From nano- to global scales: properties, processes of formation, and aftereffects of atmospheric aerosol impacts. 4. Interactions of aerosol and clouds

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The interactions between atmospheric aerosol and clouds are reviewed.

Introduction

It is well known that the dynamics of cloud cover is one of the key climate-forming factors. Nevertheless, it is also well established (though calling for more reliable quantitative validation) that cloud condensation nuclei (CCN), that is, atmospheric aerosol particles, are of critical importance in formation of the cloud cover. In addition, clouds strongly affect the content and properties of aerosol. All these aspects determine acute urgency of the problem of aerosol–cloud interaction, which involves formation of various feedbacks.[1,18,19] Therefore, the Graf's sentence that the interaction between aerosols and clouds is one of the strongest factors of climate change, but there are significant uncertainties in our current understanding of the corresponding processes,[9] seems to be quite natural.

"Anthropogenic experiments," consisting in the every-year large-scale biomass burning in tropical regions favor the study of aerosol–clouds interactions. The consequences of these "experiments" still attract considerable attention. Thus, for example, Koren et al.[20] studied the reduction of development of clouds formed below the top of the atmospheric boundary layer (ABL) under the effect of air pollution and smoke from biomass burning during the dry season, and Andreae with colleagues[2] discovered the "smoking" clouds, which appear due to emitted smoke aerosol in the period between dry and wet seasons in the Amazon region (Brazil).

Koren with co-authors[20] has shown that the main effect of the aerosol, absorbing and scattering radiation, is a decrease in water amount coming from the forest canopy to clouds. The decrease is caused by the aerosol-induced cooling of the surface and stabilization (due to warming) of the turbulent ABL. Satellite data showed that scattered cumulus cloud cover was reduced from 38% in clean conditions to 0% for heavy smoke (optical depth of 1.3). This response to the smoke radiative effect reverses the regional smoke instantaneous forcing of climate from -28 W/m^2 in cloud-free conditions to $+8$ W/m^2 once the reduction of cloud cover is accounted for.

According to the data of Andreae et al.,[2] the smoke aerosol reduces cloud droplet size and so delays the onset of precipitation from 1.5 km above cloud base in pristine clouds to more than 5 km in polluted clouds and more than 7 km in pyro-clouds. The suppression of low-level rainout and aerosol washout allows transport of water and smoke to upper levels, where the "smoking" clouds appear as they detrain much of the pollution. Elevating the onset of precipitation allows invigoration of the updrafts, causing intense thunderstorms, large hail, and a greater likelihood for overshooting cloud tops into the stratosphere. There, detrained pollutants and water vapor would have profound radiative impacts on the climate system. The invigorated storms release the latent heat higher in the atmosphere. This should substantially affect the regional and global circulation systems.

Graf[9] emphasized fairly that though the results mentioned above correspond to the Amazon region, their methodological significance is mush wider. The influence of aerosol on the cloud dynamics can manifest itself in different ways under different conditions. The cloud–aerosol interaction is a nonlinear process and can lead to bifurcation of "trajectories" of the cloud cover development. The specificity of this interaction is determined by the existence of certain critical values of the parameters governing this interaction. Nano-scale changes occurring in individual aerosol particles lead to changes in the cloud dynamics, regional cloud fields, and large-scale energy exchange in the atmosphere. This means, in particular, that the adequate reconstruction of the processes in ABL is possible only when taking into account the influence of aerosol on the radiative transfer, turbulent mixing, evacotranspiration, and plant cover, as well as the corresponding microphysical and chemical processes.

Having mentioned some newest results, let us address ourselves to consideration of the problem in more detail.

1. Empirical diagnostics of the aerosol–cloud interaction

Although there are strong grounds to believe that the sulfate aerosol exerts a considerable influence

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upon formation of global climate, the corresponding quantitative estimates are uncertain, to a high extent, as yet. This influence is direct, showing itself in the aerosol backscattering of short-wave sunlight, whose global average estimates vary from −0.2 to −0.9 W/m^2. Indirect influences of aerosol, leading to changes in the optical properties and lifetime of clouds (so-called the first and the second indirect effects), are potentially more significant (and much more uncertain).

As to the first indirect effect, its estimates range from 0 to −2 W/m^2, while those for the second one are too uncertain to be discussed on the quantitative basis. Clouds take part in the processes of both production and removal of sulfate aerosol (SA), and sulfates are produced almost exclusively through the SO_2 oxidation both in gas and in aqueous phases (in the latter case, in cloud droplets). The gas-phase production of sulfates below clouds is suppressed by such an oxidant as OH hydroxyl. Sulfates, which are well soluble in water, are removed from atmosphere mostly through precipitation scavenging (wet deposition) and, to a smaller degree, through dry sedimentation. The processes mentioned above are poorly studied yet, and, in particular, the following questions are still unanswered: 1) whether the processes in clouds favor the intensification of sulfate production or, to the contrary, scavenging of sulfate aerosol from the atmosphere; 2) whether clouds restrict significantly the process of gas-phase production of sulfate aerosol.

To analyze the relation between the cloud aqueous-phase production of sulfate, precipitation scavenging of sulfate, and inhibition of gas-phase sulfate production beneath clouds, Koch with co-authors[17] considered a correlation between clouds and sulfate aerosol. Statistical analysis of the observed daily cloud cover and sulfate surface concentrations in Europe and North America indicated a significant negative correlation between clouds and sulfate aerosol. This implies that clouds remove sulfate via precipitation scavenging and/or inhibit sulfate gas-phase production more than they enhance sulfate concentration through aqueous-phase production. Persistent sulfate/cloud anticorrelations at long timescales (8–64 days) apparently result from large-scale dynamical influences on clouds, which in turn impact sulfate.

At the same time, statistical analysis based on the general circulation model (GCM) of the Goddard Institute for Space Studies (GISS) shows a weak coherence between sulfate aerosol and cloud cover. However, there is a stronger anticorrelation between the model's sulfate aerosol generated by gas-phase oxidation and the cloud cover. The sulfate/cloud anticorrelation in the GCM strengthens if the gas-phase sulfate production beneath clouds is extinguished, as should happen under photochemical generation of OH. The only way to achieve a strong anticorrelation between total sulfate and clouds is correcting the process of aqueous-phase sulfate production in the above model.

This model, like many other global tracer models, reproduces the released dissolved species (including sulfate) from clouds at each time step rather than describes their appearance upon cloud evaporation. After corresponding correction of the GISS model, it turns out that more sulfate is rained out, the sulfate burden produced via the aqueous phase decreases to half its former amount, and the total sulfate burden and the sulfate lifetime are 25% lower (0.54 TgS and 4.2 days, respectively). In general, the sulfur circulation model should provide for more significant sulfate production under clear sky conditions and less significant under cloudy conditions, which should have a considerable effect on the radiative forcing (RF). The correction mentioned above leads to a small decrease in the indirect (anthropogenic) RF from −1.7 to −1.5 W/m^2. The change in the direct anthropogenic RF is proportional to the decrease in the sulfate aerosol production: from −0.66 to −0.47 W/m^2. As to the second indirect effect, the positive, rather than negative correlation is probable in this case. Thus, the results considered are indicative of the advisability of accounting for the negative aerosol/cloud correlation in the validation of the sulfate aerosol global model.

Using the AVHRR and POLDER satellite remote sensing data, Sekiguchi with co-authors[32] have analyzed the correlations in order to evaluate the radiative forcing of the aerosol indirect effect. The consideration of the global statistics have shown that the effective particle radius r_e and the optical thickness of low clouds τ_c correlate well with the column number concentration of the aerosol particles, indicating the aerosol indirect effect on the radiative forcing and, consequently, on climate. A correlation between the cloud fraction and the aerosol number concentration was also seen, whereas the attempts to find a significant correlation of the cloud-top temperature with the column aerosol number failed.

The regional statistics indicate that, as a rule, there is a positive correlation of τ_c and cloud fraction with the aerosol column number concentration, that is consistent with the global mean statistics. However, the effective cloud particle radius r_e shows a tendency similar to the global correlation only around the seashore regions. Using these correlations and assuming that the aerosol column number concentration has increased by 30% from the preindustrial era, the evaluated total radiative forcing of the aerosol indirect effect is about −0.6 to −1.2 W/m^2. The radiative forcing of the aerosol direct effect over the ocean from the satellite-retrieved parameters is −0.4 W/m^2.

Thus, the indirect aerosol effect on RF significantly exceeds the direct one, though we should keep in mind a considerable uncertainty in the estimates of the indirect effect. The cloud-top temperature was found to be insensitive to the change in the aerosol number concentration, although there was a distinct negative correlation between the aerosol number concentration and the cloud temperature, at

which the cloud particle radius grows to 14 μm. This particular dependence of the cloud temperature suggests that aerosols act on clouds so that the cloud near-top particle size, optical thickness, and amount change, but the cloud-top temperature keeps without causing a significant longwave radiative forcing.

Using the airborne data collected in 1997 during the second Aerosol Characterization Experiment (ACE-2) performed within the framework of the CLOUDYCOLUMN experiment, conducted over the eastern Atlantic Ocean, in the vicinity of the Canary Islands, Guibert et al.[10] obtained the estimates of the aerosol indirect effect (AIE) on climate via the aerosol-induced changes in the cloud properties. The value of the cloud-induced radiative forcing serves a criterion of AIE. For this purpose, the aircraft and surface measurements, including observations of condensation nuclei (CN), aerosol accumulation mode, and aerosol size spectra, were compared. Measured and predicted wet aerosol size spectra were also compared with the statistics of vertical velocity within and below stratocumulus clouds.

In general, the aircraft and surface measurements agreed within experimental uncertainties. However, a substantial disparity was documented in the comparison of predicted and observed wet aerosol spectra. This disparity can be attributed to either bias in the wet aerosol measurements, made with the FSSP-300 particle counter, or to errors in the sizing of dry aerosol particles. The analysis of vertical velocity indicates that the first and the third moments of the vertical velocity frequency distribution do not change substantially between below-cloud and in-cloud flight segments; however, some increase in the second moment of the distribution across cloud base is documented. Overall, the obtained results lend confidence to the use of surface data on aerosol physical and chemical properties, as well as airborne measurements of vertical velocity for modeling the aerosol activation process.

Airborne measurements of CCN were made in the vicinity of southwest Florida (USA) as a part of the Cirrus Regional Study of Tropical Anvils and Cirrus Layers-Florida Area Cirrus Experiment (CRYSTAL-FACE) field campaign in July, 2002.[34] Two carefully calibrated CCN counters measured the CCN number concentration at two supersaturation levels S of 0.2 and 0.85%. The measurements indicate that the aerosol sampled during the campaign was predominantly marine in character. The mean CCN concentrations were 233 cm^{-3} (at S = 0.2%) and 371 cm^{-3} (at S = 0.85%).

Three flights during the experiment differed from the general trend: the aerosol sampled during two flights on 18 July was more continental in character, and the observations on 28 July indicated a high spatial variability and periods of very high aerosol concentrations. The simplified aerosol/CCN closure analysis, as well as the comparison with the CCN concentrations predicted using the Kohler theory (with an idealized composition of pure ammonium sulfate) demonstrated a good general agreement between the predicted and observed CCN concentrations: at S = = 0.2%, $N_p/N_o = 1.047$ (the correlation coefficient r^2 = = 0.911); at S = 0.85%, $N_p/N_o = 1.201$ ($r^2 = 0.835$). Removing the data of the 28 July flight improved the result of the closure analysis: at S = 0.85% the ratio $N_p/N_o = 1.085$ ($r^2 = 0.770$).

The analysis of satellite observations suggest that high levels of urban and industrial atmospheric pollution, as well as concentrations of the desert dust aerosol and biomass burning smoke aerosol usually lead to decrease in size of cloud droplets and, thus, to the increase in the cloud albedo and the reduction of the cloud formation efficiency. To the contrary, large salt particles observed in the land atmosphere and sea-salt aerosol favor the growth of cloud droplets. In addition, aerosol polluting the atmosphere affects considerably the processes of cloud and precipitation formation. The results of numerical simulation have demonstrated, for example, that the black carbon aerosol, observed in China, very strongly affect the formation of precipitation on the regional scale, because of its intensified absorption of sunlight. The similar situation was also observed in the Mediterranean region.

In this context, Rudich et al.[31] studied the effects of smoke from the Kuwait oil fires in 1991 on clouds and precipitation. For this purpose, the microphysical properties of the smoke-affected and smoke-free clouds were compared using the data of satellite observations. The comparison revealed several effects: (1) clouds typically develop at the smoke plume top, probably because of solar heating and induced convection by strongly absorbing aerosols; (2) large salt particles resulted from fire smokes form giant CCNs, which initiated coalescence in the highly polluted clouds; (3) far away from the smoke source, the giant CCN were deposited, and the extremely high concentrations of medium and small CCNs dominated in the cloud development due to strong suppress of the drop coalescence and growth with altitude; and (4) small cloud droplets in the smoke-affected clouds froze at colder temperatures and suppressed both the water and ice precipitation forming processes.

These observations imply that the smoke particles above land are not washed out efficiently and can be transported to long distances, extending the observed effects to large areas. The absorption of solar radiation by the smoke induces convection above the smoke plumes and consequently leads to formation of clouds above the plume. This process dominates over the semidirect effect of cloud evaporation due to the smoke-induced enhanced solar heating.

A great amount of Saharan dust (mineral) aerosol about 200 Mt/year, which makes up 20–30% of the total aerosol mass, experiences the long-range transport in Atlantic tropics in the Northern Hemisphere. That is why, according to observations at the Barbados Islands, the concentration of the dust aerosol (DA)

appears to be 10–100 times higher in summer (in the period of the most intense long-range transport) than in winter. However, the DA concentrations at the Sal Island (northeastern Pacific) proved to be higher in winter than in summer. This contrast reflects a specificity of the global atmospheric circulation, caused by the displacement of the Azores anticyclone and the intertropical convergence zone (ICZ). The most significant aspect of this specificity is that the DA transport occurs in winter mostly at the altitudes lower than 1 km (in the layer of trade-wind inversion) and in summer in the 3–5 km layer (within the Saharan dust layer, lying above the layer of trade-wind inversion).

The earlier estimates of the DA effect on the radiative forcing suggest it to be relatively weak as compared to contribution of the anthropogenic sulfate aerosol, because the DA fraction in the aerosol optical depth is about 17%, while that of sulfate aerosol is 20%. However, the situation can change considerably, if: 1) the albedo of the underlying reflecting surface (for example, clouds) is high or 2) DA mixes with clouds.

In this connection, Pradelle et al.[28] discussed the results of Meteosat satellite observation of the outgoing radiation in the visible and IR spectral ranges for 6 years. These observations revealed a decrease in the apparent cloud albedo at any season, particularly, over the areas with high dust content (and, consequently, an increase of the aerosol optical depth). In spring, in the period of most intense emissions of Saharan dust, the decrease in the albedo can exceed 20%. In summer, the presence of DA can lead to a change in reflectance of the surface–cloud–atmosphere system due to a more intense absorption of solar radiation. The winter decrease in the cloud albedo can be partly explained by desiccation of atmosphere due to the air masses responsible for the long-range transport of the Saharan dust. Along with this, an important role can be played by mixing DA with clouds, occurring in winter. The mixing leads to changes in the microstructure of cloud droplets, affecting the cloud albedo (DA emissions always stipulate the reduction of the cloud albedo in the tropical Atlantic in the Northern Hemisphere), and, consequently, it can be considered as a significant climate-forming factor.

Organic films on deliquescent aerosols and cloud droplets lower the water surface tension and may inhibit the exchange of water vapor and gases between the gas and the liquid phase, with important implications for aerosol and cloud microphysics and heterogeneous chemistry, characterizing the interaction between aerosol and clouds. On the basis of the rheological properties of the surface films, Decesari et al.[4] studied the solubility properties of surfactants in aerosol and fog/cloud water samples. The variations of the surface tension induced by the fast expansion/compression of the films were measured by means of a drop shape tensiometer and were linked to the capacity of surfactants to exchange between the surface layer and bulk solution, and ultimately to their water-solubility. The results obtained in Ref. 4 are in good agreement with properties of standards of soluble surfactants and can be interpreted by the theory of formation of hydrophilic adsorption layers. These findings suggest that the water-soluble organic compounds (WSOC) are the main contributors to formation of films on cloud/fog droplets. The surface coverage of film-forming compounds is mainly controlled by the bulk concentration of WSOC, regardless of the available surface area. It can be concluded also that the surface tension decrease observed under laboratory conditions actually occurs in the atmosphere.

An enhancement of cloud droplet number concentration due to anthropogenic aerosols has been demonstrated previously by *in situ* measurements, but there remained a large uncertainty in the resultant enhancement of the cloud optical depth (τ_c) and reflectivity. Detection of this effect is significantly hampered by the large inherent variability in cloud liquid water path (LWP); the dominant influence of LWP on τ_c and albedo masks any aerosol influences. To determine the dependence of τ_c on LWP, Kim et al.[16] used ground-based remote sensing of τ_c by narrowband radiometry and LWP by microwave radiometry, which was limited to complete overcast conditions with single layer clouds, as determined mainly by the millimeter wave cloud radar. Measurements were conducted in north central Oklahoma for 13 different days in 2000, showing wide variation in LWP and τ_c any given day, but with nearly linear proportionality between the two quantities; variance in LWP accounts as much as 97% of the variance in τ_c a day and for about 63% of the variance in τ_c for the whole data set. The slope of τ_c vs. LWP was inversely proportional to the effective radius of cloud droplets (r_e), which ranged from 5.6 ± 0.1 to 12.3 ± 0.6 μm. This effective radius was negatively correlated with aerosol light scattering coefficient at the surface; the weak correlation ($r^2 = 0.24$) might be due, in part, to the vertically decoupled structure of aerosol particle concentration and possible meteorological influence such as vertical wind shear. Cloud albedo and radiative forcing for a given LWP were found to be highly sensitive to the effective droplet radius; for a solar zenith angle of 60° and typical LWP of 100 g/m^2, as r_e decreased from 10.2 to 5.8 μm on different days, the resultant decrease in calculated net shortwave irradiance at the top of the atmosphere (Twomey forcing) was about 50 W/m^2.

2. Numerical simulation of the aerosol–cloud interaction

Based upon recent data of European Center for Medium-Range Weather Forecasts, Pradelle et al.[27] estimated the effect of the DA layer, formed above stratocumulus clouds due to the long-range transport

of dust emitted during dust storm in Sahara, on the albedo of the ocean–atmosphere–cloud system. The calculations have demonstrated a decrease in the system albedo reaching about 15% on average, i.e., comparable to that observed by satellite in summer. In winter, when dust is concentrated in the trade-wind layer, at the same level as the cloud cover, a simple direct effect (superposition effect) cannot explain the observed albedo decrease. The agreement with observations is provided only with allowance for the complicated interaction of DA with cloud aerosol (cloud droplets) and with some trace gases.

The "processing" of dust aerosol in clouds results in a significant change of their properties. Numerical simulations revealed that dust presence in clouds may lead to a decrease in the initial cloud condensation nucleus number, an increase of the effective droplet radius, and eventually, a reduction of the cloud albedo. The albedo decrease may reach more than 10% under certain conditions, but, as a rule, the albedo reduction varies between 3% and 10%.

Simulations performed with a mesoscale model revealed a presence, at some altitude, of carbonaceous particles emitted by the African savanna burning in winter. Dust transformation associated with the presence of carbonaceous particles would decrease the system albedo by more than 10% and so would explain the satellite observations during the winter season in the southern part of the area under study. These results confirm the assumption that dust particles play an important climatic role to the west of the African coast. According to the estimates obtained, the radiative forcing associated with the dust in the presence of clouds is (1) globally always positive along the atmospheric column and (2) located either near the surface (winter case) or at some altitude (summer case). In summer, the radiative forcing remains negative near the surface but is overcompensated by positive RF contribution at the dust level.

Yin et al.[35] numerically investigated the effect of mineral dust particles, functioning as CCN, on subsequent development of cloud and precipitation, as well as on optical properties of clouds. For this purpose, a two-dimensional (2D) nonhydrostatic cloud model with detailed microphysics was used. The initial aerosol spectra used in the 2D model consisted of both background CCN and mineral dust particles.

The results obtained show that insoluble mineral dust particles become after crossing a convective cloud the effective CCN. Their effectiveness as CCN increases because of a sulfate layer that is formed on their surface as they are first captured by growing drops or ice crystals and then the sulfate is released as these hydrometeors evaporate. Upon entering clouds, these particles increase the concentration of the activated drops and widen the drop size distribution.

The calculations show that the effect of cloud-processed dust particles in continental clouds is in acceleration of formation of precipitation particles, although the amount of precipitation depends on the concentration of the large and giant CCN. In maritime clouds, the addition of the cloud-processed aerosol and mineral dust particles has a minimal effect on precipitation, because the cloud formation starts with many already existing large particles. The addition of more CCN to either maritime or continental clouds increases their optical depth, even in the cases when the precipitation amount is increased.

Due to intense biomass burning in the Amazon Basin, an inhomogeneous layer of smoke aerosol is formed in the atmosphere, which covers the area of millions km^2 during the dry season (June–December). The complex interaction of water vapor, CCN, and radiative forcing due to absorption of shortwave radiation by black aerosol strongly affects the processes of convection and cloud formation, as well as the cloud properties. Earlier measurements carried out for dry season revealed a stable spatial distribution of water vapor, but a variable distribution of the aerosol concentration. The latter implies that the optical properties of clouds in the Amazon Basin less depend on the weather conditions, than on the distribution of the CCN concentration, which strongly increases under the effect of smoke aerosol.

To assess the impact of aerosol on optical properties of clouds, Roberts et al.[30] used a one-dimensional (1D) cloud model, which allowed analyzing the dependence of the droplet growth conditions on physical and chemical properties of aerosol. CCN concentrations were measured at supersaturation values between 0.15% and 1.5%. During the wet season they were low and resembled concentrations more typical of marine conditions. During the dry season, smoke aerosol from biomass burning dramatically increased CCN concentrations. The modification of cloud properties, such as cloud droplet effective radius and maximum supersaturation, was most sensitive at low CCN concentrations. Hence, a larger interannual variation of cloud properties could be sooner expected during the wet season.

The observations have shown that differences between CCN spectra in forested and deforested regions during the wet season are modest and cause insignificant modifications of cloud properties as compared to those between wet and dry seasons. The results obtained suggest that the differences in surface albedo, rather than cloud albedo, between forested and deforested regions may dominate the impact of deforestation on the hydrological cycle and convective activity during the wet season. During the dry season, cloud droplet concentrations may increase by up to seven times, which leads to a model-predicted decrease in the cloud effective radius by a factor of two. This could imply a maximum indirect radiative forcing due to aerosol as high as -27 W/m^2 for nonabsorbing clouds.

Light-absorbing substances in smoke aerosol darken the Amazonian clouds and reduce the net radiative forcing. The approximate estimates suggest

that absorption of sunlight due to smoke aerosol may compensate for about half of the maximum indirect aerosol RF. However, further investigations are necessary, because even minor differences in chemical and physical properties of aerosol are particularly influential in the kinetics of aerosol and its activity as CCN. Knowledge of the CCN spectrum alone is not sufficient to assess reliably the climatic influence of smoke aerosol.

In the 1950s and later, different numerical models of individual clouds and cloud systems, accounting for the cloud dynamics depending on the microstructure and chemical reactions proceeding in clouds, were developed. For faster computation, all these models used various assumptions, restricting their generality. Jacobson[14] opened a new stage in numerical simulation, whose goal was, in particular, to reconstruct the cloud and precipitation dynamics taking into account, on the one hand, the various aerosol microstructure and, on the other hand, the description of the effect of the cloud and precipitation dynamics on the aerosol scavenging from the atmosphere.

The new model accounts for (1) the effect of simultaneous liquid and ice growth onto multiple aerosol size distributions, (2) diffusiophoretic, thermophoretic, gravitational, as well as coagulation processes proceeding among liquid, ice, hail particles, and their aerosol components, (3) contact freezing (CF) of drops stipulated by inter-cloud aerosols, (4) heterogeneous and homogeneous freezing, (5) liquid drop breakup, (6) coagulation of cloud drops and ice crystals (with incorporated aerosols) with inter-cloud aerosols, (7) coagulation of precipitation hydrometeors with interstitial and below-cloud aerosols (washout), (8) precipitation and removal of incorporated aerosols (rainout), (9) below-cloud evaporation/sublimation to smaller hydrometeors and aerosol cores, (10) gas washout, and (11) aqueous chemistry.

The major conclusions from the simulation were (1) hydrometeor–hydrometeor coagulation appears to play a substantial role in controlling aerosol particle number concentration globally, (2) washout (as a result of aerosol–hydrometeor coagulation) may be a more important in-plus below-cloud removal mechanism of aerosol number than rainout (the opposite is true for aerosol mass), (3) close-in diameter dual peaks in observed cloud distributions may be in part due to different activation characteristics of different aerosol distributions, (4) evaporative cooling at liquid drop surfaces in subsaturated air may be a mechanism of drop freezing (termed "evaporative freezing"), and (5) heterogeneous–homogeneous freezing may freeze more upper-tropospheric drops than CF, but neither process appears to affect substantially warm-cloud hydrometeor distributions or aerosol scavenging.

Clouds, observed in the upper troposphere at temperatures from −20 to −85°C, are usually called cirrus-like (cirrus, cirro-stratus, and cirro-cumulus). It was also established that stratus, stratocumulus, alto-stratus, and cirrus clouds exert the strongest climate effect, determined by their wide spatial extent. In the framework of the European Union-funded project "Investigation of Cloud by Ground-based and Airborne Radar and Lidar" (CARL), Mavromatidis and Kallos[24] developed the Regional Atmospheric Modeling System (RAMS), which was used to study ice crystal formation and evolution in a cold cloud formation during a field program over Palaiseau, France, in the period from April 26 to May 16 of 1999.

The Doppler radar (3.2 mm–94.9 GHz range) and lidar (532 nm wavelength) cloud sensing data were supplemented with direct measurements from aboard the Merlin flying laboratory. Numerical analysis of the model sensitivity to the shape of particle size gamma-distribution and to the initialization time of the simulation has revealed the most significant parameters, determining the cirrus dynamics, and the comparison with the observations showed the adequacy of the model.

The calculations have shown that the model was able to reproduce the cloud characteristics (e.g., the spatial and temporal variability and the cloud geometry). A detailed comparison of the model results with aircraft data showed that the model-calculated water content and number concentration deviated significantly for small-size particles (2–47 μm) but were in good agreement for medium- (25–800 μm) and large-size (200–6400 μm) particles. The differences for the smaller particles can partially be attributed to both poor performance of the microphysical algorithms and instrument inaccuracies.

Some differences for the larger particles can be attributed either to the definition of the cloud boundaries by the model or to disturbances caused by the aircraft. The time of model initialization is also an important factor affecting cloud formation during the first few hours of the simulation. In general, RAMS appeared to be able to reproduce most of the microphysical parameters of cold cloud formations satisfactorily when utilizing conventional meteorological fields and observations for initial and boundary conditions.

Kreidenweis et al.[21] have compared the models of aerosol scavenging and aqueous-phase oxidation of SO_2 by H_2O_2 and O_3 in a cloud updraft. Approximate models considering only a single droplet size were compared with size-resolved models that explicitly simulate multiple aerosol and drop sizes. All models simulate growth of cloud drops at ammonium bisulfate aerosol lognormal distribution, and subsequent aqueous-phase chemistry during adiabatic ascent.

In agreement with earlier published studies, it was found that relative to approximate models, the size-resolved cloud chemical models consistently calculate 2–3 times more oxidation via the $SO_2 + O_3$ pathway, due to calculated variability of cloud water pH among cloud drops. All models calculate high scavenging of the input dry aerosol mass, but the

calculated number concentration of cloud drops varies within 275–358 drops · cm^{-3}.

These differences result from differences in parameterization of gaseous species uptake, solution thermodynamics, and some other processes. The difference in calculated cloud drop number concentration can propagate to appreciable variations in the cloud optical depth (up to 9%) and cloud albedo (up to 2%). The numerical simulation has shown that the modifications of the aerosol size and mass spectrum are sensitive to the number concentration of cloud drops formed, and the differences in the processed aerosol spectra were found to induce up to 13% differences in calculated light extinction properties of the modified particle distributions. The results obtained demonstrate the important role played by the adequate parameterization of these processes in climate models.

Feingold and Kreidenweis[6] have analyzed theoretically the cloud processing of aerosol through both drop collision coalescence and aqueous chemistry. The numerical simulation was carried out with the aid of a large eddy simulation model that represented (1) size-resolved aerosol, (2) size-resolved microphysics, and (3) the conversion of SO_2 to sulfate via aqueous chemistry.

Earlier estimates have shown that the outcome of processing in a stratocumulus-capped marine boundary layer strongly depends on the cloud liquid water content, aerosol concentrations, trace gas concentrations, and contact time with a cloud. The new model represents spatial and temporal variability of these parameters at large eddy scales on the order of a few hundred meters and timescales on the order of a few seconds.

To analyze the role of different processes in the aerosol–cloud interaction, a number of scenarios were considered: (1) relatively low aerosol concentrations in which aqueous chemistry processing does not substantially affect drizzle, (2) intermediate aerosol concentrations of relatively large size in which drizzle is suppressed by aqueous chemistry, and (3) intermediate aerosol concentrations of relatively small size in which drizzle is enhanced by aqueous chemistry.

The results of the simulation, corresponding to the situations listed above, indicate that aqueous chemistry can modify the dynamics and microphysics of stratocumulus clouds and illustrate the complexity of the coupled system. The latter suggests that the parameterization of the effects of cloud processing of aerosol require careful consideration of a multitude of feedbacks in the cloudy boundary layer.

For reliable numerical simulation of the atmospheric aerosol dynamics and solution of related problems of assessing the meteorological range, as well as for study of heterogeneous atmospheric chemical reactions and the radiative forcing, of great importance is an adequate knowledge of the aerosol size spectrum. Within the framework of the model of shallow stratiform clouds observed during the Second Aerosol Characterization Experiment, Zhang et al.[36] studied the effect of aerosol size parameterization on modeling aerosol–cloud interactions using a one-dimensional version of a climate–aerosol–chemistry model. Both the modal (with prognostic aerosol number and mass or prognostic aerosol number, surface area and mass, referred to as the Modal-NM and Modal-NSM) and the sectional approaches (with 12 and 36 sections) were considered, which predicted the total number and mass of aerosol particles. However, the modal approach with prognostic aerosol mass but diagnostic number (referred to as the Modal-M) failed to provide for reliable results.

The reliability of the particle size distributions appeared to be sensitive to size representations, with normalized absolute differences of up to 12% and 37% for the 36- and 12-section approaches, and 30%, 39%, and 179% for the Modal-NSM, Modal-NM, and Modal-M, respectively. All the representations (except for Modal-M) permit for calculating correctly the liquid water content and the optical thickness of clouds, as well as the aerosol extinction. Further development of this work assumes a consideration of a more realistic aerosol model taking into account a number of different modes and the variable aerosol chemical composition (the latter can affect significantly the number of activated aerosol particles, which is important for reliable estimates of indirect radiative forcing). A significant role in the aerosol size distribution can be played by such processes (ignored in Ref. 36) as in-cloud scavenging of aerosol particles, droplet coalescence, and wet deposition. The parameters of the aerosol activation process call for refinement. The effect of interactions between aerosol and clouds on the microstructure of both aerosol and clouds remains unstudied yet.

The generation of maritime aerosol particles (in particular, CCN) still remains poorly studied. One of important components of this process is represented by the sea-salt particles, entering the atmosphere as a result of "explosion" of sea splashes. The size of such particles varies from 0.1 to 300 μm, and the number density depends on the wind speed. In addition, there are the processes of gas-phase formation of non-sea-salt aerosol particles in the form of the secondary and tertiary nucleation, heterogeneous nucleation, and condensation. In the marine boundary layer (MBL) and in the Arctic and Antarctic regions, *in situ* formation of ultrafine particles of nucleation aerosol less than 0.003 μm in diameter was observed. One of the possible aerosol sources in such cases is oxidation of dimethylsulfide (DMS), leading to formation of sulfate particles (DMS is emitted into the atmosphere by sea phytoplankton). On the coasts (at low tides), the formation of ultrafine particles of biogenic origin was observed almost every day, while in the boreal forests only about 50 events a year.

Based on the data of observations and numerical simulation of gas-phase chemical processes with the AEROFOR-2 model, Pirjola with co-workers[25] have

studied the near-coast formation and evolution of properties of new nucleation aerosol particles in MBL. Coastal regions are known to be a strong source of natural aerosol particles and biogenic vapor, which can condense onto aerosol particles, thus resulting in particle growth. Model simulations determined the instantaneous nucleation rate along with the source rate of a generic biogenic vapor leading to the observed particle size distributions and indicated rapid appearance of ~10^5–10^6 cm^{-3} nucleation mode particles in this environment.

The model calculations suggested values of $3 \cdot 10^5$ $cm^{-3} \cdot s^{-1}$ to $3 \cdot 10^6$ $cm^{-3} \cdot s^{-1}$ for the instantaneous nucleation rate and a value of $5 \cdot 10^7$ $cm^{-3} \cdot s^{-1}$ for the condensable vapor source rate. A significant fraction of the new particles can further be transformed into CCN during subsequent evolution for 3 days under clear-sky conditions at the observed supersaturation levels in clouds, thus contributing to the indirect radiative effect of aerosols. The amount of CCN is mainly affected by coagulation between particles and biogenic vapor condensation, and, to a lesser extent, by condensation of sulfuric acid H_2SO_4 formed by DMS oxidation.

In all simulated cases, more than 100% increase in CCN concentration for supersaturations > 0.35% was observed. In the period of low tidal level, the composition of particles belonging to the nucleation and Aitken modes undergoes significant changes, leading to almost full insolubility of particles, whereas the changes in particles of the accumulation mode prove to be less significant. Due to the following intensification of DMS emissions into the atmosphere, the soluble fraction of particles of the Aitken and nucleation modes, however, increases gradually, thus enhancing their potential as CCN sources.

The available observations indicate also that atmospheric aerosol (as well as precipitation) usually contains bacteria. "Cloud" bacteria are able to grow actively and reproduce at temperatures below 0°C. Bauer et al.[3] studied the ability of bacteria, cultivated from aerosol and cloud water samples collected at a remote Austrian mountain site, act as CCN. Average concentrations of cultivable airborne bacteria amounted to 8 colony forming units (CFU)/m^3 in aerosol samples and to 79 CFU/ml in cloud water. A set of tested bacteria comprised Gram positive and Gram negative but no known ice nucleating species.

The experiments with a CCN counter showed that at supersaturations between 0.07 and 0.11% all types of bacteria were activated as CCN. As the sizes of the bacteria are smaller than the Kelvin diameters at respective supersaturations, physical-chemical properties of their outer cell walls must have enhanced their CCN activity. Keeping in mind that the number concentration of bacteria in atmospheric aerosol is by orders of magnitude lower than the ordinary CCN concentration (about 100–200 cm^{-3}), it becomes clear that the real contribution of bacteria as CCN to formation of water clouds is likely insignificant. However, if bacteria are capable of nucleating ice particles, then they can play a significant role in the cloud icing processes.

Iversen and Seland[13a] numerically simulated climate based on the extended version of the National Center for Atmospheric Research (NCAR) Community Climate Model 3 (CCM3) including the life cycle schemes for sulfate (SO_4) and black carbon (BC). The parameterization of sulfur cycle accounted for the emissions of maritime dimethyl sulfide (DMS), SO_2, and sulfate of natural and anthropogenic origins and emissions of BC from biomass burning and fossil fuel combustion. Chemical and physical processes of aerosol generation and transformation were parameterized based on prescribed oxidant levels and background aerosols of marine, continental, and polar origins. In case of aqueous chemistry processes, the dependence on estimated exchange rate in cloudy and clear atmosphere was examined.

With emissions from International Panel on Climate Change (IPCC), the life times (atmospheric turnover) proved be equal (in days) to 1.5 (SO_2), 3.5 (SO_4), and 4.7 (BC) for 2000 year and 1.6 (SO_2), 4.0 (SO_4), and 4.7 (BC) for 2100 year (according to the A2 emission scenario). The modeled SO_x compounds agree within a factor of two with observations at the ground level in North America and Europe and for SO_4 in free troposphere. For BC, the ground-level concentrations were well within a factor of ten.

The calculated BC and SO_4 values were about a factor of ten underestimated comparative to the measured in Arctic winter, which could partly be linked to high variability of cloudiness conditions. To the contrary, the SO_4 and BC values were a factor of ten overestimated at ground-level low latitudes. These major model biases are caused by neglected transport and low scavenging efficiency in cumulus clouds. In this connection, specialized computer experiments were conducted to estimate the sensitivity of simulated results to accounting for cloud processes. The analysis of these estimates showed SO_4 and BC to be very sensitive to the vertical transport and scavenging in convective clouds. Thus, it is necessary to improve further the parameterization techniques for cloud processes, as well as for interaction of aerosol and clouds.

The atmospheric aerosol consists of hundreds and even thousands of organic and inorganic compounds, and an important fraction of aerosol particles is represented by CCN, which play a critical role in formation of cloud droplets. The understanding of the processes of CCN functioning is seriously complicated by the available inadequate information about the organic fraction of aerosol and the poorly reliable description of interactions between the organic and inorganic aerosol components. The experimental laboratory investigations of CCN were limited mostly to the consideration of either polydisperse aerosol of complex chemical composition, existing under the real atmospheric conditions, or individual laboratory-made components of aerosol.

Earlier investigations revealed a wide variability in the capability of different individual compounds to stimulate the formation of aerosol particles. If some organic compounds show this capability to only a small degree, then other compounds can be active as inorganic salts. CCN in the atmosphere are usually composed of multiple inorganic and organic chemical species, the interactions between which in the process of formation of cloud droplets remain poorly studied as yet.

In this connection, Raymond and Pandis[29] studied the capability of CCN, having a mulicomponent composition, to stimulate the process of formation of cloud droplets, as well as their properties. Laboratory experiments were performed using mixed, multicomponent particles as well as particles consisting of a core coated with hexadecane. The particles were composed of sodium chloride, ammonium sulfate, pinonic acid, pinic acid, norpinic acid, glutamic acid, leucine, and hexadecane. Activation diameters were determined combining a Tandem Differential Mobility Analyzer (TDMA) with a thermal diffusion Cloud Condensation Nucleus Counter (CCNC). The studies were performed at supersaturations of 0.3% and 1% with dry particle diameters ranging between 0.02 and 0.2 μm.

The results of laboratory measurements of parameters of cloud droplet formation were compared to a theory assuming additive behavior of the CCN constituent species. This assumption was sufficient for prediction of the CCN activation diameter of the mixed particles. The results obtained indicate that hexadecane coating, whose volume makes up to 96% of the total particle volume, does not affect the activity of particle cores as CCN, which is in agreement with the results obtained earlier for particles with cores composed of ammonium sulfate. The addition of 1% solution of sodium chloride affects strongly the activation diameter.

Kärcher and Lohmann[15] proposed a parameterization of cirrus cloud formation by heterogeneous freezing, which accounts for the effects of sizes of aerosol particles acting as CCN on the freezing process in adiabatically rising air parcels. Aerosol size effects become important when the timescale of the freezing event is fast compared to the timescale of depositional growth of the pristine ice particles. The generalized parameterization scheme was validated with parcel model simulations. The relationship between aerosol and ice crystal number concentrations in cirrus clouds formed by homogeneous freezing was analyzed. This relationship is found to be much weaker than in liquid water clouds. It is shown in Ref. 15 that even freezing of enhanced levels of sulfate aerosol originating from strong volcanic eruptions is unlikely to exert a sensible influence on cirrus formation. Thus, for crystal clouds the Twomey mechanism of the aerosol effect on the cloud properties is not so strong as for water clouds.

Gao et al.[8] conducted *in situ* measurements of relative humidity with respect to ice (RH_i) and of nitric acid (HNO_3) in both natural and contrail cirrus clouds in the upper troposphere. According to these measurements, at $T < 202$ K RH_i values show a sharp increase to average values of over 130% in both cloud types. These enhanced RH_i values were attributed to the presence of a new class of HNO_3-containing ice particles (Δ-ice). It was assumed that surface HNO_3 molecules prevent the ice/vapor system from reaching equilibrium by a mechanism similar to that of freezing point depression by antifreeze proteins. The presence of Δ-ice particles represents a new link between global climate and natural and anthropogenic nitrogen oxide emissions. Hence, it follows that including Δ-ice in climate models will alter simulated cirrus properties and the distribution of upper tropospheric water vapor.

Studying the processes in cirrus clouds, forming in anvils of vertical-development clouds, Fridlind et al.[7] have come to the conclusion that ice crystals in cirrus are usually formed on CCN, represented by mid-tropospheric rather than boundary-layer aerosols, as was believed before. Consequently, aerosol emitted from distant pollution sources may have a greater effect on anvil clouds.

The promising field of development in the context of the aerosol–cloud interactions arises in connection with the recently discovered relationship between polar mesospheric clouds and the layer of iron atoms, existing at the same altitude.[12,26]

The dominant feature of the Arctic climate is formation of temperature and humidity inversions, especially during the cold season. The temperature inversions are caused by radiative cooling, advection of warm air, downwelling flows, cloud processes, thawing at the surface layer, and topography. Formation of the humidity inversions is favored by precipitation in the form of ice crystals, which leads to a humidity decrease in the lower atmospheric layers. The information about the amount of clouds in the Arctic was mostly obtained from the data of ground-based observations, according to which the annual behavior of the total cloud amount has a maximum in summer (up to 90%) and a minimum in winter (40–60%). The comparison of the satellite data on the cloud amount with the ground-based data has shown that the former give higher (within 5–35%) estimates.

Although very complicated global circulation models were used to reconstruct the dynamics of climate in the Arctic, it is doubtless that the parameterization of the cloud and radiation dynamics, employed in these models, cannot be considered as realistic. To study the real dynamics of ice and mixed clouds and their effect on the radiative transfer, Lohmann et al.[22] carried out numerical simulation using the mesoscale model GESIMA. The results of simulation were illustrated by the consideration of particular situations from the observations dated to April 16 and 28–29 of 1998 on the SHEBA drift-ice research station within the framework of the Arctic

Cloud Experiment, being a part of First ISCCP Regional Experiment (FIRE ACE).

The comparison of the results of numerical simulation with the observations has demonstrated that the GESIMA model is able to simulate adequately the cloud boundaries, ice and liquid water content, and effective radii of cloud particles. The calculations corresponding to the conditions of clear atmosphere and the atmosphere polluted with anthropogenic aerosol have shown that the anthropogenic aerosol can alter microphysical properties of Arctic clouds (increases the number concentration of fine droplets) and, consequently, modify surface precipitation. On the contrary to the earlier results, it was found that the accretion of snow crystals with cloud droplets is increased in the polluted cloud owing to its higher cloud droplet number concentration. In addition, the autoconversion rate of cloud droplets and accretion of drizzle by snow decrease because of shutdown of the collision-coalescence process in the polluted cloud.

As a result, from combination of these two opposing effects, the precipitation decreases in the polluted atmosphere as the aerosol concentration increases from 100 to 1000 particles/cm^3. The amount of precipitation reaching the surface as snow crucially depends on the crystal shape. If crystal aggregates are assumed, then a tenfold increase in aerosol concentration leads to an increase in accumulated snow by 40% after 7 hours of simulation, whereas the snow amount decreases by 30% when planar crystals are assumed because of the larger accretion efficiency of snow crystals with cloud droplets in case of aggregates.

In the context of the subject under discussion, of great interest are polar stratospheric clouds (PSCs), which play a dual role in the process of depletion of stratospheric ozone in polar regions: 1) activation of halides occurs on the surface of PSC particles; 2) sedimentation of the largest (radius $r > 5$ μm) PSC particles, containing nitric acid, favors denitrification, which decreases chlorine deactivation and thus intensifies the catalytically induced ozone reduction. Large particles, containing HNO_3 and having a low number concentration ($N \approx 10^{-4}$ cm^{-3}, $r \lesssim 10$ μm), were observed in Arctic PSCs of synoptic scale. To the contrary, PSCs arising in orographic waves are characterized by relatively small horizontal scales (of the order of several thousands km^2) and are produced under the conditions of cold climate due to the adiabatic displacement of air masses.

Orographic PSCs are comprised of particles of different types. If the cooling process is rather slow, at the temperature 2–3 K above the ice frost point (T_{ice}), then the stratospheric sulfuric-acid (H_2O/H_2SO_4) aerosol assimilates a large amount of nitric acid and water vapor, which determines the formation of droplets of supercooled ternary solution (STS). At the temperature about 3 K below T_{ice}, the ice nucleation occurs in STS particles. In the direction of the air flow from Arctic ice clouds, solid particles were observed, consisting, most probably, of nitric acid trihydrate (NAT = $HNO_3 \cdot 3H_2O$).

According to the available observations, NAT is a part of PSCs and plays an important role in the sedimentary denitrification, enhancing the chlorine-catalyzed ozone reduction. However, the scientific understanding of NAT formation mechanisms is vague. Luo et al.[23] have shown that high cooling rates in orographic waves, though causing only moderate NAT saturation ratios ($S_{NAT} = P^{part}_{HNO_3} / P^{vap}_{HNO_3} \lesssim 30$, where P is the partial pressure due to particulate (part) and vapor (vap) NAT) within STS droplets, lead to $S_{NAT} \gtrsim 500$ within the gas phase. Laboratory measurements suggested that such extreme supersaturations promote NAT deposition nucleation on ice or other solid surfaces, as long as they are not coated by STS.

On the basis of airborne lidar PSC observations during three Arctic winters and a combined microphysical/optical model, a simple parameterization was proposed[23] for a NAT deposition and nucleation rate, which depends only on the NAT supersaturation and temperature. The results obtained indicate that deposition and nucleation of NAT on ice surfaces exposed directly to the gas phase driven by extreme NAT supersaturations is able to explain the formation of NAT particle number densities up to 10 cm^{-3}.

Luo et al.[23] discussed possibilities of meteorite matter functioning as a factor providing for the development of heterogeneous reactions and nucleation of NAT. Micrometeorite particles with sizes > 0.1 μm at the number concentration of about 10^{-5} cm^{-3} cannot provide for the formation of the high concentration of NAT particles, observed in PSCs in orographic waves. In the presence of meteorite dust with the particle radius of about 5–10 μm and at high number concentration, the total surface of the meteorite particles proves to be by two to three orders of magnitude smaller than the total surface of ice particles. Because S_{NAT} over the surface of non-ice particles is four times higher and the NAT nucleation rate on the surface of meteorite particles is unknown, NAT nucleation may be significant in this case. Observations, confirming this assumption, are still lacking.

Conclusions

A number of unsolved problems in interactions of atmospheric aerosol with clouds are indicative of the need in further investigations, whose urgency is determined, in the first turn, by the key significance of these problems for understanding and quantifying of the aerosol–cloud impact on climate. The program of the future studies should include consistent field observational experiments and numerical simulation. Good example of such efforts are developments within the framework of the Field Investigations of Budgets and Conversions of Particle Phase Organics in

Tropospheric Cloud Processes (FEBUKO) and Modeling of Tropospheric Multiphase Processes: Tools and Chemical Mechanisms (MODMEP) described by Herrmann and Wolke.[11]

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