The Transformation of Black Carbon Aerosols from Hydrophobic to Hydrophilic and the Impact on its Global Distribution

Huiyan Yang1 (yang@princeton.edu), L. L. Horowitz2, H. Levy1F
1The Program of Atmospheric and Oceanic Sciences, Princeton University, Princeton, NJ 2GFDL/NOAA, Princeton, NJ Spring 2005 Hawaii Workshop on Air Pollution as a Climate Forcing

Abstract

Most models apply a fixed transformation time of black carbon (BC) aerosols from hydrophobic to hydrophilic. Here a more physically-based parameterization scheme is developed. Three mechanisms are studied, which are the condensation by H2SO4 vapor and coagulation by H2SO4 nuclei, the coagulation with pre-existing water-soluble aerosols, and the interstitial scavenging in clouds. It is found that the transformation is more than 10 times faster in polluted than in remote areas. The Condensation of H2SO4 vapor ranges from the most important to the least important mechanism among the three, depending on the precipitation of the precipitating clouds. The coagulation of H2SO4 nuclei in polluted areas is important only in polluted areas, while the coagulation with cloud droplets is relatively more important in remote areas.

A global chemistry model, MOZART II, is applied to assess the impact of a parameterized transformation time on BC aerosol distribution. The results are compared with simulations using a global uniform transformation time (control run). It is found that by setting the cloud supersaturation at 0.8%, BC aerosol concentration is reduced in polluted areas, and is increased in biomass burning and remote areas by using a parameterized transformation time (parameterized run). The increase is especially prominent in South America and South Africa, the major biomass burning areas, Antarctica, and high atmospheric altitudes. The case by setting the cloud supersaturation at 0.1% produces a weaker reduction, and a much stronger increase, when compared with the control run. A preliminary comparison of the control run with observations indicates that the parameterized transformation time in the parameterized run (0.8% of case) improves the agreement between model and observations.

Model Description

A box model was set up for the study of the role of H2SO4 vapor. The model has four species: H2SO4 vapor, sulfate nucleus, hydrophilic BC aerosols, and pre-existing accumulation mode aerosols. Aerosol microphysical processes included are:

- nucleation and condensation of H2SO4 vapor
- coagulation of sulfate nuclei on hydrophilic BC aerosols and other aerosols.

The production rate of H2SO4 vapor by SO2 oxidation is CH2O in the input of the model. The coagulation with pre-existing water-soluble aerosols and cloud droplets are also studied. A parameterized transformation scheme of the transformation time is developed and applied to a 3D global photrealism model MOZART II.

The chemical mechanism in MOZART II used for this study includes oxidation schemes for the five major tropospheric oxidants, (NMHCs) and heterogeneous reactions of NO2 on BC aerosol.

The global distribution of transformation time in the surface layer for (1) the role of H2SO4, (2) the role of pre-existing aerosols, and (3) interstitial scavenging in clouds is shown in the figure. The transformation by interstitial scavenging (Tt3) varies inversely with the liquid water content (LWC) of clouds. Larger cloud droplets produce a slower transformation. BC aggregates have a slower transformation compared to BC aephasites due to its larger size that is closer to the size of cloud droplets. The global distribution of transformation time in January in the surface layer for (1) the role of H2SO4 vapor condensation (Tt1), (2) the role of pre-existing aerosols (Tt2), and (3) interstitial scavenging in clouds (Tt3) is shown in the graph. The overall transformation time by including all the three mechanisms is shown in the last graph.

Conclusions

- Condensation of H2SO4 vapor is found to be the most important mechanism in the transformation process by assuming a high limit supersaturation. The typical transformation time for this mechanism ranges from about 10 hours to 50 hours in urban and urban-influenced rural areas, and the transformation time in remote areas is on the order of 100 hours or even longer. The impact of this mechanism is reduced tremendously by assuming a low supersaturation of 0.8%.

- When the transformation by interstitial scavenging in clouds is included in the global model, the reduction is strengthened and the increase is weakened, because a faster transformation produces a higher atmospheric load. However, polluted areas in northern hemisphere are affected less than remote areas and high levels, due to the relative importance of clouds in the transformation processes.

- The global modeling of BC aerosol distribution in MOZART II shows a reduction in most polluted areas in northern hemisphere, and an increase elsewhere especially in the biomass burning regions and the polar areas.

Discussions

- Other mechanisms such as the oxidation of organic carbon on the surface of BC aerosols and the ice nucleation of BC aerosols will speed up the transformation process.

- Preliminary comparison with observations indicates that the base of control run, such as the low bias in biomass burning and remote areas and high bias in polluted areas couldn't be lowered by applying the physically-based parameterisation scheme to the transformation time.

References


