Impact of particle size distribution on the ability of aerosols to act as CCN

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The Goddard Chemistry Aerosol, Radiation, and Transport (GOCART) model shows that the optical depth of the global distribution of aerosols is due to the presence of different chemical components, including sulfates, dust, and sea salt. Each of these three aerosol chemical components is highly concentrated in various areas, such as the East Asia Coast, the Sahara Coast, and the Southern Ocean. Moderate Resolution Imaging Spectroradiometer (MODIS) data was used to study the characteristics of aerosols and cloud condensation nuclei (CCN) in these three ocean regions. The results show that, in all three regions, there is a clear, linear relationship between CCN number concentration and aerosol number concentration. The activation ratio, which is defined as the ratio of CCN to aerosol number concentration, increases with particle size. The results from the theoretical model calculations are consistent with satellite measurements, and they indicate that activation ratio is more sensitive to aerosol size distribution than the solubilities of the various chemical components that make up the aerosols.

1. Introduction

Marine stratocumulus clouds cover approximately 30% of ocean areas at any time (Han, 1994), and their high reflectivity of solar radiation plays an important role in the Earth's energy budget (Ramanathan, 2001). In addition to the existence of proper meteorological conditions, the formation of stratocumulus clouds strongly relies on the existence of aerosol particles (Andreae, 2008). The ability of aerosols to act as cloud condensation nuclei (CCN) is related to their chemical composition and size distribution (Cruz, 1997), and this relationship is studied here by using both satellite measurements and theoretical model calculations.

2. Satellite measurement of the aerosol/CCN relationship

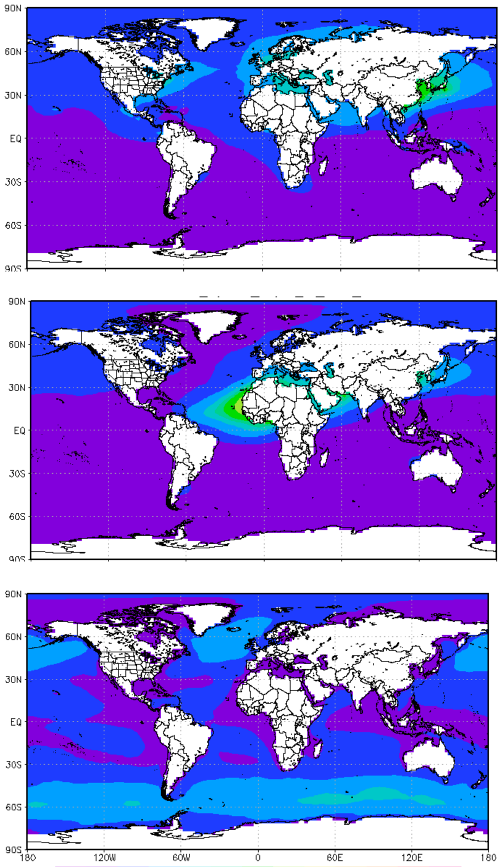

Figure 1. Aerosol optical depth over the ocean due to sulfate (top), dust (middle), sea salt (bottom) from GOCART Model (data: 2000/01-2002/06)

Aerosols have different chemical components (e.g., sulfates, sea salt, dust, and organic and black carbon), and their chemical compositions have spatial variation. In certain regions, the chemical compositions of aerosols can be dominated by specific chemical components. In order to find the regions in which the aerosols have high concentrations of these specific chemical components, we used the results related to aerosol optical depth (AOD) global distribution (Fig.1)

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obtained from the Goddard Chemistry Aerosol Radiation and Transport (GOCART) Model (Chin, 2000). AOD is a radiative property of an aerosol column that describes the extent to which aerosol scattering or absorption will reduce solar radiation before it reaches the surface of the Earth. Generally, AOD is proportional to aerosol mass within the column, and, thus, it is used here to estimate the distribution of the different aerosols. Sulfates are highly concentrated over the East Asia Coast, and AOD due to dust is greatest over the tropical Atlantic due to the dust plume from the Sahara Desert. The highest concentration of sea salt occurs over remote ocean areas, especially the Southern Ocean.

Moderate Resolution Imaging Spectroradiometer (MODIS) on board the NASA/Terra satellite provides important measurements for the study of aerosol-cloud interactions (King, 1992), including column aerosol mass concentration (g/cm^{-2}), aerosol effective radius (μm), and column CCN number concentration (cm^{-2}). In the three ocean regions mentioned above, aerosol and CCN measurements taken by MODIS over a four-month period (June 2009 – September 2009) were analyzed based on the daily average of the grid data.

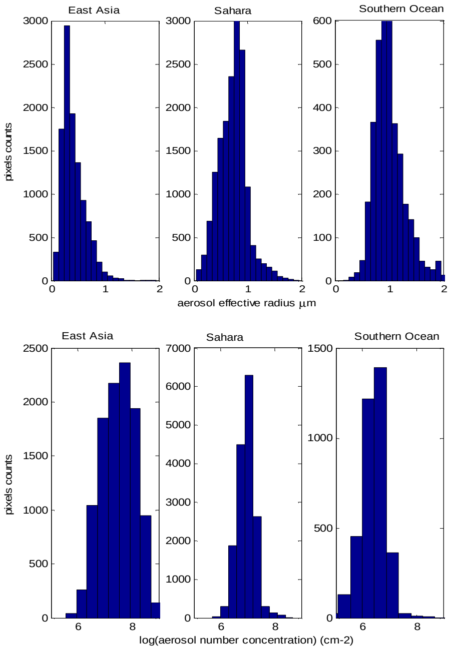

Figure 2. Pixel count distribution of aerosol effective radius (top) and aerosol number concentration (bottom)

The aerosol in East Asia was dominated by sub-micron particles, and particles larger than 1 μm were a larger fraction of the aerosols over the Southern Ocean (Fig. 2, top). Aerosol number concentration (Fig. 2, bottom) was calculated from measured aerosol mass concentration and aerosol effective radius by assuming that the aerosol particles were spherical and that they had a constant density. Aerosol number concentrations were highest over East Asia and lowest over the Southern Ocean.

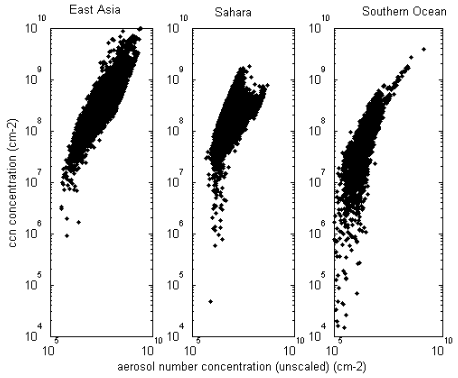

Figure 3. CCN concentration and aerosol number concentration

The relationship between CCN and aerosol number concentration was studied for each region. The results (Fig. 3) show that the CCN number concentration correlates to the aerosol concentration very well. CCN concentration over East Asia was as high as 10^{10} cm^{-2}, which corresponds to the highest aerosol loading there, while the CCN concentration over the Southern Ocean hardly exceeded 10^{9} cm^{-2}.

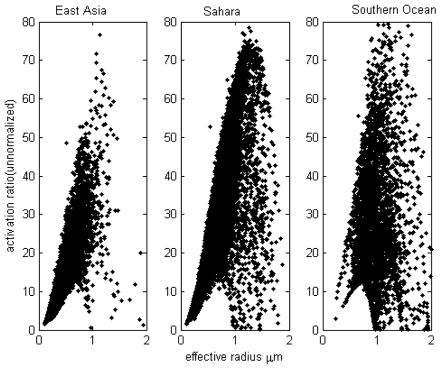

Figure 4. The relationship between activation ratio and the effective radius of the particles

Activation ratio, defined as the ratio of CCN to aerosol number concentration, was calculated to describe the ability of the aerosol to act as CCN. Since aerosol number concentration is not directly measured but is deduced using strong assumptions, the calculated activation ratio sometimes is not smaller than one (unnormalized), so that it can only be taken as an estimation of the ability of the aerosols to act as CCN. The relationship of the activation ratio with particle size (Fig. 4) indicates that the larger particles are more capable of serving as CCN, which corresponds to Köhler's theory and to the model calculations in the next section. This linear relationship is not significant for the particles with radii larger than 1 µm, probably due to the relatively larger variations that are associated with larger sizes.

It was difficult to establish the relationship between the ability of the aerosols to serve as CCN and chemical composition by comparing the data from the three regions. Although these three selected regions have high concentrations of different chemical components (sulfates, dust, and sea salt) and although these chemical components may affect the ability of the aerosols to act as CCN due to their different solubilities (sulfate greatest, dust smallest), the impact of any single component was hard to analyze because the chemical compositions in each region were very complicated. An idealized, theoretical model calculation is described in the next section to understand the effect of chemical composition on the ability of aerosols to act as CCN.

3. Model calculation of aerosol/CCN relationship

A theoretical model calculation (Roberts, 2002) based on classical Köhler's theory was made to study how the relationship between CCN and aerosol number concentration is affected by aerosol size distribution (Kelvin effect) and the chemical compositions of the aerosols (Raoult effect). The size distribution of the aerosol particles was treated as a lognormal distribution and so can be described by median radius and standard deviation. The standard deviation was assumed to be constant in following calculations, and median radius was used to describe aerosol size. In this model, the contribution of the chemical composition of the aerosols was determined from the solubility of each chemical component and its fraction. To simplify the discussion, in the following calculation, we assume that the aerosol is only composed of sulfates (high solubility) and dust (insoluble) and that the fraction of sulfates and dust can change.

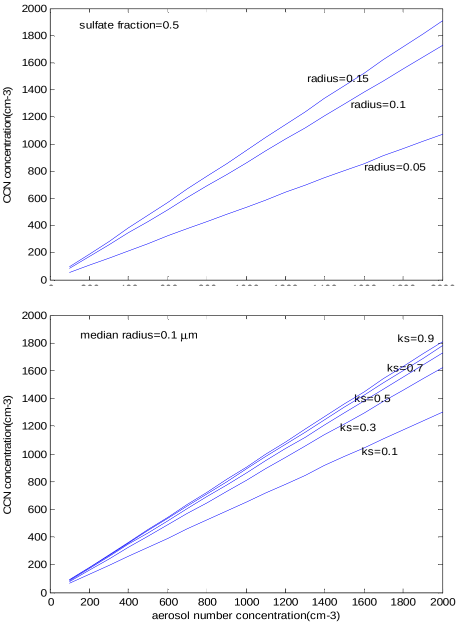

Figure 5. The relationship between CCN and aerosol number concentration for an aerosol with a certain sulfate fraction (top) and median radius (bottom)

The modeled relationship between CCN and aerosol number concentration was calculated assuming the critical supersaturation condition, at which the CCN that is formed is the typical value in the cloud (0.2) (Martin, 1994). The results (Figure 5) show a linear relationship between CCN and aerosol number concentration, similar to the observation from MODIS shown in the previous section. When sulfate fraction is fixed at 0.5 (Fig. 5, top), CCN number concentration increases faster with aerosol number concentration if the particles are larger than 0.1 µm, showing that the ability of aerosols to act as CCN is affected by aerosol size distribution. When the median radius of the aerosol is fixed at 0.1 µm, the impact of the chemical composition of the aerosol is shown (Fig. 5, bottom); a larger fraction of soluble component will increase the ability of the aerosol to act as CCN.

Activation ratio was calculated also using the model to quantify how aerosol size distribution and chemical composition affect the CCN/aerosol relationship (Fig. 6). When the median radius of the aerosol was increased to 0.2 µm, the activation ratio increased to 0.8 - 0.9, and, for the aerosols with higher sulfate fractions, the activation ratio approaches unity more rapidly. With any fixed median radius, activation ratio increases as sulfate fraction increases, but this dependence is only significant when the particles are small; for particles with radii larger than 0.1 µm, the activation ratio approaches unity easily even at low sulfate fraction (0.1), so the activation ratio is less sensitive to sulfate fraction.

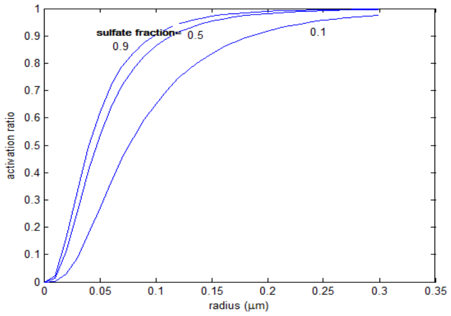

Figure 6. The relationship between activation ratio and particle radius at different sulfate fractions

4. Conclusions

According to Köhler’s theory, activation ratio, as an indicator of the ability of aerosols to act as CCN, is related to aerosol size distribution and chemical composition. Satellite observations clearly show that activation ratio is sensitive to aerosol size distribution, but the dependence of activation ratio on chemical composition is hard to see due to the complicated composition of the aerosol. Even when the chemical composition of the aerosol is simply assumed to be a mixture of sulfate and dust, the model results indicate that the activation ratio is sensitive to the sulfate fraction only when the aerosol particles are smaller than 0.1 µm, indicating that the major impact on the ability of the aerosol to act as CCN results from the size distribution of the aerosol particles.

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