AEROSOL–CLOUD–CLIMATE INTERACTIONS. PART 1. AEROSOL

(to the results of the Symposium of the International Association of Meteorology and Atmospheric Physics held in Vienna, August 13–20, 1991)

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The reports devoted to the investigations of the microphysical and radiative characteristics of atmospheric aerosol as well as to the evaluation of the effect of aerosol on the climate are reviewed. An interaction between aerosol and clouds is analyzed. Significant contribution of atmospheric aerosol to the climate formation is shown.

The Third Symposium on aerosol–cloud–climate interactions, organized under the auspices of the International Association of Meteorology and Atmospheric Physics, was held in Vienna, August 11–24, 1991 as a part of the 20th General Assembly of the International Union of Geodesy and Geophysics. This Symposium was the longest (6 days) in comparison with all other Symposiums of the Assembly, although a lot of contributions were not accepted by the Program Committee. Such an attention to the problems being discussed is not occasional. As is well known, two basic unsolved problems of global climatic changes are: 1) necessity for deeper insight into both the cloud dynamics (and its parameterization on this basis) and the interaction of clouds with radiation; 2) need for further investigations (and taking them into account more comprehensively in numerical modeling of climate) of the interaction of the atmosphere with the ocean (primarily, determination of the contribution of the deep ocean circulation). Therefore, the problems associated with study of the cloud cover are considered in USA as the most important part of the national program on Global Changes. Although we can hardly agree with such an order of priorities in studying the global changes, the priority of investigations of the aerosol–cloud–climate interactions is obvious.

It should be noted at once that the organization of the Symposium (and its program) could not be admitted as irreproachable: the range of problems of numerical modeling of the global climate – the main part of the aerosol–cloud–climate system – was practically lacking (leading specialists in the field of numerical modeling took part in other Symposiums, while the scale of the Assembly and spread of places of meetings made extremely difficult even the informal contacts). The Symposium was opened by Pr. P. Hobbs (USA), the Chairman of the Symposium, who restricted himself to a formal report about organizational problems instead of a conceptual substantiation of the problems of this Symposium. Inspite of a great number of reports (classified into 21 sessions, with Chairmen) the main point of the problem – an analysis of the contribution of atmospheric aerosol to the climate formation and an account of aerosol in numerical models of climate – were elucidated extremely inadequately.

Instead of comprehensive discussions of the problem with well–defined priorities, the work was reduced to the fragmentary consideration of its separate aspects (in a number of cases being of secondary importance). The invited speakers who had doubled time at their disposal (30 minutes instead of 15) failed to give the Symposium a proper lead and, as a rule, restricted themselves to biased reviews of rather a special character. Since no Soviet specialists were among the invited speakers, the results of investigations of Soviet scientists in the field in which our contribution was of great importance and a number of cases –– even unique, were not mentioned at all. It was especially sorrowful because the number of reports of Soviet scientists was small. The reports were not discussed at all (and the time needed to answer the questions was short). As a result, it was impossible to supplement the reports.

Although the above–mentioned facts do not bring to an end my critical remarks (I will return to them at the end of this review), I will focus my attention on the brief description of the Symposium following the structure of its program.

TROPOSPHERIC AEROSOL

1. Spatial distribution. The invited report presented by Pr. R. Jaenicke (Germany) incorporated a lot of interesting information including new data on the effect of *in situ* aerosol formation, the important contribution of biogenic aerosol, and the evidence that the contribution of a desert in the formation of the atmospheric aerosol is more important than that of an ocean. New estimates of the formation of atmospheric aerosol in clouds (due to evaporation of droplets) are of great interest. According to these estimates, the global mass of aerosol reaches 3000 tetragram per year, while the rest of the sources are characterized by the following figures (in tetragram/year): 1500 (*in situ*), 450 (biosphere), 2000 (underlying surface without vegetation), and 1000–2000 (oceans). The data on the global distribution of the condensation nuclei (CN) are of great importance, as well as the results of a three–dimensional numerical modeling of aerosol distribution. These results revealed the contribution of deserts (in this connection, the complete ignorance of the results obtained by Soviet scientists and repeatedly discussed, in particular, in the publications written in English). H. Rodhe and I. Langner (Sweden) described an interesting three–dimensional model of formation, transformation, and transfer of sulphate aerosol in the altitude range corresponding to 1000–100 GPa on a grid 10° in latitude and 10° in longitude (unfortunately, the speakers were unfamiliar with analogous but more comprehensive models developed by the Soviet scientists). The transfer was calculated on the basis of the well–known wind field given that both natural (DMS, H_2S) and antropogeneous (SO_2) sources of sulphate aerosol formation were prescribed. The processes of transformation of sulphate aerosol depend strongly on the concentration of oxidants (OH) and the cloud amount (the problem of interaction of aerosol with clouds drew much attention during the Symposium and formed one of the new current tendencies of its development). As to the aerosol sinks, they are primarily determined by the processes

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of dry and moist (washing out by precipitations) sedimentations. The calculations of the global distribution of the number density of sulphate aerosol near the ground showed the presence of maxima over the West Europe and European territory of the USSR (it is strange but this maximum is absent over the Eastern USA). The number density of the sulphate aerosol at the altitude corresponding to 900 GPa increased during the last century more than twice over ~ 50% of the territory of the Northern Hemisphere. Probably, this process had an appreciable effect on climate.

A. Clarke (USA) analyzed the large quantity of vertical profiles of the CN obtained from airborne observations within the layer 8–12 km (extended from 70° n.l. to 58° s.l. over the Pacific ocean) and discovered the negative correlation between the number density of the ultrafine fraction of the CN (with the diameter varying in the limit 3–20 nm) and the surface area of the largest particles. The main source of the CN is the upper troposphere. The evolution and growth of the CN size points to the fact that entering into the atmospheric boundary layer (ABL), they form "new" CN of clouds. R. Husar and V. Wilson (USA) having processed the data on of the meteorological visibility range obtained at the network stations over the territory of USA during the last 40 years as well as the available evidence about the chemical composition of aerosol made a conclusion that over the Northeast USA the trend of decreasing the number density of aerosol took place, while over the Southwest USA the opposite trend was established. The aerosol attenuation coefficient B_H, used as an indicator, increased almost twice and was correlated with intensification of emissions of the sulphur gas. Accompanying changes in the climatic parameters were manifested in the decrease of the difference between the surface air temperature (SAT) at midday and midnight, in elevation of the dew point and in increasing the relative humidity and recurrence of fogs.

2. Technique, measurements, and origin. O. Preining et al. (Austria) drew attention to the discrepancy of the data of various counters of atmospheric aerosol (AA) and CN due to the lack of unified techniques and proposed to conduct an international intercomparison of instruments at the Vienna University. Data on the number density of the AA over the Pacific ocean obtained aboard the DS–8 aircraft in November, 1989 with the help of a photoelectric counter and a wire impactor were discussed by R. Pueschel et al. (USA). Processing of the data obtained for the aerosol particles with radii varying in the limits 0.03–3.0 μm resulted in the following values of the parameters of the lognormal size distribution: $0.05 < r_g < 0.6$ μm, $12 < N < 64$ cm^{-3}, and $1.23 < \sigma < 1.30$. The following elements predominated in aerosol composition (according to the particle number density): S (about 50%), Cl (31%), Si (15%), Na (7%), and Mg (5%). Sufficiently uniform distribution of the sulphate AA over the troposphere indicates spatially uniform process of their formation. On the contrary, the number density (ND) of particles containing chlorine and sodium decreases faster with increase of altitude, which, apparently, results from the fact that they are formed near the earth's surface. Sulphate component of the AA in the free atmosphere amounts to less than 20% of total, while the volume number density of particles with diameter varying in the limits 0.3–20 μm, as a rule, fluctuates about 0.01 μm^3/cm^3. Sometimes (at temperature close to the point of saturation above ice and in the case of air movement caused by orography or by gravitational waves), the sudden increase in the ND of particles by several orders of magnitude occurs.

Shipborne observations performed by K. Dowd et al. (Great Britain and Ireland) in the Northeast Atlantic have shown that the air mass of Arctic origin can be characterized by very low number density, predominantly, of the sulphate aerosol of accumulation mode, while in the marine air the ammonium sulphate (up to 80–90% of total) predominates also with low number density. Radical change of the aerosol composition and number density of aerosol particles is caused by air mass intake from Great Britain and Central Europe, namely, the ND increases at least by an order of magnitude, and sulphate of ammonia predominates in the composition of the AA (65–85% of total). The relative content of the soot component varies from 2–6% (clear Polar or marine air) to 10–30% (air masses of continental origin).

Emphasizing the strong spatio–temporal variability of the properties of the AA, A. Jayaraman and B. Subbaraya (India) presented the results of investigations of the optical parameters of the AA in the range of wavelength from the UV to the IR at the altitudes up to 30 km from the data of the rocket and balloon photometric probing started in 1980 on the polygons of Thumba (8.5° n.l.) and Hyderabad (17.5° n.l.). The measured vertical profiles of the aerosol attenuation coefficient with various models were compared. F. Valero et al. (USA) succeeded in reconstruction of the aerosol optical thickness (AOT) of the atmosphere at different altitudes from the data of airborne measurements of vertical profiles of total and scattered radiation over the Arctic with the help of a 7–channel radiometer in the wavelength range 780–1064 nm (outside the strong bands of molecular absorption). Spectral dependence of the AOT was then used for determining the aerosol microstructure.

3. Effect on climate. O. Preining (Austria) used a zero–dimensional heat–balance model of climate for the comparative estimate of the contribution of CO and aerosol to the change of the averaged global surface temperature of air. Having obtained the contributions of approximately equal values (but with opposite signs), the speaker drew attention to the necessity for a correct account of the AA as a climate–forming factor.

More complicated one–dimensional model (5–layered model of the atmosphere and 2–layered model of the soil) for estimating the aerosol contribution to the climate formation was implemented by Yongfu Qian and Young Yu (China). The results of calculations showed the decrease of the surface and soil temperatures as well as the decrease of the temperature at the altitude corresponding to 100 GPa due to aerosol. Climatic effect of aerosol was studied as a function of aerosol optical characteristics and albedo of the underlying surface. The important contribution of desert aerosol as a regional climate–forming factor was illustrated. M. Legrand (France) analyzed the contribution of the atmospheric aerosol in the scope of the problem of the atmospheric correction in the IR range. T. Hayasaka et al. (Japan) presented the results of ground–based nephelometric observations (in Sapporo and Sandai during 1986–1987) of the scattering phase functions for the orthogonally polarized components of radiation with subsequent use of these data for retrieving the microstructure of the AA. An analysis of the filtered air samples can be used to obtain the data on the content of various components of the AA (soot, organic carbon, sulphates originated not from the marine salts, nitrates, sulphate of ammonia, marine salts, soil component, and water). The soot component exhibiting a pronounced annual behavior most strongly affects the optical characteristics of the AA.

A. Deepak and G. Vali (USA) summarized the results of the vast work on the foundation of the International Global Aerosol Program (IGAP) reflecting an increasing urgency of aerosol investigations in the scope of the problem of interactions in the "geosphere–biosphere" system. The main purposes of the program are as follows: developments in the field of global aerosol climatology, a comprehensive study of the processes of interaction of aerosol with clouds, and, finally, adequate account of the AA as a reason for the global climatic changes. Various theoretical and experimental investigations must be performed to achieve these purposes. At

the Meeting of the International Comission on Radiation the proposal was approved to form the Working Group for further development of the program.

L. Stowe et al. (USA, USSR) discussed the results of the comparisons of the AOT retrieved from the satellite data (a modified radiometer with very high spectral resolution MRVHSR) and those found from the data of shipborne observations obtained with the help of a cloud photometer (placed aboard the scientific research ship *Akademik Vernadskii*). The main result of the study carried out in the scope of Soviet–American collaboration (Working Group of the Earth Sciences) was the establishment of considerable disagreements. To make clear the reason for these disagreements the further investigations are needed. This fact can be illustrated, for example, by the following series of the AOT at a wavelength of 0.5 μm:

AOT (satellite)	0.03	0.08	0.00	0.17	0.28	0.25
AOT (ship)	0.07	0.03	0.03	0.26	0.42	0.48

As can be seen, the satellite values of the AOT are systematically underestimated.

S. Kreidenwais et al. (USA) described an interesting three–dimensional global model of the transfer, transformation, and sedimentation of the accumulation and nucleation modes of the sulphate aerosol (GRANTOUR), with the following reactions: $DMS + OH \rightarrow SO_2$; $SO_2 + OH \rightarrow SO_4^=$; $SO_2 \rightarrow SO_4^=$ providing the basis for this model. Approximate parameterization of a gas–phase process of sulphate aerosol formation $S\ (gas) \rightarrow SO_x^=$ (aerosol), as a result of which new particles are formed and the existing particles grow, can be used in the interactive model "aerosol–climate" based on the climatic model CCM–1 developed by the scientists of the National Center of Atmospheric Research (USA). The model GRANTOUR was implemented for calculation of the global fields of the CN and AA for the given antropogeneous emissions of sulphur gas due to the combustion of mineral fuels and natural emission of dimethyl sulphide in order to estimate the contribution of these emissions to the formation of the global fields of the AA. One of the regions of the most intensive generation of the sulphate aerosol is in the East Mediterranean.

The purpose of lidar sensing of aerosol and *in situ* observations carried out by E. Dutton et al. (USA) in Boulder was the estimate of the optical parameters of the cloud of smoke resulting from the forest fires. The maximum AOT of the cloud at a wavelength of 0.5 μm reached 2, while the decrease of the net radiation associated with this cloud was about 28%. The effect of the cloud of smoke on the heat radiation fluxes was negligible.

A. Robock (USA) performed new estimates of contribution of the "volcanic signal" to the climatic changes using the data on the latitudinal variability and annual behavior of the SAT for the last 140 years keeping in mind that it is of great interest for identifying the "greenhouse signal". If the changes in the SAT caused by the ENSV (El Nin'o Southern variation) is filtered out, the distinct correlation between the SAT and decay of volcanic index (DVI) appears, namely, during a few years after large eruptions the SAT falls, and this effect is stronger in the Polar regions in winter, thereby demonstrating the inverse relation between the SAT and the albedo. The most distinct correlation DVI ¢ T took place during 1928–1988.

4. Aerosol–cloud Interactions. As was noted, this important subject drew much attention. In the review presented by P. Hobbs (USA) two aspects of this problem were analyzed: 1) effect of the AA on the structure and characteristics of clouds; 2) effect of clouds on the AA. It is well known that the AA determines initial concentration and microstructure of cloud droplets (consequently, the optical characteristics of clouds). However, it is not clear to what extent the effect of aerosol is weakened as clouds of different types evolve and age. There are a lot of uncertainties concerning the contribution of clouds to the formation of ice particles. An important (and already mentioned) point is the relationship between the concentration of dimethyl sulphide and the CN. On the one hand, entering of aerosol, which acts as the CN, into the droplets engenders the changes in the chemical composition of clouds and precipitations (resulting in the acidic precipitations). On the other hand, the product of chemical reactions proceeding in the droplets is an aerosol residue after evaporation of droplets. Since this process can recur in the cloud layer, the considerable transformation of aerosol microstructure occurs. During the precipitations, removal of aerosol from the atmosphere occurs. Thus, the clouds can serve as sources and sinks of aerosol (note that in this connection the interaction of aerosol with cloudiness and radiation is of great importance). Moistening of air surrounding the cloud (which spreads at long distances from the clouds) favours nucleation of new particles.

I. Kaufman and Ming–Da Chou (USA) implemented a two–dimensional multilayered climatic model with energy balance in order to estimate the effect of antropogeneous sulphur gas on the climate and to compare it with the contribution of the greenhouse effect. The calculation of increase in the optical thickness (and albedo) of clouds caused by the gas–phase formation of the CN resulted in the following conclusion: "greenhouse" warming is compensated to a considerable extent due to the growth of the albedo of clouds. In this connection the fact that each SO_2 molecule is 50–1100 times more efficient as a cooling agent in comparison with the SO_2 molecule treated as a heater, is of great importance. Therefore, the current emissions of sulphur gas can compensate up to 70% of "greenhouse" warming and by 2060 the SAT increase will be only 60% of "greenhouse" warming. Since the lifetime of the SO_2 molecules in the atmosphere is much longer than that of the SO_2 molecule, at first the sharp reduction of the amount of mineral fuels will accelerate the global process of warming, and only later the undisturbed condition will return. Note that it is very hard to estimate the reliability of the results being considered. However, it is clear that the aerosol–climate problem is of great urgency.

S.L. Halsa (USA) studied various ways of penetration of aerosol into the clouds from airborne sensing, and E. Carter and R. Borys (USA) carried out the field experiment in order to compare the chemical composition of aerosol and cloud droplets as a function of their size. The comparisons revealed a similarity of the composition of clouds and aerosol but the difference depending on the size of droplets took place. It should be taken into account in developing the models of moist sedimentation.

F. Parungo et al. (USA) studied the effect of concentration of the CN on the number of clouds and short–wave (SW) and long–wave (LW) cloud–radiative forcing (CRF) from the data of the MRVHSR observations (3.7 μm channel) in September, 1982. Simultaneously the shipborne observations of the concentration of the CN near the southeast coast of China, in the West and Central Pacific ocean were carried out. Strong correlation of the CN concentration with the number of clouds as well as with their albedo was shown. Specific data on radiative fluxes and the CRF were not discussed.

In an interesting (from the conceptual point of view) report presented by G. Shaw some ideas about the contribution of dimethyl sulphide entering into clouds from the ocean as a modulator of the cloud cover and, hence, the climate, were formulated. The purpose of the report presented

by I. Kaufman and R. Fraser (USA) was an analysis of the effect of a haze engendered by the forest fires in the Amazon basin on the microstructure and albedo of clouds. According to the data of the MRVHSR (0.63 and 8.7 μm channels), the values of mean particle size, optical thickness τ, and cloud top height were retrieved. At $\tau < 1.0$, the decrease of the size of cloud particles with increase in the number density of haze particles took place. The presence of a soot component in the haze determined the nonuniformity of the effect of haze on the variations of the albedo of clouds. A. Ackerman et al. (USA) implementing a one–dimensional nonstationary model, discussed the effect of the ship smoke emissions on the marine stratocumulus clouds as an example of aerosol–cloud action.

The same theme was concerned in the report presented by M. King and L. Radke (USA) who analyzed the data of airborne measurements of angular distribution of radiation scattered within the stratocumulus clouds (at 13 wavelengths in the range 0.5–2.3 μm) whose optical characteristics were transformed under the action of smoke emissions of two ships near the coast of South California. An increase of the optical thickness of clouds but a decrease of the radiation absorbed by the clouds occurred.

The report presented by D. Baumgardner and S. Twohy (USA) was devoted to the study of the spatial distribution and microstructure of aerosol in the atmosphere surrounding the clouds from the data of airborne observations. Simultaneously, the microstructure of clouds was measured in marine air masses near the coasts of Hawaiian Islands and California as well as in the continental air masses over North Germany and East Colorado. The results of measurements of the particle number density and microstructure of convective cloudiness in the process of its development show that the number density and mass of aerosol decrease considerably outside the clouds.

In view of great interest to the analysis of the possible effect of the antropogeneous sulphate aerosol on the optical characteristics of the cloud cover and climate, R. Chuan et al. (USA) performed the calculations of the effect of the cloud microphysics and chemical composition of the AA as well as of the meteorological conditions on the microphysics of clouds having implemented Lagrange's approach for this purpose. For the given global distribution of the emissions of sulphur gas (g/m^2), the distribution of concentration of ammonium sulphate near the ground was calculated. The transformations of the microstructure and optical characteristics of clouds caused by the sulphate gas–phase aerosol were rather considerable. Global averaged estimates indicate an increase of the albedo of the "Earth's surface–atmosphere" system approximately by 2% which results in cooling equivalent to 1.55 W/m^2 (these estimates refer to the present time). Therefore, the aerosol–cloud effect compensates completely the greenhouse effect. Research work is carried out in order to obtain more detailed and reliable estimates based on the composite model CCM–1 + GRANTOUR.

In conclusion let us note that recently a significant progress has been achieved in investigating the aerosol as a climate–forming agent not only due to its direct effect on the climate but also due to the aerosol–induced transformation of changing the microstructure and optical characteristics of clouds.