

The Relaxed Eddy Accumulation for Estimating Aerosols Dry Deposition above Tropical Forest

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Abstract The relaxed eddy accumulation (REA) method was applied to estimate dry deposition fluxes and velocities of SO_4^{2-} and NO_3^- above a tropical forest. The monthly measured concentration of NO_3^- was found to be greater than that of SO_4^{2-} . The average dry deposition flux of NO_3^- was found to be greater than that of SO_4^{2-} . The average deposition velocities of sulfate were evaluated to be 0.30 cm s^{-1} during the day and 0.22 cm s^{-1} at night, while the highest deposition velocities of nitrate were 0.93 cm s^{-1} during the day and 0.54 cm s^{-1} at night. The deposition velocities of aerosols (SO_4^{2-} and NO_3^-), calculated by using the resistance model, were found to be 0.49 cm s^{-1} during the day and 0.29 cm s^{-1} at night.

Keywords: aerosols dry deposition, relaxed eddy accumulation, tropical forest

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1. Introduction

Acid deposition is a trans-boundary environmental problem. The transfer of strong acids and acid forming substances from the atmosphere to the Earth has become a critical stress affecting forested landscapes. Atmospheric aerosols containing sulfate and nitrate are secondary pollutants which result from chemical reactions of sulfur dioxide and nitrogen oxide to form sulfate and nitrate in association with other constituents of the atmosphere such as fine particles and water. Dry deposition of such aerosols has an important and direct impact on forests and agricultural areas, as well as on soil and water in various ecosystems [1].

Dry deposition fluxes of aerosols, monitored on natural surfaces, have been measured or estimated using a variety of methods and techniques. An alternative way of measuring fluxes by micrometeorological methods uses empirical relationships between fluxes and gradients of quantities measured in the inertial sublayer [2]. The relaxed eddy accumulation (REA) method is one of several techniques that directly measures turbulent transport of air pollution. It is a micrometeorological technique that can be used to measure the air-surface exchange of atmospheric pollutants [3]. Some of the current methodologies used for measuring concentrations of aerosol sulfate and nitrate are based on accumulation devices such as annular denuders and filter packs, in which concentrations can be measured very accurately. In many operational air quality models, dry deposition of

atmospheric aerosols is quantified as the product of the modeled concentration and value of deposition velocity that is often modeled using the concept of the resistance analogue. A comparison of several models revealed that they differ from each other greatly, with the largest uncertainty being the size range of aerosols.

The objective of this study is to measure the dry deposition flux of sulfate and nitrate over a deciduous forest using the relaxed eddy accumulation method and to utilize the data to estimate dry deposition velocities. Deposition velocities measured in the field were compared with values predicted by the model used.

2. Methodology

2.1. Experimental Method

The experiment was conducted above the deciduous forest which is located adjacent to the King Mongkut's University of Technology Thonburi, Ratchburi province in Thailand at latitude $13^\circ 35' 13.3''\text{N}$, Longitude $99^\circ 30' 3.9''\text{E}$. The dominant canopy tree species are *Dipterocarpus obtusifolius*, *D. tuberculatus*, *Shorea obtuse*, and *S. siamensis*. This is a regenerated forest. Most of its trees are young (5-7 years old). The height of the trees ranges from about 5 to about 7 meters. The experiment was conducted from June of 2009 to May of 2010. The climatic condition can be classified as the Summer season (February-October), the rainy season (May-October) or the Winter season (November-January). The aerosol concentrations were collected by two four-stage filter packs with the REA system positioned at a

height of 10 meters on the micrometeorological tower. A vacuum pump (GAST model DOA-P504- BN) was used to draw in air samples. The temperature and relative humidity were measured by a Wisco-HT120 sensor, also at a height of 10 meters. The 3D ultrasonic anemometer was also located at a height of 10 meters above the ground.

A flow rate of 20 L min⁻¹ for air samples was continuously drawn through the 4-stage filter pack by a vacuum pump. The collection time was set for every 6 hours (06.00-12.00 (morning), 12.00-18.00 (afternoon), 18.00-24.00 (evening) and 24.00-06.00 (night)) for three days per month. The 4-stage filter pack, starting from the inlet, consisted of a Teflon filter (Pall) to collect SO₄²⁻ and NO₃⁻, a nylon membrane to collect gases, potassium carbonate impregnated cellulose (Whatman, No. 41) to collect gases, and phosphoric acid impregnated paper to collect NH₃. This research focused on SO₄²⁻ and NO₃⁻ only. The collected filters were then placed in a polyester bag and preserved in a refrigerator. To analyze the chemical species, each filter was placed in a separately put in 50 ml polypropylene tubes which was then filled with up to 20 ml of distilled water. Twenty ml of 0.1% V/V H₂O₂ was added to the second-stage filter. The tubes were shaken for 20 minutes in an ultrasonic shaker. The solution was then filtered using a membrane 0.45 micrometer filter and preserved in a refrigerator at 4°C. Ion chromatography was used to analyze the anion species of SO₄²⁻ and NO₃⁻. The quality assurance/quality control (QA/QC) program for the air concentration monitoring in East Asia was performed at the regional laboratory, King Mongkut's University of Technology Thonburi, Bangkok, Thailand.

2.2. Theory of Relaxed Eddy Accumulation

The relaxed eddy accumulation method is applied to chemical species for which a fast response instrumentation is not yet available [4,5,6,7]. The main instrument used consists of a 3D ultrasonic anemometer, a vacuum pump and two filter packs. The method employs isolated filter reservoirs to sample vertical wind eddies based on two directions "updraft" (+w) or "downdraft" (-w). Wind velocities are measured using the 3D ultrasonic anemometer. (and the vertical wind exceeds a threshold (dead - band) velocity [8]. The vertical flux of sulfate and nitrate is determined by

$$F = \beta \sigma_w (\bar{C}_{up} - \bar{C}_{down}) \quad (1)$$

where

$$\beta = \frac{\overline{w'T'}}{\sigma_w (T^+ - T^-)} \quad (2)$$

β is calculated using a probability distribution of vertical wind speed and sensible heat fluxes. σ_w is the standard deviation of the vertical wind velocity, and \bar{C}_{up} and \bar{C}_{down} are the average concentrations of pollutant in the updraft and downdraft direction, respectively. In this study we applied a deadband of ± 0.5 where is computed as a running mean over a period of time. The deadband is the pre-set value of the vertical wind velocity where velocities below the set are rejected. The deadband is controlled by a microcontroller and C++ program which orders the solenoid valves to open or close the inlet line (through the reservoirs line and the deadband line. The switching

frequency between updraft, downdraft and deadband was 1 second. This deadband will cut off the lower threshold of the vertical wind around zero which significantly reduces the frequency of valve switching, and increases the ambient concentration difference [9].

The direct measurements of vertical flux near the surface can be used to characterize dry deposition velocity (V_d). This velocity is commonly used in parameterization of dry deposition flux or rate (F) and the concentration (C) of an airborne pollutant. It can be expressed as

$$V_d = \frac{F}{C} \quad (3)$$

The dry deposition velocity, in Eq. 3 is an observed value which depends on the local micrometeorological conditions.

2.3. The Resistance Model

In the trans-boundary transport of pollutants, the value of V_d of an atmospheric particle, used by the model, is generally computed from the resistance model as shown below [10]

$$V_d = \frac{1}{R_a + R_b + (R_a R_b V_t)} + V_t \quad (4)$$

where V_t is the terminal settling velocity of a particle

$$V_t = \frac{D_p^2 \rho_p g C}{(18\mu)} \quad (5)$$

Here, D_p is the particle diameter, ρ_p is the particle density, g is acceleration due to gravity, μ is the absolute viscosity of air and C is the Cunningham slip correction factor for small particles given by

$$C = 1 + \frac{2\lambda}{d_p} (1.257 + 0.4e^{-0.55 \frac{d_p}{\lambda}}) \quad (6)$$

where λ is the mean molecular free path of air.

The aerodynamic resistance is calculated by

$$R_a = \frac{\ln \left[\frac{z_{ref} - d}{z_0} \right] - \Psi_H}{K u_*} \quad (7)$$

Where d is the reference height at which V_d is calculated, z is the surface roughness length, K is von Kaman's constant (≈ 0.41), u* is friction velocity and Ψ_H is dimensionless stability function for heat.

The quasi-laminar boundary layer resistance is calculated by

$$R_b = \frac{\ln \left[\frac{z_0}{z_{0c}} \right]}{K u_*} \quad (8)$$

Where z_{0c} is a secondary sink height or the height of the canopy [11]. The parameters infer in the resistance model will employ the measured values from the field experiment. The deposition velocity obtained by the resistance model method will be investigated and compared with the value obtained by the REA method.

3. Results and Discussion

3.1. Measurements

The monthly average concentrations of SO_4^{2-} and NO_3^- are shown in Figure 1. The average concentration of nitrate was observed to be higher than that of sulfate for each month, except April. The highest concentrations of nitrate occurred during the dry season (Nov 09 to Jan10) when certain anthropogenic activities and events, such as open/biomass burning and wild fires occurred with greater frequency. Near the site, that there are farming activities and animal husbandry, such as swine production and dairy production, that produce NH_3 from animal waste. The NO_3^- can be produced by the gas phase reaction of HNO_3 with NH_3 to form NH_4NO_3 .

The dry deposition fluxes were calculated by Eq. 1 in which the turbulent flux is proportional to the standard deviation of vertical velocity and the difference between mean concentrations under conditionally sampling during updraft and downdraft wind flows. The β coefficient is the slope of the linear line (Eq. 2) and determined to be 0.49. The monthly variation of dry depositions of SO_4^{2-} and NO_3^- are shown in Figure 2. The average dry deposition flux of NO_3^- was larger than SO_4^{2-} . This indicated a great contribution of NO_3^- to acid deposition. Nitrogen oxides, such as NO and NO_2 , can be transformed to NO_3^- in the atmosphere by photo-oxidation reactions with fine particles in intense sunlight. NO_2 can also form HNO_3 in the atmosphere by reaction with OH radical [12].

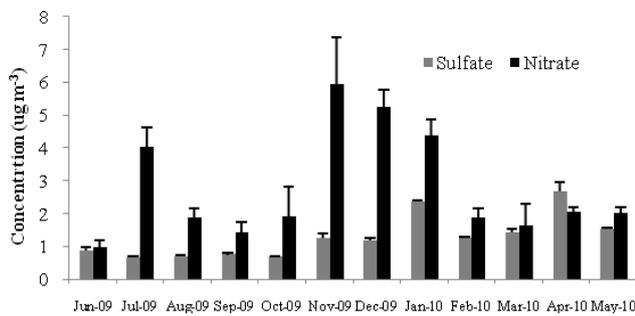


Figure 1. Monthly average concentrations of SO_4^{2-} and NO_3^-

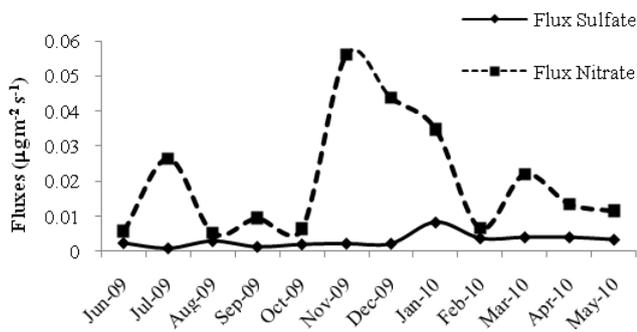


Figure 2. Monthly average fluxes of SO_4^{2-} and NO_3^-

3.2. Model-Measurement Comparison

The observation of deposition velocity is based on an assumed steady-state relationship $V_d = F/C$, where dry deposition flux or rate (F) is a product of the dry deposition velocity (V_d) and the concentration (C) of an airborne pollutant. The resistance model involves indirect estimations of dry deposition rates on the basis of

routinely measured air concentrations and meteorological parameters. The deposition velocity is estimated on the basis of resistance models and is defined as the inverse of the sum of aerodynamic resistance (R_a), boundary-layer resistance (R_b), and where V_t is the terminal settling velocity of the particle. The dry deposition velocity can be estimated from its conventional definition as the ratio of the vertical flux of a material at or near the surface of the earth to the mean concentration of the material at a reference height. The deposition velocities for sulfate and nitrate were calculated using Eq. (3) and the resistance model by Eq. (4) in the daytime and nighttime. Aerosols size according to a preliminary determination by electrical low pressure impactor was assumed to be $0.1 \mu\text{m}$ for calculation of terminal settling velocity. A comparison of the V_d values calculated using the model vs. measured values is shown in Figure 3. The average deposition velocities of sulfate were 0.30 cm s^{-1} during the day and 0.22 cm s^{-1} at night, while the highest deposition velocities of nitrate were 0.93 cm/s during the day and 0.54 cm s^{-1} at night. The deposition velocities of aerosol were calculated by using the resistance model to be 0.49 cm s^{-1} during the day and 0.29 cm s^{-1} at night. The trend of depositions in the daytime was larger than the nighttime in both modeled estimation and observation due to the high atmospheric turbulence. The observed nitrate deposition velocity was higher than the observed sulfate deposition velocity. This is likely due to the larger size of nitrate particles relative to sulfate particles. Zhang et. al. [13] found the size distribution of nitrate was to be $4\text{-}7 \mu\text{m}$, while the size distribution of sulfate is $0.3\text{-}0.6 \mu\text{m}$. Thus, it appears that the dry deposition velocity of particles depends significantly on their diameters. Some studies found that most sulfate, nitrate and ammonium particles fall within the $0.1\text{-}1.0 \mu\text{m}$ size range [14]. However, dry deposition velocities exhibit a pronounced dependency on surface types, in particular with regard to the roughness of various surface structures. For example, dry deposition velocities in the range of 0.01 to 0.02 cm/s are found over grasslands, while velocities in the range of $0.1\text{-}1 \text{ cm s}^{-1}$ are found over forests [8]. Model validation for aerosol deposition is of concern in acid deposition. Current measuring equipment is not able to detect small changes in concentration rapidly enough to detect or to measure the deposition phenomena.

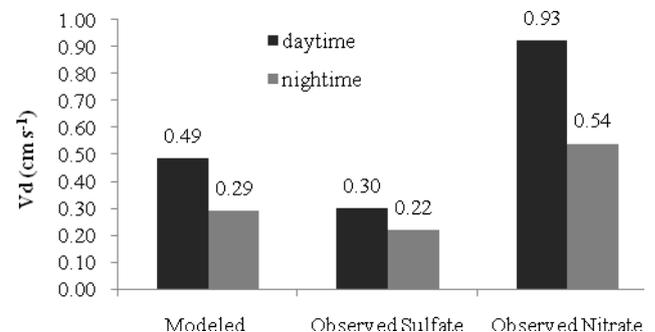


Figure 3. Comparison of V_d values calculated using the resistance model vs. observed values

Figure 4 shows the high correlation between the deposition velocity and the friction velocity (u^*) ($R^2 = 0.914$). The friction velocity, as an atmospheric turbulence mechanism, can influence the deposition velocity simultaneously. Gaman et. al. [15] applied a relaxed eddy accumulation method to estimate the number flux of nm

particles above the Scots Pine forest and found that the deposition velocity of 50 nm particles increased with friction velocity from approximately 5 mm s^{-1} at friction velocities $< 0.5\text{--}15 \text{ mm s}^{-1}$ for friction velocities $> 1.0 \text{ ms}^{-1}$. The sensible heat flux is also a very important parameter of the atmospheric turbulence because of its thermal properties. A similar correlation for the deposition velocity and the sensible heat flux is shown in Figure 5. This study also shows a good correlation of $R^2 = 0.855$ which clearly shows the effect of sensible heat flux on the dry deposition values. Accordingly, the deposition velocity increases with physical turbulence and thermal turbulence. The result obtained corresponds well with a study by Coppalle et. al. [16].

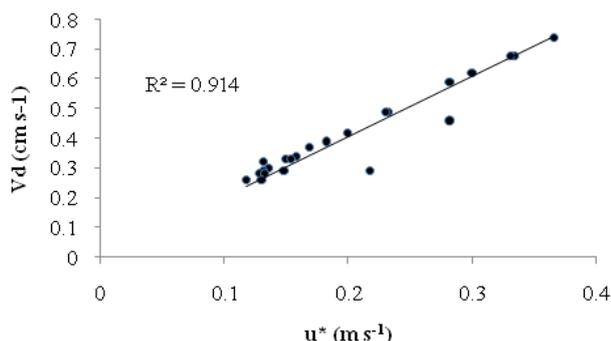


Figure 4. Correlation between deposition velocity and friction velocity

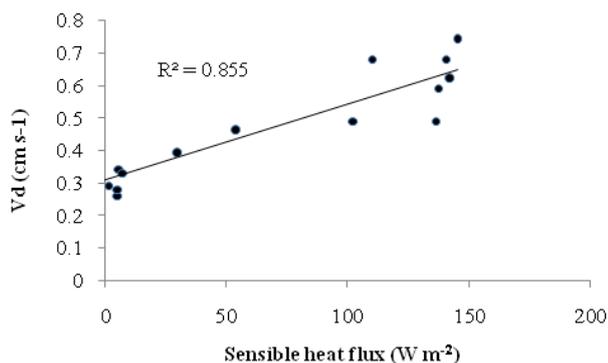


Figure 5. Correlation between deposition velocity and sensible heat flux

4. Conclusions

In this study, sulfate and nitrate dry deposition over a tropical forest was carried out for 12 months using the relaxed eddy accumulation method. Concentrations of nitrate were found to be greater than sulfate concentrations throughout the year. The average dry deposition flux of NO_3^- was greater than that of SO_4^{2-} . This indicates that NO_3^- is an important acid compound affecting the ecosystem. The average deposition velocities of sulfate were evaluated to be 0.30 cm s^{-1} during the day and 0.22 cm s^{-1} at night. The highest deposition velocities of nitrate were determined to be 0.93 cm s^{-1} during the day and 0.54 cm s^{-1} at night. The deposition velocities of aerosol calculated by the resistance model were estimated to be 0.49 cm s^{-1} during the day and 0.29 cm s^{-1} at night. Predicted values were close to measured values for sulfate but less than measured values for nitrate. The deposition velocity increases with the friction velocity and the sensible heat flux.

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