Aerosol Deposition Measured by Eddy-Correlation Mass Spectrometry

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Background and Motivation

Dry deposition is an important mechanism for the removal of aerosol particles from the atmosphere and for the addition of materials to terrestrial ecosystems. This may cause a significant perturbation of natural biogeochemical cycles, contributing to extratropical marine and terrestrial bodies and to nutrient loading of sensitive ecosystems (Figure 1).

![Simplified diagram of the Nitrogen cycle.](image)

Deposition rate is highly uncertain and the literature includes a wide range of particle deposition velocity estimates. Semi-empirical models of deposition velocity have been developed from wind tunnel experiments and a number of research groups have measured aerosol fluxes using optical particle counters (e.g., Gallagher et al., 1997). Measured deposition velocity values differ from predicted values by an order of magnitude (Figure 2).

![Particle deposition velocity as a function of size to forest canopies.](image)

Eddy-Correlation Technique

The well-known eddy-correlation technique can be used to calculate the turbulent flux of solute quantities, S, such as nutrients, heat, gaseous species, and particulate species. From time series measurements of the scalar concentration, C, and wind speed, u, the turbulent flux, \( F = \rho C u' \), can be estimated from the eddy correlation technique. The covariance of the vertical wind velocity, v, with S, \( \rho' v = < S S > / < S > \), is the turbulent vertical flux.

![Schematic diagram of the Aerodyne Aerosol Mass Spectrometer (AMS).](image)

Eddy-Correlation Mass Spectrometry

Aerosol concentrations were measured using an Aerodyne Aerosol Mass Spectrometer (AMS) (Yue et al., 2000). In the AMS, vacuum aerodynamic diameter (\( D_{\text{vac}} \)) was measured by particulate time-of-flight (tof) transmission efficiency (100% for \( D_{\text{vac}} = 40 \)–700 nm) chemical composition was determined by flash desorption of the non-volatile (\( D_{\text{p}} < 300 \text{nm} \)) components, which are then ionized by electron impact and detected using a quadrupole mass spectrometer. The AMS signals were validated using a sonic anemometer to measure wind velocity and direction at 10 Hz. Aerosol deposition velocities for 30-min periods were calculated as the covariance of the vertical wind and the AMS signal, divided by the average of the AMS signal

\[ F = \rho C u' \]

At the time response required for the eddy-correlation technique, the AMS is capable of measuring one ion species at a time. During the study, three different species were measured for 1-3 day periods.

![Schematic diagram of the Aerodyne Aerosol Mass Spectrometer (AMS).](image)

Results and Discussion

The field experiment was conducted during the Prognosis Program for Research on Aerosol Photochemistry, Emission, and Transport in August 2001. The site is located in a mixed deciduous/coniferous forest near Pellston, Michigan.

![PROPHET towers at the University of Michigan Biological Station.](image)

Cyclone

11 L/min

(2.1 μm)

AMS

0.09 L/min

![Cyclone sample, cascade anemometer, and CO\textsubscript{2}/H\textsubscript{2}O analyzers on the PROPHET tower (left). Air sample flow schematic diagram (right).](image)

Momentum and sensible heat fluxes were calculated using the eddy-correlation technique. Both fluxes show a strong diurnal pattern associated with increases in daytime solar heating and wind velocity. Momentum and sensible heat fluxes (Figures 4 & 7) are in agreement with published values for similar sites (Molden, 1998; Schmid et al., 2000).

![Schematic diagram of the Aerodyne Aerosol Mass Spectrometer (AMS).](image)

Deposition velocities measured by eddy-correlation mass spectrometry are within the range of published results. Differences in the approach deposition velocities appear to be due to changes in meteorological conditions, especially for sulfuric particles measured during two different time periods. The results presented are the first to report measurements of particulate fine aerosol deposition velocities and can be used to evaluate parameterizations of deposition velocity.

Conclusions

Deposition velocities measured by eddy-correlation mass spectrometry are within the range of published results. Differences in the approach deposition velocities appear to be due to changes in meteorological conditions, especially for sulfuric particles measured during two different time periods. The results presented are the first to report measurements of particulate fine aerosol deposition velocities and can be used to evaluate parameterizations of deposition velocity.

Deposition to the CAP Ecosystem

Plant are currently undergoing to conduct a field study in the Phoenix, Arizona metropolitan area. We plan to deploy the Aerosol Mass Spectrometer (AMS) and monitor aerosols in an agricultural field at the Salt River Indian Community to collect eddy-correlation mass spectrometry data. Utilize data from this study can be used to improve on HY deposition estimates for the CAP biogeochemical research program.

![Schematic diagram of the Aerodyne Aerosol Mass Spectrometer (AMS).](image)

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![Schematic diagram of the Aerodyne Aerosol Mass Spectrometer (AMS).](image)

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![Schematic diagram of the Aerodyne Aerosol Mass Spectrometer (AMS).](image)

Figure 1: Okeham length (L) for 30-min periods in August 2001.

The Okeham length (L) is a parameter characteristic of the mixing in the planetary boundary layer. Deposition Okeham length (Figure 8) are negative, indicating unstable conditions in which buoyant flows dominate over mechanical shear production of turbulence. Note that the buoyant driven mixing is lower on Aug. 3-4 than the remaining days. This is consistent with low deposition velocities observed on Aug. 3-4 in comparison with the other days.

![Schematic diagram of the Aerodyne Aerosol Mass Spectrometer (AMS).](image)

The median vacuum aerodynamic diameter (\( D_{\text{vac}} \)) of the AMS signal varies little during study period (Figure 9). While particle size is important in determining deposition velocity, these results do not explain the wide variation in deposition velocities shown in Figure 8.

![Schematic diagram of the Aerodyne Aerosol Mass Spectrometer (AMS).](image)

The characteristic deposition velocities were 0.25 cm/s for sulfur (Aug. 3-4), 1.0 cm/s for organic compounds (Aug. 3), 1.2 cm/s for nitrate (Aug. 6-7), and 1.1 cm/s for oxidized (Aug. 7-10). The range of deposition velocity values measured by eddy-correlation mass spectrometry is in agreement with those in the literature. The deposition velocity for Aug. 3-4 is an order of magnitude lower than those for the remaining days sampled.

![Schematic diagram of the Aerodyne Aerosol Mass Spectrometer (AMS).](image)

Figure 3: PROPHET towers at the University of Michigan Biological Station. Tower height is 31 m, forest canopy height is approx. 20 m.

Figure 4: Schematic diagram of the Aerodyne Aerosol Mass Spectrometer (AMS).