Aerosol Deposition Measured by Eddy-Correlation Mass Spectrometry

Daniel A. Gonzales[1], Jonathan O. Allen[1,2]

[1]Chemical and Materials Engineering, Arizona State University; [2]Civil and Environmental Engineering, Arizona State University

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 downwind ecosystems. This may cause a significant perturbation of natural biogeochemical cycles, contributing to eutrophication of water bodies and to nutrient loading of sensitive ecosystems (Figure 1).

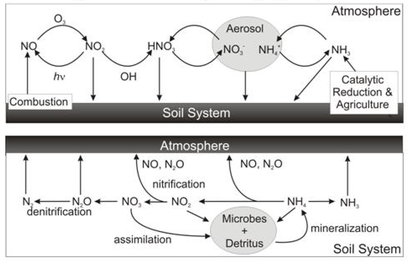

Figure 1: Simplified diagram of the Nitrogen cycle.

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).

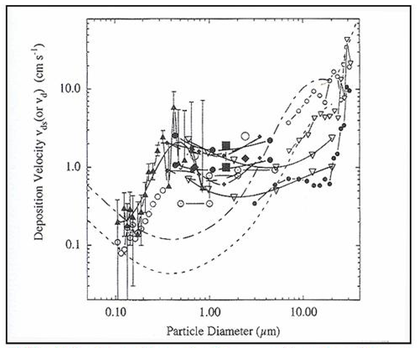

Figure 2: Particle deposition velocities as a function of size to forest canopies (Gallagher et al., 1997).

Eddy-Correlation Technique

The well known eddy-correlation technique can be used to calculate the turbulent flux of scalar quantities, S, such as momentum, heat, gaseous species, and particulate species. From time series measurements of the scalar of interest, the mean, <S>, is calculated over an averaging period. The deviation, S', is calculated by subtracting the mean from each instantaneous measurement. The covariance of the vertical wind velocity, w, with S, <w'S'> is the turbulent vertical flux.

Eddy-Correlation Mass Spectrometry

Aerosol concentrations were measured using an Aerodyne Aerosol Mass Spectrometer (AMS) (Jayne et al., 2000). In the AMS, vacuum aerodynamic diameter (D_{va}) was measured by particle time-of-flight (transmission efficiency ~100% for D_{va} = 40 - 700 nm); chemical composition was determined by flash vaporization of the non-refractory (NR) components, which are then ionized by electron impaction and detected using a quadrupole mass spectrometer.

Co-located with the AMS inlet was a sonic anemometer to measure wind velocity and direction at 10 Hz. Aerosol deposition velocities for 30-min periods were calculated directly as the covariance of the vertical wind with the AMS signal, divided by the average of the AMS signal:

$$v_d = -\langle w'S' \rangle / \langle S \rangle$$

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

Species	m/z	Characteristic Ion
Sulfate	64	SO_2^+
Organic	57	$C_4H_9^+$
Nitrate	46	NO_2^+

PROPHET 2001

The field experiment was conducted during the **P**rogram for **R**esearch on **O**xidants: **PH**otochemistry, **E**missions, and **T**ransport experiment in August 2001. The site is located in a mixed deciduous/coniferous forest near Pellston, Michigan.

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

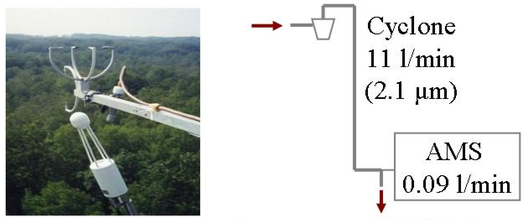

Figure 4: Cyclone sampler, sonic anemometer, and CO_2/H_2O analyzer on the PROPHET tower (left). Air sample flow schematic diagram (right).

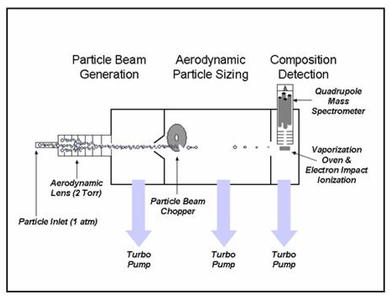

Figure 5: Schematic diagram of the Aerodyne Aerosol Mass Spectrometer (AMS).

Results and Discussion

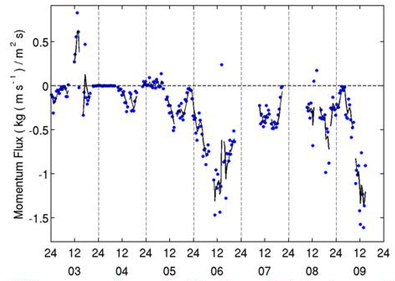

Figure 6: Momentum fluxes for 30-min periods in August 2001.

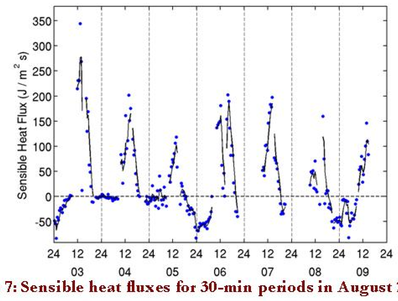

Figure 7: Sensible heat fluxes for 30-min periods in August 2001.

Momentum and sensible heat fluxes were calculated using the eddy-correlation technique. Both fluxes show a strong diurnal pattern associated with increased daytime solar heating and wind velocity. Momentum and sensible heat fluxes (Figures 6 & 7) are in agreement with published values for similar sites (McMillen, 1988; Schmid et al., 2000).

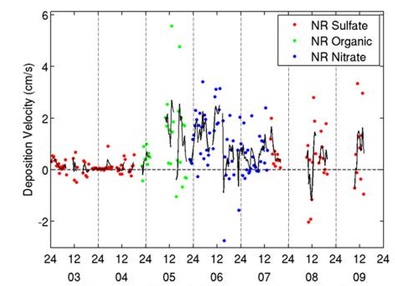

Figure 8: Deposition velocities (v_d) for 30-min periods in Aug 2001.

The characteristic deposition velocities were 0.25 cm/s for sulfate (Aug 3-4), 1.9 cm/s for organic compounds (Aug 5), 1.2 cm/s for nitrate (Aug 6-7), and 1.1 cm/s for sulfate (Aug 7-9). The range of deposition velocity values measured by eddy-correlation mass spectrometry are in agreement with those in the literature. The deposition velocities for Aug 3-4 are an order of magnitude lower than those for the remaining days sampled.

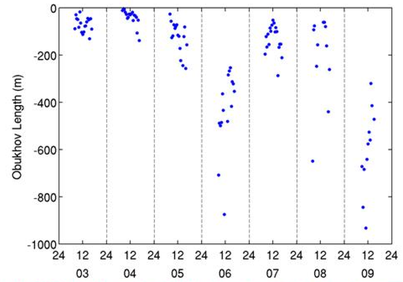

Figure 9: Obukhov length (L) for 30-min periods in August 2001.

The Obukhov length (L) is a parameter characteristic of the mixing in the boundary layer. Daytime Obukhov lengths (Figure 9) are negative, indicating unstable conditions in which buoyant forces dominate over mechanical shear production of turbulence. Note that the buoyancy 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.

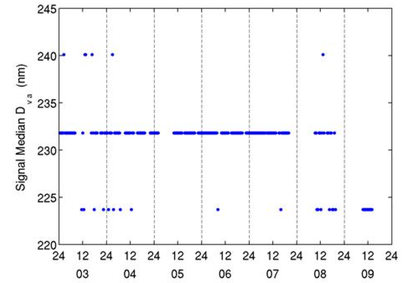

Figure 10: Signal median diameter for 30-min periods in August 2001.

The median vacuum aerodynamic diameter (D_{va}) of the AMS signal varies little during study period (Figure 10). While particle size is important in determining deposition velocity, these results do not explain the wide variation in deposition velocities shown in Figure 8.

Conclusions

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

Deposition to the CAP Ecosystem

Plans are currently underway to conduct a field study in the Phoenix, Arizona metropolitan area. We plan to deploy the Aerosol Mass Spectrometer (AMS) and sonic anemometer in an agricultural field at the Salt River Indian Community to collect eddy-correlation mass spectrometry data. Nitrate data from this study can be used to improve upon N dry deposition estimates for the CAP biogeochemical research program.

Figure 11: Proposed sampling site at the Salt River Indian Community.

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