On the Dry Deposition of Submicron Particles*

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ON THE DRY DEPOSITION OF SUBMICRON PARTICLES

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1. INTRODUCTION

The air-surface exchange of particles can have a strong role in determining the amount, size, and chemical composition of particles in the troposphere. Here we consider only dry processes (deposition processes not directly aided by precipitation) and mostly address particles less than about 2 \( \mu \text{m} \) in diameter (often referred to as "submicron" particles because most of such particles are less than 1 \( \mu \text{m} \) in diameter). The processes that control the dry exchange of particulate material between the atmosphere and the surface of the Earth are numerous, highly varied, and sometimes poorly understood. As a result, determining how to simulate in modeling the tropospheric mass budget of a particulate substance can be a significant challenge.

Dry deposition, for example, can be controlled by a combination of Brownian diffusion, impaction, interception, and gravitational settling, depending on the size of the particles, the roughness of the surface on both micrometeorological and microscopic scales, the geometrical structure of vegetative canopies, and other surface characteristics such as wetness. Particles can be added to the lower atmosphere by resuspension from land surfaces and sea spray. The roles of rapid gas-to-particle conversion and growth or shrinkage of particles as a result of water condensation or evaporation in the lower few meters of the atmosphere can also have a significant impact on particle concentrations in the lower atmosphere. Here, a few micrometeorological observations and inferences on particle air-surface exchange are briefly addressed.

2. SOME PAST OBSERVATIONS

Different studies have produced widely varying estimates of particle dry deposition velocities (vertical flux density divided by concentration at a specified height). Eddy correlation measurements in the 1980s showed that the deposition velocities for submicron sulfate to short vegetation during the daytime were much larger than originally expected and that the velocities varied more strongly with atmospheric stability than had been predicted on the basis of theoretical and wind tunnel studies (e.g., Wesely et al. 1985; Hicks et al. 1989). The deposition velocities tended to be small at night but increased substantially during the daytime with dynamically unstable conditions near the surface. Micrometeorological measurements above forests in the 1990s confirmed a strong stability dependence for deposition of submicron particles and yielded even larger deposition velocities for sulfate and nitrate than were seen in the 1980s; variations associated with friction velocity, surface wetness, and relative humidity were parameterized (Erisman et al. 1997; Gallagher et al. 1997; Ruijgrok et al. 1997). Although considerable theoretical and modeling research has been done on particle dry deposition (e.g., Slinn and Slinn 1980; Ibrahim et al. 1983; Williams 1982; Davidson et al. 1982; Slinn 1983; Peters and Eiden 1992; Binkowski and Shankar 1995), the results neither fully explain the observations nor always conform to the parameterizations based empirically on the observations.

Results from several eddy correlation experiments in which the fluxes of particles were measured without regard to chemical composition remain difficult to interpret. The observed particle deposition velocities vary greatly for different types of surfaces, ambient conditions, and particle size ranges. The fluxes of particles smaller than about 0.1 mm in diameter have been observed to be directed upward during the daytime and downward at night over lush vegetation (e.g., Wesely and Hicks 1978; Hicks et al. 1989). A possible source of the upward fluxes is rapid gas-to-particle conversion in and just above the vegetative canopy. Other potential confounding processes include the effects of vertical gradients in relative humidity near the surface in changing size distributions (DePaul et al. 1987; Lee and Wesely 1989) and suspension of fine particulate material from dry surfaces (Wesely et al. 1983). Very few measurements of the vertical flux of submicron particles have been made over natural water bodies. In some cases, fluxes unrealistically large in magnitude have been seen, possibly as a result of sea spray or the rapid changes in particle size in the presence of strong vertical gradients in humidity (Wesely et al. 1982; Lee and Wesely 1989).

3. FUTURE OBSERVATIONS NEEDED

The theoretical interpretation of measurements of vertical particle fluxes would be facilitated if measurements were made according to both size and chemical species of particles, especially for micrometeorological techniques such as eddy correlation and gradient methods. Parameterizations of the deposition velocity for particulate sulfate and, to a lesser extent, nitrate have been developed for aerodynamically rough surfaces as a result of field observations, but the parameterizations are somewhat empirically based and are not easily applied to other particulate substances whose particulate size distributions deviate significantly from that for sulfate. Field observations of particle deposition to relatively smooth surfaces are very rare, but they could be carried out to a limited extent with existing techniques. Major efforts would be needed to study quantitatively the role of rapid gas-to-particle conversion in generating vertical fluxes near vegetation that emits reactive compounds and the effects of humidity variations near the surface on the particle size and chemical composition of the particles.

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4. REFERENCES


