On the optical properties of polluted clouds

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Received July 2, 2001

Some possible mechanisms of interaction of cloud droplets with soot particles and the optical properties of the corresponding systems are considered. It is shown that for high, but quite realistic soot concentration, the cloud single scattering albedo may reach a range sufficient to reproduce the effect of anomalous cloud absorption.

The experimental and theoretical studies of radiation regime of a cloudy atmosphere show that the optically active anthropogenic aerosols influence substantially the optical characteristics of clouds. For instance, one of the key problems in modern atmospheric optics is the problem on anomalous absorption in stratus clouds. The experimental measurements of optical properties of stratus clouds give cloud single scattering albedos in the visible range much lower than Mie scattering calculations predict.[1,2] In line with recent effort to explain the anomalous absorption in nonstandard way (ranging from claims that it is due to misinterpretation of experimental data to suggestions to revise the mechanisms of interaction of electromagnetic waves with cloud particles), the present paper tries to answer the question if the possibilities to explain the anomalous absorption in the framework of classic scattering theory are completely exhausted.

Since the absorption by pure water in the visible spectral range is vanishingly small, it remains to assume that an absorbing aerosol is present in the cloud, either as independently mixed with water droplets or interacting with them. The numerical estimates show that, for real concentrations, the only possible aerosol can be soot particles.

Both field and laboratory studies of the behavior of soot particles in the presence of liquid-phase fraction indicate the variety of states in such a two-phase medium, depending on the source, mechanism of soot formation, and on the presence of different admixtures (ozone, sulfurous gas, sulfuric and nitric acids, etc.), and their residence time in the cloudy medium.

In the polluted clouds, the following morphologic structures can be envisioned:

1) finely dispersed soot particles modeled as spheres;

2) cloud droplets covered with tiny soot particles;

3) water droplets with soot core, that can be approximately modeled as a sphere;

4) soot particles of fractal type;

5) soot fractals packed by high humidity with a few large globules or transformed to a particle that can be modeled as an oblate spheroid.

The present paper considers the models 1–3. The model 1 is a standard model of independently mixed water and soot particles assumed to be homogeneous spheres. Model 2 simulates in the approximation of a two-layer sphere, whose cover has a complex refractive index calculated as an average weighted by mass of soot and water in the surface layer of the drop (determined by the radius of soot particles). The laboratory studies performed indicate that, at the initial stage, the soot adsorption on droplet proceeds piecewisely rather than uniformly.[3] Hence, the two-layer spheres with very thin soot layers are unlikely to exist. Lastly, model 3 is again the standard model of a two-layer spherical particle with soot core and water shell. The electron microscope analysis of aerosol particles suggests that such structures are quite frequently observed.[4] To calculate the optical characteristics of two-layer particles (models 2 and 3), the algorithms from Refs. 5 and 6 were used.

To minimize the number of calculated parameters, a single particle radius was assumed, which reduced the number of varied parameters to three:

a) Q is the ratio of soot to water mass in the unit volume;

b) R is the water droplet radius; and

c) r is the soot particle radius.

Obviously, the transition to particle ensembles will not substantially change the result, especially considering that we aim not at performing exact quantitative calculations, but rather at the qualitative estimates, for which the cloud absorption is close to experimentally measured anomalous value.

As most representative optical characteristic, we chose the quantity $w = 1 - \Lambda$, where Λ is single scattering albedo (hence w is the scattering to extinction ratio).

Some calculated results are presented in Figs. 1 and 2. Note that in model 2 (large drops with strongly absorbing shell) for quite large core radius, the algorithm from Ref. 6 has been modified to use asymptotic rather than exact optical radiation calculations for the computation time reason. Because of this, the optical characteristics change

0235-6880 / 02 / 02 151-03 $02.00

abruptly. The change is not significant for extinction and scattering cross sections; but it becomes considerable (tens of a percent) for absorption cross section (which is a small residual of the two first). To circumvent this discontinuity, the calculations by asymptotic formulas were œtailoredB with the exact calculations. In Figs. 1 and 2, such asymptotic regions are indicated by dashed line (meaning that the dashed lines correspond to less confident results than the solid ones).

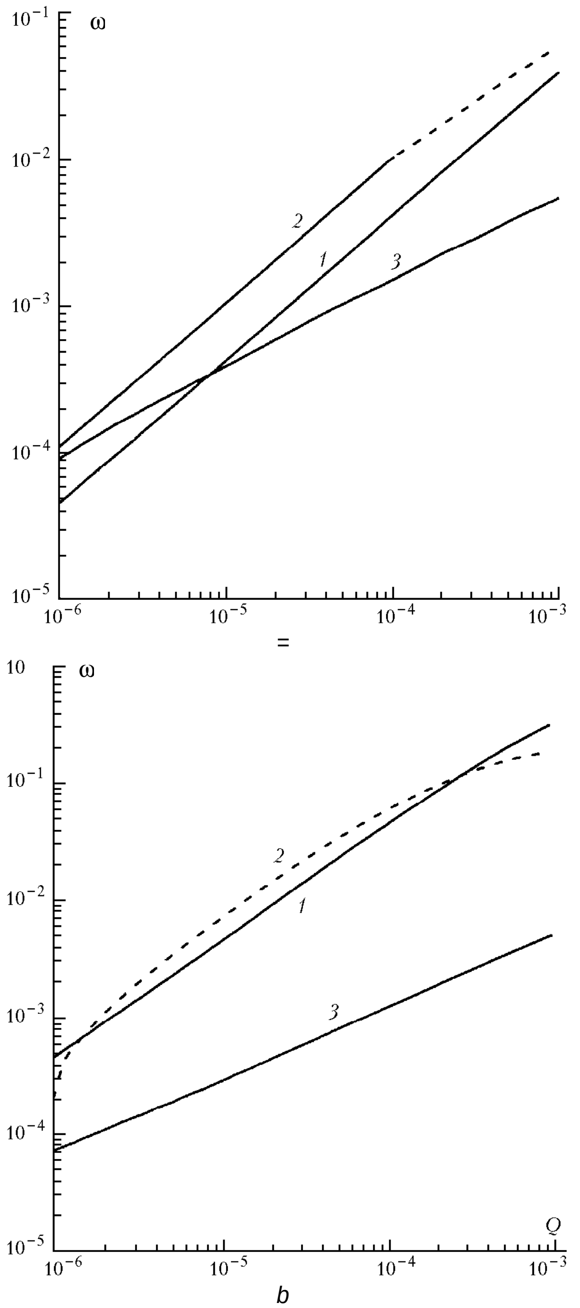

Fig. 1. The $w(Q)$ dependence for three models of interaction of droplets with soot particles. Calculations were made for $r = 0.005$ μm. (Dashed line shows region of asymptotic calculations according to Ref. 2). $R = 10$ μm (*a*) and $R = 100$ μm (*b*).

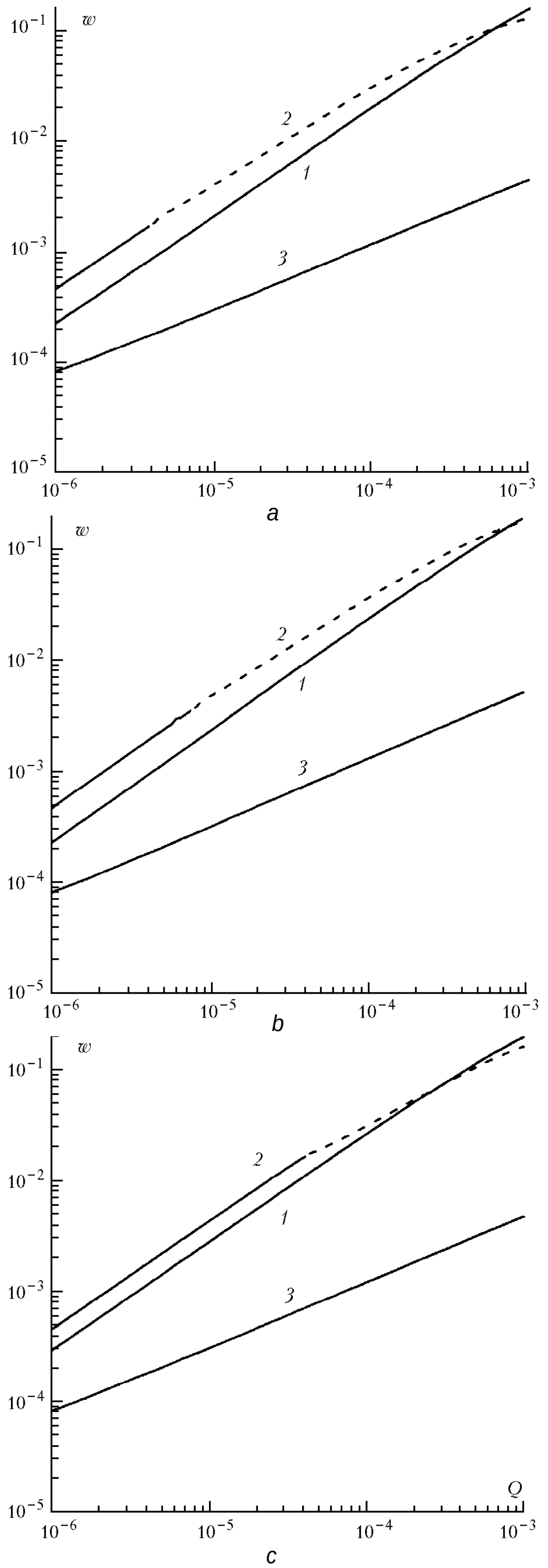

Fig. 2. The $w(Q)$ dependence for three models of interaction of droplets with soot particles. Calculations were made for $R = 50$ μm. (Dashed line shows region of asymptotic calculations according to Ref. 2). $r = 0.005$ μm (*a*), $r = 0.01$ μm (*b*), and $r = 0.05$ μm (*c*).

Upon analysis of calculated results it is found that the smallest value of single scattering albedo (i.e., largest w) is produced by model 2, the largest value by model 3, and model 1 gives a value in between. The literature data available suggest[7] that soot content in the atmosphere depends on its pollution and may range from $10^{\$8}$ to $10^{\$5}$ g / m^3; whereas characteristic liquid water content of stratus clouds[8,9] is of the order of $10^{\$1}$ g / m^3. Hence, the possible values of parameter Q (the ratio of soot to water concentrations) lie in the range $10^{\$7}\$10^{\$4}$. From Figs. 1 and 2 it is seen that, for Q close to $10^{\$5}\$10^{\$4}$, single scattering albedos of 0.999, or even 0.99, can be readily observed, which, by the modern estimates,[1,7] is quite sufficient to reconcile the theory and experiment about absorption by stratus clouds. It is interesting to note that the œdesiredB value of single scattering albedo is produced not only by the œspecialB model 2 of water particle with soot shell, but also by classic model 1 of independently mixed homogeneous water and soot particles. Thus, for sufficiently high, but really possible soot concentration in strongly polluted clouds, model 2, and even model 1 can give single scattering albedos coinciding with those observed in the experiment.

Therefore, the anomalous absorption in strongly polluted clouds can be explained in the framework of classic theory. However, it is still unclear how often such clouds can occur, how far from pollution sources they can travel, and how effective the soot accumulation inside stratus cloud could be.

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