Dry Deposition of Fine Aerosol Nitrogen to an Agricultural Field Measured by Eddy-Correlation Mass Spectrometry

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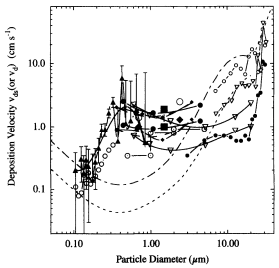
Background and Motivation

Dry deposition is an important mechanism for the removal of aerosol particles from the atmosphere, often accounting for more than half of the total deposition of particulate nitrate (Lovett, 1994). This may cause a significant perturbation of natural biogeochemical cycles, contributing to eutrophication of water bodies and to nutrient loading of sensitive ecosystems. Measured and predicted deposition velocity values often differ by an order of magnitude.



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

Experimental Site

The field experiment was conducted at an agricultural site in the Phoenix, Arizona, metropolitan area at the Salt River Pima-Maricopa Indian Community in January and February 2005. The site is a level agricultural field 0.8 km square. The crop was mature broccoli with a mean height of 0.5 m, which was flood irrigated once per week during the study.

Sampling tower at the agricultural field site with a homogeneous fetch of 0.8 km. The tower height is 7 m and the mean crop height is 0.5 m. The mountain in the background is about 12 km to the west.

References

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Jayne, J.T., Leard, D.C., Zhang, X., Davidovits, P., Smith, K.A., Kolb, C.E., and Worsnop, D.R., 2000. Development of an Aerosol Mass Spectrometer for Size and Composition Analysis of Submicron Particles. *Aerosol Sci. Tech.* 33:49-70.

Lovett, G.M., 1994. Atmospheric Deposition of Nutrients and Pollutants in North America: An Ecological Perspective. *Ecol. Appl.* 4(4):629-650.

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- Henry Guzman, Rousseau Ranch

Method: Bulk Deposition Collectors

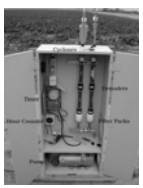
Bulk deposition collectors consist of two buckets: one is exposed to the atmosphere while the other is covered during periods without precipitation (dry deposition). A rain sensor triggers a motor which moves the cover during rain events to collect precipitation (wet deposition).

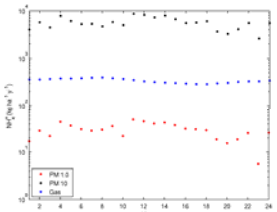
Bulk Samples	Sample Volume (ml)	Flux NO_3^- (kg ha^{-1} y^{-1})	Flux NH_4^+ (kg ha^{-1} y^{-1})
1/18 – 1/24 Wet	144.6	0.64	1.74
1/24 – 1/28 Wet	988	0.94	5.00
1/28 – 2/1 Wet	76.2	0.45	0.86
2/1 – 2/14 Wet	3073	1.11	3.19
1/18 – 2/14 Dry	500	0.58	0.23

Bulk wet and dry deposition results.

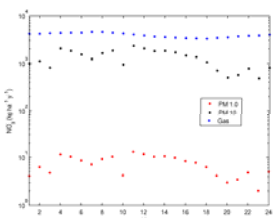
Method: Filter and Denuder Banks

Atmospheric fluxes were inferred from filter and denuder bank measurements of gas and particle (PM 1.0 and PM 10) phase concentrations and deposition velocities calculated from the US EPA's Community Mulitscale Air Quality (CMAQ) modeling system.





Mean ammonia dry deposition flux Jan 19, Jan 25, & Feb 1.



Mean nitrate dry deposition flux Jan 19, Jan 25, & Feb 1.

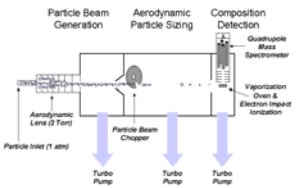
The deposition velocities for PM 1.0 and PM 10 particles differ by two orders of magnitude, resulting in large differences in flux estimates. Gas phase deposition velocities are strongly dependent on chemical species.

Method: Eddy-Correlation Technique

The eddy-correlation technique can be used to calculate the turbulent flux, F, of scalar quantities, S, such as momentum, heat, gaseous species, and particulate species. From time series measurements of the scalar, the mean (denoted by overbars) is calculated over an averaging period. The deviation (denoted by primes) is calculated by subtracting the mean from each instantaneous measurement. The covariance of the vertical wind velocity is the turbulent vertical flux

$$F = \overline{w'S'}$$

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.



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

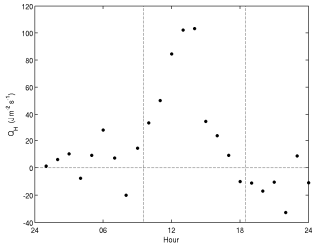
Co-located with the AMS inlet was a sonic anemometer to measure 3-dimensional wind velocities at 10 Hz. Aerosol deposition velocities for 30-min periods were calculated directly as the covariance of the vertical wind, w, with the AMS signal, S, divided by the average of the AMS signal

$$v_d = -\frac{\overline{w'S'}}{\overline{S}}$$

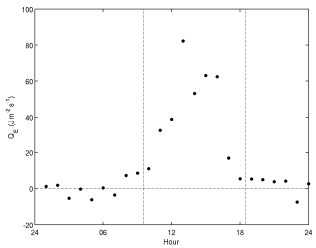
In eddy-correlation mode, the AMS measures one ionic species at a time. Three ions characteristic of environmentally relevant species were monitored during the experiment.

Species	m/z	Characteristic Ion
Sulfate	64	SO_2^+
Organic	43	$C_3H_7^+$
Nitrate	30	NO^+

Eddy-Correlation Results

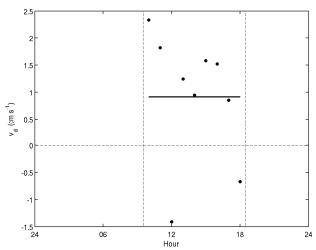


Ensemble mean daily sensible heat flux Jan 16 – Feb 1, 2005.



Ensemble mean daily latent heat flux Jan 16 – Feb 1, 2005.

Sensible and latent eddy heat fluxes were calculated for 30-min averaging periods, then ensemble averaged for each hour of the day. The heat fluxes show a strong diurnal cycle, reaching maximums in the early afternoon, demonstrating increasing vertical mixing during midday.



Ensemble mean daily deposition velocities Jan 16 – Feb 1, 2005.

Dry deposition velocities were calculated for 30-min periods during periods of unstable atmospheric conditions. The ensemble mean deposition velocities for nitrate and organic species for the entire study are in the range –1.5 to 2.5 cm s^{-1} during unstable periods. The mean of the ensemble deposition velocities is 1 cm s^{-1}.

Conclusions

Bulk collectors sample both wet and dry deposition on a surrogate surface. Estimates of nitrogen deposition based on bulk collector results are prone to errors due to the inclusion of foreign materials and volatilization loss of aerosol species. In addition, it is difficult to interpret dry deposition estimates since the buckets are not representative of natural deposition surfaces, such as leaves and soils.

Filter and denuder banks measure the concentration of gas and particle phase nitrogen species, eliminating sampling biases associated with volatile aerosol species. The resulting concentrations provide a method to estimate nitrogen dry deposition fluxes using the inferential method.

Modeled deposition velocities were used to estimate dry deposition fluxes based on readily available meteorological data. This method is limited by the state of the art of deposition velocity modeling. Several models exist for estimating gas and particle deposition velocities, however, there is a two-order of magnitude difference between modeled and measured deposition velocities that is yet to be reconciled.

The eddy-correlation mass spectrometry technique has been demonstrated to directly measure dry deposition velocities of fine aerosol particles. For the particle size range measured (40 - 700 nm), the deposition velocity measured by eddy-correlation mass spectrometry is 1 cm s^{-1}, in agreement with published measurement results. This method is the first to measure speciated and size-segregated particle deposition velocities.