and models to reduce the uncertainty in calculations of the climate forcing due to aerosol particles. As was noted by Huebert et al.,[30] ACE-Asia, the fourth in this series of experiments, consisted of two focused components: (1) An intensive field study that sought to quantify the spatial and vertical distribution of aerosol concentrations and properties, the processes controlling their formation, evolution, and fate, and the column-integrated radiative effect of the aerosol (late March through May 2001). (2) A longer-term network of ground stations that used *in situ* and column-integrated measurements to quantify the properties of aerosols (aerosol optical thickness, etc.) in 2000–2003.

Three aircraft, two research ships, a network of lidars, and many surface sites gathered data on Asian aerosols. In addition, satellite observations were widely used. Numerical simulations of aerosol formation, transport, and transformation on the basis of chemical transport models (CTMs) were integrated into the program. The model results were used in the forecast mode during the intensive observation period to identify promising areas for airborne and shipborne observations, when the content of dust aerosol in the atmosphere was anomalously high. The testing and improvement of a wide range of aerosol models (including microphysical, radiative transfer, CTM, and global climate models) was one of the important tasks of the observations.

The main result of ACE-Asia is the various information about aerosol, illustrating the wide variability of its physical and optical properties, determined by the complex chemical composition of aerosol as a mixture of the natural dust aerosol and anthropogenic components such as black carbon, sulfates, nitrates, and organic compounds. The observations revealed the key role of aerosol in the formation of radiative forcing. Thus, for example, under clear sky conditions, the direct ARF in April achieved -30 W/m^2. The results of ACE-Asia give rise to new questions to be addressed thus dictating the need in continuation of such studies.

The Yangtze delta region of China is a key agricultural area that experiences relatively high aerosol loadings. Xu and co-authors[68] have discussed the measurements conducted with a multichannel (at the wavelengths of 380, 440, 500, 675, and 875 nm) sun photometer. The measured values were the aerosol optical depth, τ_λ, scattering coefficient, σ_{sc}, and absorption coefficient, σ_{ab}, as well as the downward photosynthetically active radiation (400–700 nm), DPAR. The measurements were conducted from October 28 to December 1 of 1999 in China (Linan, 30°17′N, 119°45′E at the background station of atmospheric monitoring). The measurements aimed at characterization of the aerosol radiative properties and estimating the direct aerosol radiative forcing in this region. The direct aerosol radiative forcing for photosynthetically active radiation (PAR) and total solar radiation (0.2–4.0 μm, TSR) at both the ground surface and at the top of the atmosphere (TOA) were estimated based on the measurements using two radiative transfer models.

The model estimate of the mean cloud-free instantaneous direct aerosol radiative forcing (at the solar zenith angle $< 70°$) is -73.51 W/m^2 for PAR ($0.2 < \tau_{500} < 1$) at the surface, what agrees with the value of -74.4 W/m^2 derived directly from the measurements of DPAR and aerosol optical depth (τ_{500}). Based on measured mean τ_{500} value of 0.61, and the estimated cloud optical depth and cloud cover of 5.0 and 50%, respectively, the 24-h mean direct aerosol radiative forcing at the surface for PAR was estimated to be approximately -11.2 W/m^2. This suggested that the amount of PAR reaching the surface over the Yangtze delta region is reduced by 16% under the direct radiative effect of aerosols.

The model results also indicated that the cloud-free 24-h average direct aerosol radiative forcing at the TOA for TSR is -30.4 W/m^2. When the presence of clouds was considered, the mean direct aerosol radiative forcing at the TOA for TSR was estimated to be approximately -12.1 W/m^2. This value is roughly an order of magnitude greater than the estimated global mean aerosol radiative forcing of -0.3 to -1.0 W/m^2 suggested by the *Intergovernmental Panel on Climate Change* [1996]. Overall, this study indicated that aerosols have a substantial impact on the amount of radiation reaching the surface as well as on the short-wave radiation balance at the TOA.

Radiative forcing of aerosols and clouds in the region of East China Sea were studied using data from surface radiation measurements, satellite remote sensing, and model simulation conducted in April 2001.[53] The estimates of the aerosol direct forcing obtained strongly depended on the method used for estimation of the aerosol properties, especially on the single scattering albedo, generating the method dependent difference about 40%.

The monthly mean whole sky radiative forcing of the direct aerosol effect was derived from various methods as −5 to −8 W/m^2 at the top of the atmosphere (TOA) and −10 to −23 W/m^2 at the Earth's surface of Gosan (33.28N, 127.17E) and Amami-Oshima (28.15N, 129.30E) sites. The corresponding mean RF values were (−5.6 ± 0.9) W/m^2 (Gosan) and (−7.5 ± 1.5) W/m^2 (Amami-Oshima) at the TOA and (−15.8 ± 6.6) W/m^2 and (−18.2 ± 5.9) W/m^2 at the surface level.

There was a large regional difference caused by changes in the aerosol optical thickness and single scattering albedo. The cloud forcing was estimated as −20 to −40 W/m^2, so that the aerosol direct forcing can be comparable with the cloud radiative forcing at surface. The indirect aerosol radiative forcing was roughly estimated from satellite method and SPRINTARS long-range transport model to be −1 to −3 W/m^2 at both TOA and surface. Since these values exceed the global mean value (about −1 W/m^2), this indicates that the latter is formed as a result of averaging of negative RF over the ocean and the positive one over the land.

A three-dimensional aerosol transport–radiation model coupled with a general circulation model, Spectral Radiation–Transport Model for Aerosol Species (SPRINTARS), simulates the spatiotemporal variability of the aerosol concentration and aerosol optical properties by taking into account the processes responsible for the formation of the global atmospheric circulation and radiative transfer. Takemura et al.[63] have performed the numerical simulation, whose results were compared with the aerosol sampling and optical observations from ground, aircraft, and satellite acquired by intensive observation campaigns over the east Asia (ACE-Asia) in spring 2001.

The analysis of results obtained has shown a good agreement of the observed and calculated temporal variations of the aerosol concentrations, optical thickness, and Angström exponent. The values of the Angström exponent suggested that the contribution of the anthropogenic aerosol, such as carbonaceous substances and sulfates, to the total optical thickness has the same order of magnitude as the Asian dust, even at the Asian dust storm events.

The radiative forcing by the aerosol direct and indirect effects was also calculated. The negative direct radiative forcing was simulated to be over −10 W/m^2 at the tropopause in the air mass during the large-scale dust storm, to which both anthropogenic aerosols and Asian dust contribute almost equivalently. The direct radiative forcing, however, largely depends on the cloud water content and the vertical profiles of aerosol and cloud. The simulation showed that not only sulfate and sea salt aerosols but also black carbon and soil dust aerosols, which absorb solar and thermal radiation, produce a strong negative radiative forcing by the direct effect at the surface, which may exceed the positive forcing by anthropogenic greenhouse gases over the east Asian region.

Measurements[12] of elemental carbon (EC) during the Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia) showed that significant amounts of EC were found in the coarse particle phase during yellow sands events. These particles come into the atmosphere with the dust storms in deserts or are of eolian origin. Coagulation during long-range transport is also consistent with this observation. The daily averaged specific mass absorption efficiencies of EC were calculated, yielding the values of 12.6 ± 2.6 and 14.8 ± 2.3 m^2/g for PM10 (particulate matter <10 μm diameter) and PM1, respectively. On a limited number of days, the absorption efficiency for the coarse particles only (PM10 − PM1) was found to be 5.7 ± 1.6 m^2/g during dusty days and 2.0 ± 1.0 m^2/g for nondust days. These measurements suggested that fine particulate EC was internally mixed and that the dust was possibly somewhat absorbing. Specific mass absorption efficiency was observed to be inversely related to the EC mass concentration. Only the air mass aging effects cannot explain this result.

If this observation holds on a global scale, it would reduce the effectiveness of a strategy for mitigating climate change by reducing EC emissions. Model simulations of idealized nonspherical dust radiative properties predict that scattering is strongly (by nearly a factor of 3) dependent on particle geometry, while absorption is a very weak function of particle geometry. The net change in absorption of short waves by polluted dust layers due to coagulation of EC with dust was predicted by model calculations to vary between −42% and +58%, depending on the assumption of the initially mixed state of the EC and on the dust optical properties. The observations, however, supported the variations to be in the range from −10 to −40%, with the value of −10% being most probable.

Actinometric surface radiation measurements[9] were conducted at Gosan, Jeju, Republic of Korea (33.29°N; 126.16°E), during the ACE-Asia field campaign aimed at studying the formation of direct ARF, especially, during the dust aerosol events caused by strong dust storms in Asia. Downwelling total, direct, and diffuse radiative fluxes were measured in the total solar spectrum as well as near-infrared and visible portions of the spectrum. Aerosol optical depth measurements at 500 nm (τ_{500}) were also made using a scanning shadow band radiometer. Surface radiative forcing values were determined under clear-sky conditions in the period from 25 March to 4 May 2001.

The averaged RF/τ_{500} values were found[9] to be −73.0 ± 9.6, −35.8 ± 5.5, and −42.2 ± 4.8 W/m^2 for the total solar, near-infrared, and visible spectral regions. A new radiative forcing parameter, the fractional forcing efficiency, defined to express the radiative forcing relative to the total energy incident on the top of the atmosphere, was also determined. The fractional diurnal forcing efficiency at Gosan during ACE-Asia was −18.0 ± 2.3, −16.2 ± 2.4, and −26.7 ± 3.3%/τ_{500} for the same band passes,

indicating that a larger percentage of the flux at visible wavelengths is radiatively forced compared to the total and near-infrared portions of the solar spectrum.

Conant et al.[17] have justified the model of vertical profiles of aerosol size, composition, and hygroscopic behavior based on observations by Center for Interdisciplinary Remotely Piloted Aircraft Studies (CIRPAS) Twin Otter and National Oceanic and Atmospheric Administration R/V *Ronald H. Brown*. The observations were obtained during the Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia) in the Asian–Pacific region. The model accounted for sulfate, black carbon, organic carbon, sea salt, and mineral dust. The effects of relative humidity and mixing assumptions (internal versus external, coating of dust by pollutants) were explicitly accounted for.

The aerosol model was integrated with a Monte Carlo radiative transfer model to compute direct radiative forcing in the short-wave solar spectrum. The predicted regional mean surface aerosol forcing efficiency (change in clear-sky short-wave radiative flux per unit aerosol optical depth at 500 nm) during the ACE-Asia intensive period was -65 W/m^2 for pure dust and -60 W/m^2 for pure pollution aerosol (clear skies). A three-dimensional atmospheric chemical transport model (Chemical Weather Forecast System (CFORS)) was used with the radiative transfer model to derive regional radiative forcing during ACE-Asia in clear and cloudy skies.

Net regional short-wave solar direct radiative forcing during April 5 to 15, 2001 dust storm was -3 W/m^2 at the top of the atmosphere and -17 W/m^2 at the surface for the region from 20°N to 50°N and 100°E to 150°E when the effects of clouds on the direct forcing were included. The model fluxes and forcing efficiencies were found to be in a good agreement with the surface radiometric observations from aboard the *R.H. Brown* research vessel. Mean cloud conditions were found to moderate the top of atmosphere (TOA) radiative forcing by a factor of three compared to clear-sky calculations, but atmospheric absorption by aerosol was not strongly affected by clouds in this study.

The regional aerosol effect at the TOA ("climate forcing") of -3 W/m^2 was comparable in magnitude, but of opposite sign, with the present-day anthropogenic greenhouse gas forcing. The main effect of aerosol on the transfer of short-wave solar radiation manifested itself as a redistribution of the solar heating from the surface to the atmosphere ("in favor" of the atmosphere). As compared with the Indian Ocean Experiment (INDOEX) findings, according to which the average RF efficiency at the surface was -72 W/m$^2 \cdot \tau_a$ (τ_a is the aerosol optical depth), the value obtained during ACE-Asia was more moderate: -60 W/m$^2 \cdot \tau_a$. At the top of the atmosphere, the values turned out different too: -25 and -27.5 W/m$^2 \cdot \tau_a$, respectively.

The main difference in the RF due to dust aerosol plumes in Eastern Asia and Southern Asia (INDOEX) manifested itself in the case of ACE-Asia in the following: 1) the effect of the dust aerosol appeared to be more significant (the anthropogenic contribution, however, remains unknown in this case); 2) the effect of upper- and medium-layer clouds was stronger (unlike the prevalence of low-layer clouds, typical of INDOEX); 3) the vertical aerosol distribution was more complex due to the influence of mid-latitudinal frontal systems. An interesting result of observations during ACE-Asia was that dust aerosol particles, combined with secondary aerosol consisting of sulfates and organic carbon, scatter the short-wave radiation more weakly than the aerosol, being an external mixture of the dust aerosol and the secondary aerosol. If, however, dust particles are covered with black carbon, then there is no marked change in absorption.

Markowicz et al.[45] presented results on the aerosol chemical composition and optical properties observed during the ACE-Asia experiment (March and April of 2001) from the NOAA *Ronald H. Brown* ship in the western Pacific Ocean (mostly, in the Sea of Japan) in order to determine the aerosol radiative forcing. The mean value of aerosol optical thickness observed during the ACE-Asia cruise in the Sea of Japan was 0.43 ± 0.25 at 500 nm, while the single-scattering albedo was 0.95 ± 0.03). A high correlation ($r^2 = 0.69$) was found between the single-scattering albedo and the relative humidity.

Aerosols in the atmosphere caused a mean decrease in the diurnally averaged solar radiation of 26.1 W/m^2 at the surface, while increasing the atmospheric solar absorption and solar radiation reflected from the top of the atmosphere by 13.4 W/m^2 and 12.7 W/m^2, respectively. The mean surface aerosol forcing efficiency (forcing per unit optical depth) over the Sea of Japan was -60 W/m^2. Decrease in the relative humidity to 55% enhanced the aerosol forcing efficiency by 6 to 10 W/m^2. This dependence on the relative humidity had implications for comparisons of aerosol forcing efficiencies between different geographical locations.

Based on the AVHRR and POLDER satellite data, Sekiguchi et al.[62] have estimated the quasi-global direct RF over oceans (-0.4 W/m^2) and indirect RF (from -0.6 to -1.2 W/m^2). These estimates indicate that indirect RF was more significant than direct RF. According to the estimate by Lesins and Lohmann,[39] the clear-sky mean global ARF is -2.2 W/m^2.

Applying the NJUADMS model of long-range transport of sulfate aerosol along with the RegCM2 regional climate model, Wang et al.[65] have obtained the average (over China) direct RF due to the sulfate aerosol to be equal to -0.85 W/m^2, whereas RF in the Central and Eastern China may achieve -7 W/m^2. The RF variability is characterized by the strong annual dependence.

Menon et al.[47] noticed that, in recent decades, there has been a tendency toward increased summer floods in south China, stronger drought in north China, and moderate cooling in China and India

while most of the world has been warmed. With the use of a global climate model, the possible aerosol contributions to these trends were investigated. It was found that precipitation and temperature changes in the model were comparable to those observed if the aerosols included a large proportion of absorbing black carbon ("soot"), similar to observed amounts. Absorbing aerosols heat the air, alter regional atmospheric stability and vertical motions, and affect the large-scale circulation and hydrological cycle with significant regional climate effects.

Summarizing the results of the ACE-Asia project, Seinfeld et al.[61] emphasized that the available estimates of the climatic effect of huge emissions of dust and anthropogenic aerosol from China and Mongolia to the Pacific Ocean (as was noted earlier, the products of these emissions may even reach North America) are still burdened by large uncertainties, although their significant impact on the radiative transfer, climate, chemical processes in the atmosphere, and cloud dynamics is beyond any doubts. The interaction of dust aerosol with atmospheric pollutants may lead to the increase in the solubility of Fe and other biogens, which may affect considerably the ocean biodynamics (it was found, for example, that almost double increase of biomass in the mixed ocean layer occurred after the income of dust storm products, which can be explained by the induced "fertilization" of the ocean).

The data in Table 2 illustrate the estimated contributions of different factors to ARF at the surface level and at the top of the atmosphere, as well as to the short-wave radiation absorbed by the atmosphere.

Table 2. Estimated contributions to ARF and short-wave radiation absorbed by the atmosphere (W/m^2) in the period of April 5–15 of 2001 in Eastern Asia (20–50°N; 100–150°E)

Factor	Surface	Rad.	TOA
Dust aerosol	–9.3	3.8	–5.5
Sulfates	–3.6	0.3	–3.3
Organic carbon	–3.9	1.7	–2.2
Black carbon	–4.1	4.5	0.4
Sea salt	–0.4	0.0	–0.4
Internal mixture	–2.2	3.5	1.3
Long-wave RF	3.0	–2.3	0.7
Total RF (clear sky)	–20.5	11.5	–9.0
Total RF (real cloudiness)	–14.0	11.0	–3.0

Myhre with co-workers[51] have performed a detailed estimation of the errors in ARF calculations due to insufficient reliability of data on the sulfate aerosol.

The main objective of the INDOEX Experiment was to study the anthropogenic aerosol coming to the Indian Ocean from Southern Asia and to estimate the effect of this aerosol on the radiative conditions of the atmosphere. Extensive and long-term multistation measurements of aerosol properties and radiative fluxes were carried out in the haze plume off the South Asian continent. These experiments have been carried out at Kaashidhoo Climate Observatory (KCO) (4.95°N, 73.5°E), Minicoy (8.5°N, 73.0°E), and Trivandrum (8.5°N, 77.0°E). In addition, the fluxes were measured at the top of the atmosphere simultaneously with the CERES radiation budget instrument.

The analysis[60] of the long-term observations (more than 15 years) over Trivandrum showed that there is a gradual increase in aerosol visible optical depth from about 0.2 in 1986 to about 0.4 in 1999. The contribution of anthropogenic sources to the aerosol optical depth exceeded 70%. Pre- and post-monsoon aerosol characteristics were examined to study seasonal variations. These variations were found to be quite significant. The impact of aerosols on short-wave radiation budget was estimated from direct observations of solar radiation using several independent ground-based and spaceborne instruments.

It was observed that "excess absorption" is not needed to model diffuse fluxes. The lower atmospheric heating due to absorbing aerosols was as high as ~ 20 W/m^2, which translates to a heating perturbation rate of ~ 0.5 K/day. The effect of aerosol mixing state (internally and externally) on aerosol forcing appeared to be negligible. A sensitivity study of the effect of aerosols over land in contrast to that over the ocean showed an enhancement in lower atmosphere heating by about 40% occurring simultaneously with the reduction of ~ 33% in surface cooling. For the ocean ARF was –10 (TOA) and –29 W/m^2 (surface), while the radiation absorbed by the atmosphere amounted to 19 W/m^2. For the land the corresponding values were equal to –7 W/m^2 (TOA) and –21 W/m^2, whereas the absorbed radiation was 28 W/m^2. Increasing sea-surface winds increase aerosol cooling due to increased sea salt aerosol concentrations, which can partly offset the heating effect due to absorbing aerosols.

From ground-based observations in Kyoto, Yabe et al.[69] have obtained the ARF values very close to those observed during INDOEX. The mean value of ARF per unit optical depth was –85.4 W/m^2. The calculations of ARF at the top of the atmosphere gave the values, which turned out to be roughly a factor of three lower than the ARF values at the surface level (this means the presence of strong absorption of the short-wave radiation by the atmosphere).

The Saharan Dust Experiment (SHADE) campaign was carried out in September 2000 over the Atlantic Ocean near the western coast of Africa in order to study the aerosol radiative forcing from the data of airborne measurements of the characteristics of the Saharan dust aerosol and the radiation fields in the short- and long-wave regions. An important part of the SHADE scientific program was reconstruction of the observations through numerical simulation based on the CTM-2 model of the long-

range transport of aerosol with the allowance for its chemical transformation, having as the input meteorological information that provided for by the European Center of Medium-Range Weather Forecasts for September of 2000.

Myhre et al.[50] have calculated ARF for the short- and long-wave spectral regions. The results of numerical simulation of the long-range transport of the dust aerosol were in a good agreement with the observations. The calculated maximum negative value of the ARF for the short-wave region was -110 W/m^2 (it took place at the local noon of August 26 of 2000). The diurnal mean ARF values during SHADE varied from -5 to -6 W/m^2, while the conditionally global average ARF is -0.4 W/m^2. The results discussed illustrate the urgency of consideration of the aerosol impact on climate. Certainly, the effect of Saharan dust should be also seen on the global scales, and it is obvious that since the short-wave radiative forcing dominates, this effect is seen as radiative cooling.

3. Some observations in Western Europe

In connection with the remaining inconsistency between the estimates of the so-called "excess" absorption (underestimation of the calculated net radiation as compared with the observed one), Wendisch et al.[66] calculated the integral and spectral fluxes of short-wave radiation at the surface level, given the vertical profiles of the aerosol microstructure and meteorological parameters (from the data of airborne, radiosonde, and lidar observations).

The calculated flux values were compared to measured ones gathered with broadband solar pyranometers and pyrheliometers, and a fixed-grating photodiode array spectroradiometer with 512 spectral channels between 500 and 920 nm wavelength. The measurements were obtained during the joint field campaign within the frameworks of Lindenberg Aerosol Characterization Experiment (LACE) 98 near Berlin/Germany in summer of 1998. A measurement-based sensitivity analysis was carried out focusing on the influence of particle composition (complex refractive index) and of microphysical and humidity growth uncertainties on the calculated surface insolations.

Assuming a spectral refractive index of ammonium sulfate for the aerosol particles, the average total solar irradiance was 11–20 W/m^2 (2–3%) greater than the measured values; the direct solar radiation was 17–28 W/m^2 (4–5%) higher, while its diffuse component was 6–7 W/m^2 (4–10%) lower than the measured value. The measured and calculated spectral surface insolations (global portion) agree well in the center of the visible region (500–600 nm wavelength). Toward longer wavelengths (near infrared), the calculated spectral surface insolations are increasingly higher than the measured ones. Thus, the results discussed did not remove completely the inadequacy of numerical simulation of the short-wave radiation transfer in the atmosphere.

To study the aerosol effect on the short-wave radiation transfer in the atmosphere, Lindenberg Aerosol Characterization Experiment (LACE 98) was carried out over the eastern part of Germany in summer 1998.[67] The main goal of this experiment was to conduct integrated ("closure") measurements of chemical, physical, optical, and radiative properties of aerosol under conditions of the clean and strongly polluted atmosphere. By coordinated flights of three aircraft, measurements of aerosol microphysical and optical properties, of spectral surface reflectance, and of upwelling and downwelling solar irradiances were performed at altitude levels between 200 m and 11 km. The measurements were used for a comparison with the results obtained using a radiative transfer model. The results for the daily averaged solar radiative forcing due to aerosol particles revealed cooling of the total Earth-atmosphere system ranging from -4.7 to -12.7 W/m^2 under cloud–free conditions. Over the tropical North Atlantic Ocean in summer, plumes of aerosol extend from Saharan Africa in the east to the Caribbean in the west. Garrett et al.[26] have studied the microphysical and radiative evolution of such plumes, formed during dust storms in Northern Sahara. The mass of aerosol and its radiative properties refer to two layers that overlay one another over much of the Atlantic Ocean. Mineral dust dominates in the lower free troposphere, and sea-salt aerosol, produced near the rough ocean surface, dominates in the maritime boundary layer. Carbonaceous, sulfate, and nitrate (CSN) aerosols are a minor component of mass but contribute significantly to total column optical depth.

Combined CSN aerosols and sea-salt contribute to more than half of the total aerosol clear-sky short-wave forcing associated with such plumes. Satellite and model data suggest that the reduction of plume forcing between the African coastline and the Caribbean is less than about 20%. The reduction is due principally to settling of large dust particles. However, the reduction of forcing remains small because (a) boundary layer trade winds provide a steady source of sea-salt, (b) dust particles that initially have been lifted up to 2.5–5.5 km from the surface and therefore have long settling distances before removal, and (c) small CSN and dust particles in the free troposphere have high specific extinction and are not involved in any significant removal processes. Measurements and climatology suggest that the CSN aerosols in the free troposphere are the anthropogenic pollution from Europe. Further investigations of these aerosols are needed to obtain the reliable data about their contribution to the radiative forcing.

The complexity of the processes of direct and indirect (through clouds) effect of aerosol on climate is the main cause of the significant uncertainty in the existing estimates of the aerosol contribution to the formation of climate.

The key aspect of this complexity is the strong spatiotemporal variability of aerosol properties. Power and Goyal[58] have performed the calculations in order to analyze the regularities of the spatiotemporal variability (including annual and interannual variations) of the atmospheric turbidity (the total column amount of aerosol) at eight stations in the South Africa and 13 stations in Germany.

According to the observations in Germany, the Angström turbidity coefficient β (equivalent to the aerosol optical depth of the atmosphere at the wavelength of 1 μm) is characterized by a strong seasonal turbidity trend of summer maxima and winter minima. In South Africa the seasonal turbidity trend is weaker, but, on average, the turbidity peaks in spring and is lowest in fall and winter. Over the past several decades, turbidity over Germany has decreased significantly (within –5.62 to –16.56% for 10 years). The impacts of the El Chichon and Pinatubo volcanic eruptions are evident in the turbidity signatures over Germany. Four stations in South Africa showed long-term trends in turbidity, but there was no consistent trend. Turbidity in Germany appeared to be more influenced by aerosols of non-local origin than in South Africa, where the aerosol load may be mostly of local origin. Long-term monthly averages of Angström turbidity coefficient β over Germany ranged between 0.019 and 0.143, and long-term annual mean values ranged between 0.064 and 0.116. Over South Africa, turbidity was noticeably lower: long-term monthly mean β ranged between 0.004 and 0.071, and the long-term annual mean β ranged between 0.013 and 0.047.

When averaged over all stations in each of the countries, turbidity was, on average, between 2.6 and 5.8 times higher in Germany than in South Africa, depending on the time of a year. The climate in South Africa is clearly more humid than in Germany, although both countries have strong seasonal trends in humidity. South Africa has become more humid over the past several decades. The decrease in turbidity at stations in Germany appeared to be due to a reduction in aerosol emissions and/or changes in climatic factors other than humidity. According to data of different observations (mostly, in Western Europe) for the past 50 years the total radiation has decreased by 0.51 W/m^2 (2.7% for 10 years), which is caused most likely by the aerosol effect.

4. Numerical simulation of the three-dimensional distribution of aerosol and climate

The results on the three-dimensional fields of the aerosol concentration and radiative properties have contributed significantly to the numerical simulation of climate formation in the context of justification of the air quality models.

The aerosol component of the Community Multiscale Air Quality (CMAQ) model, described by Binkowski and Roselle,[5] was designed to be an efficient and economical reproduction of the aerosol dynamics in the atmosphere. This approach represents the particle size distribution as a superposition of three lognormal distributions, called modes, which include the fine PM2.5 mode (particle diameter < 2.5 μm), consisting of two submodes: Aitken nuclei (diameter < 0.1 μm) and accumulation submode (diameter of 0.1–2.5 μm), as well as the coarse PM10 mode of particles with a diameter of 2.5–10 μm. The evolution of aerosol properties is described taking into account the processes of coagulation, particle growth by the addition of mass, and formation of new particles.

The component considers both PM2.5 and PM10 and includes estimates of the primary emissions of elemental and organic carbon, dust, and other species. Secondary species considered are sulfate, nitrate, ammonium, water, and secondary organics from precursors of anthropogenic and biogenic origin. Extinction of visible light by aerosols is represented by two methods: a parametric approximation to Mie extinction and an empirical approach based upon data of field measurements. The algorithms that simulate cloud interactions with aerosols are also described in Ref. 5. Results from box model and three-dimensional simulations are also presented to illustrate the results obtained.

Mebust et al.[46] have carried out preliminary analysis of the adequacy of the Models-3 Community Multiscale Air Quality (CMAQ) model by comparing the calculated and observed visibility and concentration of different aerosol components. The visibility evaluation, using National Weather Service observations from 139 airports for 11–15 July 1995, has shown that CMAQ reasonably captured the general spatial and temporal patterns of visibility degradation, including major gradients, maxima and minima.

However, CMAQ's two visibility prediction methods by Mie theory approximation and mass reconstruction, both underestimated the visibility degradation (i.e., overestimated the visibility). The normalized mean bias (NMB) and normalized mean error (NME) for the Mie calculations were –21.7%, and 25.4%, respectively. For the reconstruction simulations, these statistics were –35.5% and 36.2%, respectively. Most of the simulated values fall within a factor of two of the observations, although the correlation coefficient $r^2 = 0.25$ (Mie) and $r^2 = 0.24$ (reconstruction).

Special aerosol evaluation was carried out in Ref. 46 using the observations of sulfate, nitrate, PM2.5, PM10 and organic carbon obtained from 18 stations in June 1995. This evaluation revealed that, with the exception of sulfate (mean bias: 0.15 μg/m^3, NMB: 3.1%), the model systematically underestimated the aerosol concentration of nitrate (–0.10 μg/m^3, –33.1%), PM2.5 (–3.9 μg/m^3, –30.1%), PM10 (–5.66 μg/m^3, –29.2%) and organic carbon (–0.78 μg/m^3, –33.7%). Sulfate was simulated best by the model ($r^2 = 0.63$, mean error = 1.75 μg/m^3, NME = 36.2%), followed by PM2.5 (0.55,

5.00 μg/m^3, 38.5%), organic carbon (0.25, 0.94 μg/m^3, 40.6%), PM10 (0.13, 9.85 μg/m^3, 50.8%) and nitrate (0.01, 0.33 μg/m^3, 104.3%). Except for nitrate, 75–80% of simulated concentrations fall within a factor of two of the observations.

Black carbon (BC) aerosol absorbs sunlight that might have otherwise been reflected to space and changes the radiative heating of the atmosphere and surface. These effects may alter the dynamical and hydrological processes governing the cloud formation. In this connection, Conant et al.[16] have considered a new, microphysical, effect of BC on climate, in which solar heating within BC-containing cloud condensation nuclei (CCN) slows or prevents the activation of these CCN into cloud drops. Solar-heated BC-containing droplets have elevated temperature by fractions of a degree above the ambient, thus raising the droplet vapor pressure and inhibiting activation of the most absorptive CCN.

In this connection, a theory describing the alteration of the Kohler curve (i.e., the equilibrium vapor pressure over a droplet as a function of water uptake) as a function of CCN size and BC fraction was developed. The effect is most significant in those CCN that contain volumes of BC larger than a 500 nm diameter sphere. For the aerosol population with 10% BC mass fraction per particle, solar heating can cause a 10% reduction in the CCN concentration at 0.01% critical supersaturation. On the other hand, the effect of heating by BC absorption on CCN activation above –0.1% critical supersaturation is negligible.

Analysis of observations suggested the cooling of the global stratosphere in the past decade, at the cooling rates depended on the length of observation series and were specific for different regions of the globe. If in high latitudes of the Southern Hemisphere the cooling trend has been observed since 1980, then in the Northern Hemisphere the most significant changes took place in 1990. The commonly accepted point of view concerning the nature of the stratospheric cooling in the Southern Hemisphere is that it is mostly connected with the chemically caused ozone depletion. The mechanism of the radiation-chemical feedback is formulated, because the cooling leads to the ozone depletion. A different situation is observed in the Northern Hemisphere, where the internally caused dynamics of the climatic system also should play a significant role.

Manzini et al.[44] has assessed the sensitivity of the middle atmosphere circulation to ozone depletion and increase in greenhouse gases by performing many year simulations with a chemistry-climate model. Three simulations with fixed boundary conditions have been carried out: one simulation for the near-past (1960) and two simulations for the near-present (1990 and 2000) conditions, including changes in greenhouse gases, in total organic chlorine, and in average sea surface temperatures. Changes in ozone were simulated interactively by the coupled model.

It was found that in the stratosphere, ozone decreases, and that in the Antarctic, the ozone hole develops in both the 1990 and the 2000 simulations but not in the 1960 simulation, as observed. The simulated temperature decreases in the stratosphere and mesosphere from the near past to the present, with the largest changes at the stratopause and at the South Pole in the lower stratosphere well agree with the current knowledge of temperature trends. In the Arctic lower stratosphere, a cooling in March (with respect to year 1960 simulation) was found only in the simulation for year 2000.

Wave activity emerging from the troposphere was found to be comparable in winters of 1960 and 2000 simulations, suggesting that ozone depletion and greenhouse gases increase contribute to the 2000–1960 March cooling in the Arctic lower stratosphere. These results therefore provide support to the interpretation that the extreme low temperatures observed in March in the last decade can arise from radiative and chemical processes, although other factors cannot be ruled out.

The comparison of the 1960 and 2000 simulations showed an increase in downwelling in the mesosphere at the time of cooling in the lower stratosphere (in March in the Arctic; in October in the Antarctic). The mesospheric increase in downwelling can be explained as the response of the gravity waves to higher winds associated with the cooling in the lower stratosphere. Planetary waves appear to contribute to the downward shift of the increased downwelling, with a delay about one month. The increase in dynamical heating associated with the increased downwelling may limit the cooling and strengthening of the lower stratospheric polar vortex from above, facilitating ozone recovery and providing a negative dynamical feedback. In both the Arctic and Antarctic the cooling due to depletion of the ozone was found to affect the area covered with polar stratospheric clouds in spring, which is substantially increased from the 1960 to the 2000 simulations. In turn, increased amounts of polar stratospheric clouds can facilitate further ozone depletion in the 2000 simulation.

Using the global climate model National Center for Atmospheric Research Community Climate Model Version 3 (NCAR CCM3), Kristjánsson[38] has obtained new estimates of the indirect effect of the sulfate and black carbon aerosol on climate, caused by the aerosol effect on the cloud dynamics. Two manifestations of the aerosol effect on clouds were considered. One of them (the first indirect effect or the radius effect) is connected with the fact that the appearance of additional aerosol particles as cloud condensation nuclei leads to the decrease in the size of cloud droplets. The second indirect effect manifests itself as a suppression of droplet coagulation, caused by the decrease of the droplet size, and, consequently, the increase of the cloud lifetime (lifetime effect). Both these effects increase the cloud albedo.

The global concentrations and horizontal distributions of the aerosol were reconstructed in Ref. 38 from simulations with a life-cycle model

incorporated into the global climate model. They were then combined with size-segregated background aerosols. The aerosol size distributions were subjected to condensation, coagulation, and humidity swelling. By making assumptions on supersaturation, cloud droplet number concentrations in water clouds were determined. Cloud droplet sizes and short-wave radiative fluxes at the top of the atmosphere (TOA) were in good agreement with satellite observations.

Using aerosol data for 2000 from the Intergovernmental Panel on Climate Change (IPCC-2001), globally averaged, a 5.3% decrease in cloud droplet radius (by 0.58 μm at the average droplet radius of 10.31 μm) and a 4.9% increase in cloud water path due to anthropogenic aerosols were found. The largest changes were found over SE Asia (where the content of the sulfate aerosol is maximum, while the zenith distance of the Sun is minimum), followed by the North Atlantic, Europe, Siberia, and the eastern United States. This is also the case for the radiative forcing ("indirect effect"), which has a global average of -1.8 W/m^2, and the contributions of the radius effect and the lifetime effect were respectively, -1.3 and -0.46 W/m^2.

When the experiment was repeated using data for 2100 from the IPCC A2 scenario, an unchanged globally averaged radiative forcing was found, but the horizontal distribution has been shifted toward the tropics. Sensitivity experiments showed that the radius effect is about three times as important as the lifetime effect and that black carbon contributes only marginally to the overall indirect effect. The approximate character of the estimates obtained is determined, first of all, by the neglect of some types of aerosol, for example, the organic carbonaceous aerosol (this is connected with the lack of the information needed), as well as by the exclusion of ice clouds and the effect of the considered processes on the long-wave radiation transfer from the consideration. In the future, it is planned to complicate the climate model by taking into account the ocean–atmosphere interaction and pay more attention to regional effects.

Cook and Highwood[18] have simulated, for the first time, the effect of aerosol absorbing solar radiation on climate with the Reading Intermediate General-Circulation Model (IGCM). The 22-layer general-circulation model of the system "atmosphere–mixed 2-m deep ocean layer" was realized on a 5° latitude × 5° longitude grid. It provides for the reconstruction of the equilibrium climate after integration over the term about 5 years (actually, the calculations were performed for 30 years). The results obtained in Ref. 18 have shown that the sign of the direct aerosol radiative forcing is not a good indication of the sign of the resulting global and annual mean surface temperature change.

A related important circumstance is that the climate sensitivity parameter for aerosols which absorb solar radiation is much larger than that for greenhouse warming due to CO_2. The reason is that significant changes in cloud amount occur, some of which appear most influenced by the change in surface temperature. A reduction in low cloud amount occurs when the aerosol single-scattering albedo is less than 0.95, which determines the corresponding positive feedback. On the other hand, the changes in the amount of upper-level clouds manifest themselves as a process of formation of the negative feedback.

The total effect of clouds is caused by the balance between two effects having opposite signs and strongly depends on the scheme of parameterization of the cloud dynamics. The so-called "semi-direct" effect of aerosols is clearly seen in this model. In this connection, it was concluded in Ref. 18 that this aerosol–cloud feedback is implicitly present in all GCMs which include absorbing tropospheric aerosol but remains largely undiagnosed. This situation suggests that adequate parameterization of the cloud dynamics (in particular, in the context of the hypothesis of semi-direct ARF[28]) is of critical importance. According to the IGCM model, clouds play an extremely important role in formation of the reaction of a climatic system to the dynamics of atmospheric aerosol.

The idea published by Jacobson[32] that the control of atmospheric emissions of black carbon and organic aerosols, being products of fossil fuel burning, can be an efficient method for slowing global warming problem, gave rise to a wide discussion. In this connection, Feichter et al.[22] noticed that, according to the Jacobson's estimates, the atmospheric emissions of black carbon (BC) and organic carbon (OC) from the fossil fuel burning can cause the increase in the global mean surface air temperature by 0.35 K. Some doubts, however, were expressed concerning the justification of conclusions drawn in Ref. 32. They were connected, in particular, with the fact that these emissions contain also the sulfur dioxide (which determines the formation of sulfate aerosol) and affect both the absorption and the scattering of the short-wave radiation, that is, may give rise to both warming and cooling. Even more important circumstance is that the GATOR-GCMM model used by Jacobson did not pass tests for adequacy through the comparison with other models and with observations, which is necessary, especially because the calculations for 6 years cannot provide for the reconstruction of the equilibrium climate. A number of objections were raised by Penner in Ref. 55.

Chock et al.[11] remarked upon the Jacobson's results,[32] concerning the adequacy of the climate model used to obtain the discussed estimates, as well as the reliability of the obtained information, and the justification of the needed environmental policy. As to the input information, it was accepted that the annual emissions of black carbon and organic carbon are 6.4 and 10.1 Tg C/yr, respectively, and the fraction of global emissions of submicron black carbon and organic carbon aerosol is, respectively, 5.1 and 7.0 Tg C/yr. The OM/BC ratio was taken to be 3.1:1, which is equivalent to the global

emissions of OM and OM submicron aerosol, achieving 19.9 and 15.9 Tg C/yr, respectively, that is, much higher than the estimates mentioned above. In addition, disadvantages such as incomplete consideration of the indirect aerosol radiative forcing, which leads to climate cooling, and the unacceptable simplification of the model, were mentioned. Replying to these and other remarks, Jacobson[33–35] rejected them as incorrect and unjustified.

Concerning this discussion, it should be noted that the most important conclusion from the above overview is the high degree of uncertainty of all the obtained estimates of the aerosol effect on global climate.

Conclusions

In connection with the Symposium "OUR HAZY ATMOSPHERE: AEROSOLS AND CLIMATE," which took place on February 14 of 2004 in the University of California, San Diego, (http://scrippsnews.ucsd.edu/article_detail.cfm?article_num=620), it was fairly noted: "it's likely that scientists will look back at the early part of the 21st century and regard it as a fundamental stage in understanding the importance of the effects of aerosols on the Earth's climate." This phrase is, certainly, only a metaphor. There are no doubts, however, that the effect of natural and anthropogenic aerosol on climate is significant, but the available quantitative estimates should be considered only as tentative. It is also obvious that we deal with the problem of extreme complexity, which requires the study of physical, chemical, and biological processes on exceptionally wide spatial (from nano- to global) and temporal scales. Assessing the significance of this problem, we can draw two conclusions: 1) the problem "aerosol–cloud–climate" should occupy the key place in the course of preparation of the IPCC Fourth Report; 2) the analysis of the results, obtained in the course of this preparation, should lead to formulation and justification of a new interdisciplinary program, which should provide for the adequate understanding of the climatic system dynamics in the context of the aerosol–cloud–climate problem. Meanwhile, it is certainly necessary to continue the investigations aimed at more complete and reliable observations and further development of numerical simulation.[1–3]

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