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Trends in Sulfate Dry Deposition over Mixed Dipterocarp Forest in Thailand using Relaxed Eddy Accumulation Method

(Corak Pemendapan Kering Sulfat di atas Hutan Dipterokarpa Campuran di Thailand menggunakan Kaedah Akumulasi Eddy Santai)

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ABSTRACT

The relaxed eddy accumulation method (REA) was applied for direct measurement of sulfate fluxes over mixed dipterocarp forest. The sampling system using a four-stage filter pack was designed to take updraft or downdraft air samples in the direction of the vertical wind velocity. The deadband of the velocity profile was set at ±0.5 σ_w in which an air sample with velocity within the set value is rejected. The forest site was located at a latitude of 13° 35' 13.3"N and longitude of 99° 30' 3.9"E Ratchaburi province in Thailand. The sampling period was set in 4 time intervals: 6-12, 12-18, 18-24 and 24-6 h for 3 consecutive days/month (n=144) throughout one year. All the micrometeorological parameters for flux estimation were measured in real time. The concentration and flux of sulfate were found to reach a peak value of 1.32 $\mu g\ m^{-3}$ and 8.35 $\mu g\ m^{-2}\ h^{-1}$, respectively, at noon time which indicated the effect of atmospheric instability caused by a high heat transfer during the day. The one-year average deposition velocity was observed to be 0.24 cm s^{-1}.The coefficient β obtained under high ambient temperature and a humid environment in this tropical climate was 0.49. It has also been observed that $\beta$is relatively insensitive to atmospheric stability.

Keywords: Dry deposition; forest; sulfate

ABSTRAK

Kaedah akumulasi eddy santai (REA) telah digunakan untuk pengukuran langsung fluks sulfat di atas hutan dipterokarpa campuran. Sistem pensampelan dengan menggunakan satu pek penapis empat peringkat telah direka untuk mengambil sampel udara draf atas atau bawah yang bergerak dalam arah halaju angin menegak. Paras akhir bagi profil halaju ditetapkan pada ±0.5 σ_w dan sampel udara dengan halaju di bawah nilai yang ditetapkan adalah ditolak. Tapak hutan tersebut terletak di kedudukan latitud 13° 35' 13.3 ''N dan longitud 99° 30' 3.9" E di wilayah Ratchaburi, Thailand. Tempoh persampelan telah dijalankan dalam 4 selang masa: 6-12, 12-18 , 18-24 dan 24-6 jam dalam 3 hari berturut-turut /bulan (n=144) dalam tempoh masa satu tahun. Semua parameter meteorologi mikro untuk anggaran fluks diukur dalam masa sebenar. Sulfat didapati mencapai paras puncak pada kepekatan dan fluks 1.32 $\mu g\ m^{-3}$ dan 8.35 $\mu g\ m^{-2}$ jam^{-1} masing-masing pada waktu tengahari yang menunjukkan kesan ketidakstabilan atmosfera disebabkan perpindahan haba yang tinggi pada waktu siang. Purata satu tahun kelajuan pemendapan didapati adalah 0.24 cm s^{-1}. Koefisien β yang diperoleh dalam keadaan suhu ambien yang tinggi dan humiditi dalam iklim tropika adalah 0.49. Hasil kajian juga mendapati $\beta$is secara relatif tidak sensitif terhadap kestabilan atmosfera.

Keywords: Hutan; pemendapan kering; sulfat

INTRODUCTION

The majority of SO_2 and NO_x emissions contributing to acid deposition are a product of human activities (Driscoll et al. 2003; Mortimer 2009). Acid deposition primarily results from the transformation of air pollutants such as sulfur dioxide and nitrogen oxides into secondary pollutants such as sulfuric acid, ammonium nitrate and nitric acid. Acidic particles and vapors can be deposited on the earth's surface as acid precipitation (wet deposition) or via particles such as fly ash, sulfates, nitrates and gases (dry deposition) (Mortimer 2009). In particular, the acidic sulfates are major components of atmospheric anthropogenic activities. Sulfates may be either primary or secondary. They are strongly acidic substances and condense to form particulate sulfate. Secondary sulfates are produced in the atmosphere through gas to particle conversion of SO_2 (Hazi et al. 2003). Acidification of ecosystems occurs when the deposition of acidic compounds exceeds the neutralizing capacity of the receiving environment. In forest soils, excess acid deposition increases the susceptibility of forests to stresses from pests, pathogens and climate change, resulting in poorer forest health, lower timber yields, leaf damage and eventually changes in the composition of forest species (Driscoll et al. 2003; Lindroos et al. 2006).

Weather is an important factor in the formation and transportation of acidic compounds in the atmosphere (Driscoll et al. 2003). SO_2, NO_x and the resulting acidic compounds can remain in the air for long periods of time where they are subject to the prevailing weather patterns of the region. Accordingly, fluxes of aerosol 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 the empirical relationship between fluxes and gradients of quantities measured in the inertial sub – layer (Monteith & Mike 2008). The attractiveness of using the relaxed eddy accumulation (REA) method is that it can be applied to chemical species for which fast response instrumentation is not yet available and allows flux measurements for which analyzer response time is not rapid enough to apply the eddy correlation method (EC) (Businger & Oncley 1990; Fotiadi et al. 2005; Meyer et al. 2006; Zemmelink et al. 2002). Desjardins (1972) originally developed eddy accumulation as modification from the eddy correlation technique. Businger and Oncley (1990) combined eddy accumulation with flux-variance and considered using the switching of valves to collect concentration in two separate reservoirs to determine updraft and downdraft vertical wind velocity.

In the REA method, turbulent flux is simply expressed as the product of the standard deviation of vertical wind velocity, the difference between mean scalar concentration in the updrafts and downdrafts and an empirical coefficient, β (about 0.63 as based on simulations with a Gaussian distribution and 0.58 as derived from experimental data) (Fotiadi et al. 2005). Hicks and Macmillen (1984) simulated eddy accumulation for measuring pollutant flux and recommended the determination of coefficient β should be $0.05\sigma_w$. Hamatoni et al. (1996) developed the system of using the relaxed eddy accumulation method to evaluate carbon dioxide flux over plant canopies. They determined β by using a temperature fluctuation signal to $\beta = 0.57$. Most studies use a constant literature value for β between 0.56 and 0.60 or derive β from eddy covariance (EC) measurements. Meyer et al. (2006) used the relaxed eddy accumulation method to estimate deposition velocity of sulfate over maize. The average sulfate deposition velocities were 2 and 3 cm s^{-1} for the low and fully developed maize canopies, respectively.

Myles et al. (2007) measured particulate sulfate using relaxed eddy accumulation. The deposition velocity estimated from the measured concentration and flux data was 0.42 cm s^{-1}. The relaxed eddy accumulation method has been implemented primarily in agricultural areas (Held et al. 2008; Meyer et al. 2006; Zemmelink et al. 2002). Grönholm et al. (2007) estimated aerosol particle deposition by using a REA technique with dynamic deadband over Scots pine forest. The results found that the 80–100 nanometer size particles had the lowest deposition velocity, about 0.4 cm s^{-1}. Deposition velocity increased with decreasing or increasing particle diameter from 80–100 nanometer size and also found that deposition velocity increases as a function of friction velocity. Few studies have been conducted in the forest, especially in tropical climate regions. The aims of this study were to develop the REA method using the filter pack system and to estimate the flux of sulfate and the deposition velocity over tropical forest.

Materials and Methods

Thailand has a warm, tropical climate affected by an annual monsoon, with a rainy season from June to October and a dry season for the rest of the year. Temperatures average 75 to 92°F or 24 to 33°C, with the highest and the lowest temperatures lies between April and December/ January, respectively. The tropical, rainy, warm and cloudy southwest monsoon starts in mid-May until September. The dry, cool northeast monsoon starts from November and continues to mid-March. The experimental field and tower were located at the latitude of 13° 35' 13.3'' N and the longitude of 99° 30' 3″. The area is covered by mixed dipterocarp forest. The dominant canopy tree species are *Dipterocarpus obtusifolius*, *D. tuberculatus*, *Shorea obtusa* and *S. siamensis*. They are regenerated forest and the tree height was approximately 5–7 m. The meteorological variables were obtained from a 10 m height micrometeorological tower placed in the site. A four – stage filter packs to collect sulfate aerosols, a 3D ultrasonic anemometer to measure wind magnitude and direction and a temperature and relative humidity sensor (Wisco – HT120) were installed on a tower at 10 m above the ground. The air was drawn in by a vacuum pump at a constant flow rate of 20 L/min. The sampling period was set at every 6 h (0600, 1200, 1800 and 2400 h), four times per day and 3 days/month from June 2009 to May 2010. Hence the total number of samples was 144 (n=144).

The 4-stage filter pack, starting from the inlet consisted of a Teflon filter (Pall) to collect SO_4^{2-} and NO_3^-, the second filter was nylon membrane to collect gas the third filter was potassium carbonate impregnated cellulose (Whatman, No. 41) to collect gas and the fourth filter was phosphoric acid impregnated paper to collect NH_3. This research was discussed SO_4^{2-} and NO_3^- only. The collected filters were then placed in a polyester bag and preserved in a refrigerator. To analyze the chemical species, each filter was separately put in 50 mL polypropylene tubes and filled with distilled water up to 20 mL. For the second stage filter, 20 mL of 0.1%V/V H_2O_2 was added. The tubes were shaken for 20 min in an ultrasonic shaker. The solution was then filtered using membrane 0.45 micrometer filter and preserved in a refrigerator at 4ºC. The ion chromatography (Metrohm:761 Compact IC) was used to analyze the anion species of SO_4^{2-} and NO_3^-. The data quality objectives (DQOs) of EANET was specified as ±15% for every constituent by the QA/QC program in EANET (Acid Deposition Monitoring Network in East Asia 2000).

The REA Method

The relaxed eddy accumulation method combines the eddy accumulation (EA) technique with the flux-variance similarity to obtain the vertical flux of chemical species (Businger & Oncley 1990),

$$F = \beta\sigma_w\left(\bar{C}_{up} - \bar{C}_{dow}\right), \quad (1)$$

where β is determined by the probability distribution of vertical wind speed and is based on sensible heat fluxes.

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

Figure 1 shows the relaxed eddy accumulation method uses isolated reservoirs to sample vertical wind eddies based on the direction 'updraft'(w^+) or 'downdraft'(w^-), which were determined by the 3D ultrasonic anemometer and on the vertical wind exceeding a threshold (deadband) velocity (Pryor et al. 2008). σ_w is standard deviation of the vertical wind velocity and $\bar{C}_{up}$ and $\bar{C}_{down}$ are the average concentration of pollutant in the updraft and downdraft direction, respectively. The term $w'T'$ represents multiplication of the fluctuation of the vertical component of the wind velocity and the ambient temperature, respectively. In this study we applied a deadband of ±0.5 σ_w where σ_w is computed as a running mean over a period. The deadband is the pre-set value of the vertical wind velocity whereby the air inlet below the set value will be rejected. The deadband is controlled by a microcontroller and C++ program which signals the solenoid valves to open or close the inlet line through the reservoirs line and the deadband line. The switching frequency between the updraft, downdraft and deadband was 1 s. This deadband will cut off the lower threshold of the vertical wind around zero which significantly reduces the frequency of valve switching and increase the ambient concentration difference.

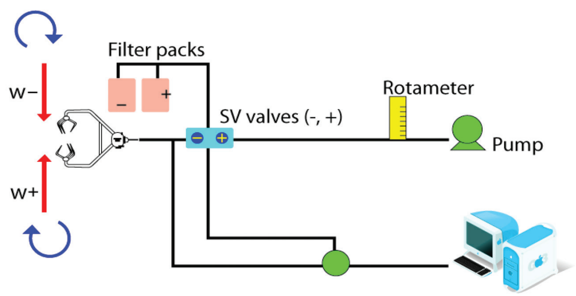

FIGURE 1. A schematic diagram of the REA system

The direct measurements of vertical flux near the surface can be used to characterize dry deposition velocity (V_d). F is Flux of pollutants (ug m^{-2} s^{-1}) and C is concentration of pollutant (ug m^{-3}). This velocity is commonly used in parameterization of dry deposition rate and can be expressed as:

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

Results and Discussion

CONCENTRATIONS AND MICROMETEOROLOGICAL PARAMETERS, β COEFFICIENT, FLUXES AND DEPOSITION VELOCITIES

The diurnal variation of sulfate concentration is shown in Figure 2(a). The average concentrations were 1.27, 1.32, 1.26 and 1.28 µg m^{-3} in the early morning (6 h), noon (12 h), evening (18 h) and night (24 h), respectively. The average concentration measured in every 6 h was observed to increase in the daytime and again, at nighttime. Figure 2(a) and 2(b) show the same direction of sulfate concentration increases with ambient temperature but inverse relationship with the relative humidity. Temperature and relative humidity are considered to be important meteorological parameters influencing the formation of atmospheric aerosols. The secondary sulfates were produced in the atmosphere through the gas to particle conversion of SO_2 under high temperature. Relative humidity seems to play an insignificant role in this observation, even though theoretically, acid particle can absorb water. This phase transition is known as deliquescence relative humidity (Seinfeld & Pandis 2006).

Average temperature of 28.5, 32.2, 24.4 and 23.2°C were observed in the morning (6 h), noon (12 h), evening (18 h) and night (24 h), respectively (Figure 2(b)). The ambient temperature reached the maximum value at noontime. Average relative humidities of 71.6, 59.5, 79.4 and 85.3% were observed in the morning (6 h), afternoon (12 h), evening (18 h) and night (24 h), respectively (Figure 2(b)). The minimum relative humidity occurred around noontime.

The local diurnal wind speeds detected by 3D ultrasonic anemometer at 10m above the canopy were observed to be 1.17, 1.84, 1.10, and 0.92 m s^{-1} in the morning (6 h), afternoon (12 h), evening (18 h) and night (24 h), respectively (Figure 2(c)). The diurnal cycle of heating and cooling of the canopy surface causes varying thermal stability, which strongly effects turbulent mixing of the air and momentum exchange process and hence influencing stability of the atmosphere. Diurnal wind speed is clearly increased to a maximum value at noontime indicating a typical daytime convective heat condition. It tended to stabilize at nighttime when the ambient environment was cooling down.

DETERMINATION OF β COEFFICIENTS

The diurnal variability of β coefficient depends on the sensible heat flux and the atmospheric stability. It was verified by the relationship of the conditionally sampled temperature difference multiplied by standard deviation of vertical wind speed with eddy correlation heat flux as expressed in (2) and graphically presented in Figure 3. The β coefficient is the slope of the linear line in which it was 0.49 and R^2 = 0.92. In this study, we applied a deadband of ±0.5 σ_w based on a vertical wind speed at each 0.1 s measurement.

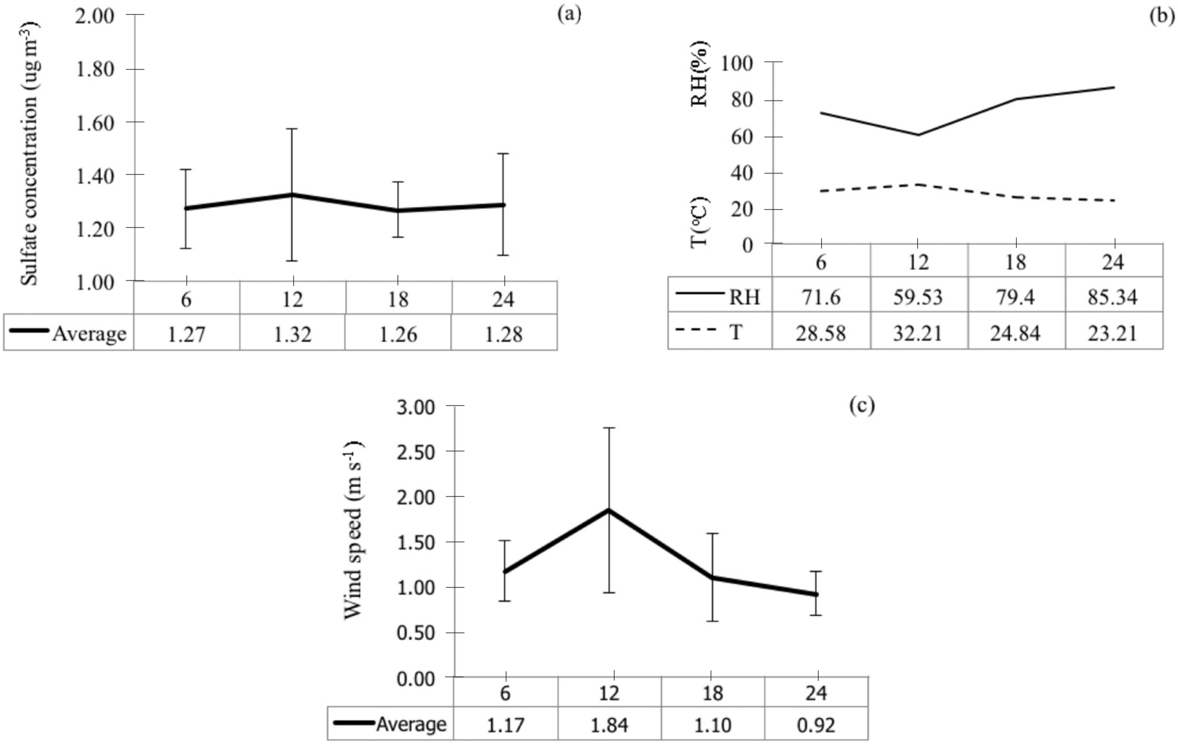

FIGURE 2. The diurnal variation of profile parameters of (a) sulfate concentrations (b) temperature and relative humidity and (c) wind speed

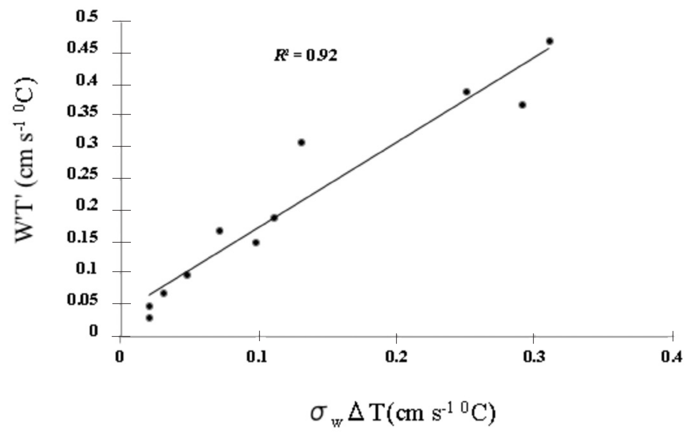

FIGURE 3. The relationship between conditionally sampled temperature difference and eddy correlation flux above the forest canopy

The diurnal variation of the β coefficient by this study was likely to increase with higher temperature and lower relative humidity (Figure 4). The values were 0.49, 0.54, 0.47 and 0.44 in the morning (6 h), afternoon (12 h), evening (18 h) and night (24 h), respectively. Recent studies suggest the β value ranged from 0.51 – 0.62 (Held et al. 2008). Katul et al. (1996) found that the experimental values of β are smaller than the theoretical ones (0.627) which a mean value of $\beta = 0.56 – 0.58$. These values are agreement with a wide range of value.

GrÖnholm et al. (2008) simulated β_{CO2} with different deadband widths. His study showed that the median value

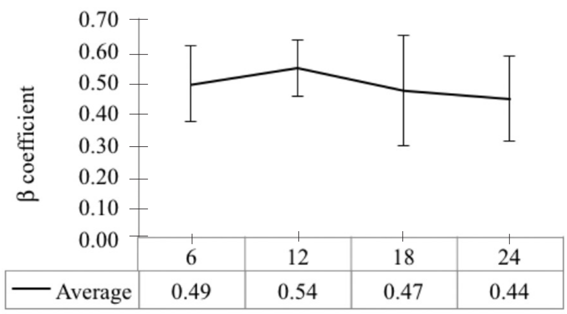

FIGURE 4. The diurnal variation of β coefficient

of the β coefficient decreases and the distribution narrows with increasing deadband width (Table 1). The β coefficient is significantly important in estimating the deposition flux value because it relates the turbulent flux to accumulated concentration difference between updrafts and downdrafts. The value varied between 0.39 and 0.63 (Table 1) and can affect the flux estimation by 30-40% as referred to (1).

TABLE 1. Deadband widths and β coefficients in REA reported in the literature and in this study

Study	Deadband	β
Zemmelink et al. (2002)	$0.3\sigma_w$	0.55
Gaman et al. (2004)	$0.5\sigma_w$	0.39
Haapanala et al. (2005)	$0.5\sigma_w$	0.41
Lee et al. (2005)	$0.4\sigma_w$	0.45
Olofsson et al. (2005)	$0.5\sigma_w$	0.42
Graus et al. (2006)	$0.6\sigma_w$	0.39
Meyer et al. (2006)	$0.5\sigma_w$	0.63
Skov et al. (2006)	$0.3\sigma_w$	0.52
Myles et al. (2007)	$0.3\sigma_w$	0.54
GrÖnholm et al. (2008)	$0.2\sigma_w$	0.50
	$0.4\sigma_w$	0.45
	$0.5\sigma_w$	0.43
	$0.6\sigma_w$	0.41
	$0.8\sigma_w$	0.38
Ren et al. (2011)	$0.5\sigma_w$	0.42
This study	$0.5\sigma_w$	0.49

DETERMINATION OF SULFATE FLUXES AND DEPOSITION VELOCITIES

The diurnal fluxes of sulfate estimated by (1) were observed to be 13.85, 13.95, 9.85 and 8.35 μg $m^{-2}h^{-1}$ in the morning (6 h), afternoon (12 h), evening (18 h) and night (24 h), respectively (Figure 5(a)). The fluxes of sulfate increased in the daytime due to high instability during the day caused by the higher heat flux in terms of the β coefficient as shown in Figure 3.

The deposition velocities of sulfate were calculated by (3) to be 0.30, 0.29, 0.22 and 0.18 cm s^{-1} in the morning (6 h), afternoon (12 h), evening (18 h) and night (24 h), respectively (Figure 5(b)). The average value of Vd with a total of 144 samplings was determined to be 0.24 cm s^{-1}. Matsuda et al. (2010) estimated a deposition velocity of PM2.5 sulfate above a deciduous forest in the summer in which the deposition velocities increased during the daytime (0.9±1.0 cm s^{-1}) and decreased during nighttime (0.3±0.3 cm s^{-1}). Accordingly, the height of tree and aerodynamic resistance can affect the deposition velocity. The Vd obtained over the dipterocarp forest in tropical climate was observed to be lower than the Vd found in the deciduous forest in central Japan. The reported values of sulfate deposition at various location and method including this study are summarized in Table 2. These listed values were not directly comparable to each other because of their differences in experimental procedures, estimation techniques, field studies and physical properties of the aerosol.

CONCLUSION

The relaxed eddy accumulation method is the most practical technique to be used for estimating the flux of sulfates in forest for a temporal analysis. The concentration and flux of sulfate were found to reach a peak value of 1.32 μg m^{-3} and 8.35 μg m^{-2} h^{-1}, respectively, at noontime which indicated the effect of atmospheric instability caused by a high heat transfer during the day. The one-year average deposition velocity was observed to be 0.24 cm s^{-1}. In tropical climatology where the ambient temperature is not varied throughout the year, the average value of β coefficient was evaluated to be 0.49.

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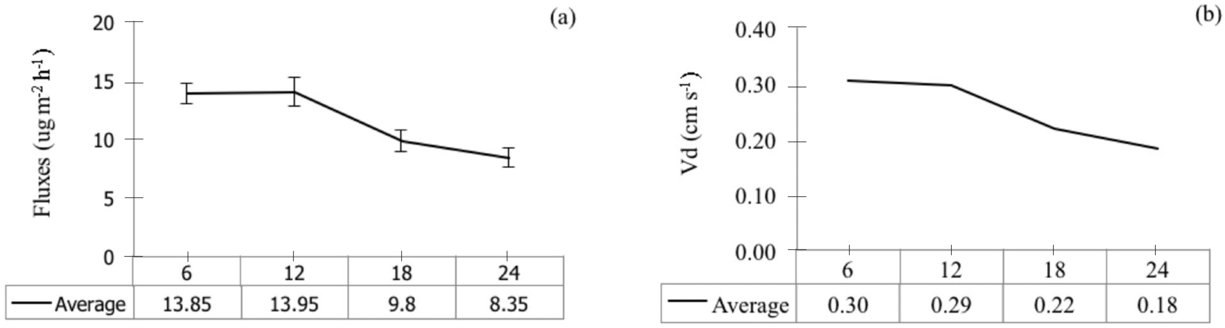

FIGURE 5. The diurnal variation of flux and deposition velocity

TABLE 2. Review of measured dry deposition velocities for sulfate

V_d (cms^{-1})	Method	Surface	References
6.3	Surrogate	Roof of four story building	Odabasi & Bagiroz (2002)
3.0 campus 7.5 city	Surrogate	Roof of two story building	Tasdemir & Gunes (2006)
<0.1	Gradient	Grass	Atkins & Garland (1974)
1.0-2.0	Gradient	Grass (rough)	Everett et al. (1979)
0.3-0.4	Gradient	Grass	Doran & Droppo (1983)
0.7 stable 2.7 unstable	Gradient	Speulderbos forest	Duyzer et al. (1994)
0.7	Gradient	Coniferous forest	Wyers & Duyzer (1997)
1.01	Gradient	Agriculture	Nemitz et al. (2000)
0.9 day time 0.3 night time	Gradient	Deciduous forest	Matsuda et al. (2010)
0.38	Gradient	Deciduous forest	Khoomsab & Pojanie (2010)
0.7	EC[1]	Coniferous forest	Hick et al. (1982)
0.6	EC	Deciduous forest	Hick et al. (1989)
0.22	EC	Grass	Wesely et al. (1985)
0.1-0.6	EC	Semi - arid	Lamaud et al. (1994)
2.0-3.0	REA[2]	Agriculture	Meyer et al. (2006)
0.42	REA	Tampa bay	Myles et al. (2007)
0.24	REA	Deciduous forest	This study

1 Eddy covariance 2 Relax eddy accumulation

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