

## Dry Deposition of SO<sub>2</sub> Over Dry Dipterocarp Forest, Thailand (Pemendapan Kering SO<sub>2</sub> ke atas Hutan Kering Dipterokarpa, Thailand)

PHUVASA CHANONMUANG\*, POJANIE KHUMMONGKOL & KAZUHIDE MATSUDA

### ABSTRACT

The aerodynamic gradient method was applied to estimate dry deposition flux of SO<sub>2</sub> over dry deciduous forest in Nakorn Ratchasima province, Thailand. The meteorological parameters and concentrations of SO<sub>2</sub> were measured in real time for one year on the experimental tower at 36 and 27 m high. The flux observed in the wet season were found two times higher than the value observed in the dry seasons, i.e. 20±7.58 and 10±11.05 ng m<sup>-2</sup> s<sup>-1</sup>, respectively. The leaf area index and the ambient humidity were believed to assert the rate of SO<sub>2</sub> deposition. The average friction velocities were estimated to be 0.26±0.02 and 0.48±0.06 m s<sup>-1</sup>, for the dry and the wet season, respectively. The friction velocity was very much depended on the surface roughness of the forest canopy. The V<sub>d</sub> of SO<sub>2</sub> evaluated by the aerodynamic gradient was compared with V<sub>d</sub> calculated by the resistance model. It was found that the observed V<sub>d</sub> was in close proximity with the model prediction in daytime in all conditions, i.e. wet, dry and annual average. The annual average V<sub>d</sub> determined by the Aerodynamic gradient was 0.43±0.06 cm s<sup>-1</sup>.

*Keywords:* Aerodynamic gradient; deposition velocity; resistance model; sulfur dioxide

### ABSTRAK

Kaedah kecerunan aerodinamik digunakan untuk menganggarkan pemendapan kering SO<sub>2</sub> ke atas hutan kering di wilayah Nakorn Ratchasima, Thailand. Parameter meteorologi dan kepekatan SO<sub>2</sub> diukur pada masa sebenar untuk satu tahun di menara eksperimen pada ketinggian 36 dan 27 m. Fluks yang diperhatikan pada musim hujan mempunyai nilai dua kali ganda lebih tinggi daripada nilai yang diperhatikan pada dalam musim kering, iaitu masing-masing pada 20±7.58 dan 10±11.05 ng m<sup>-2</sup> s<sup>-1</sup>. Indeks kawasan daun dan kelembapan sekeliling digunakan untuk mendapatkan kadar pemendapan SO<sub>2</sub>. Geseran purata kelajuan dianggarkan masing-masing pada 0.26±0.02 dan 0.48±0.06 m s<sup>-1</sup> untuk musim kering dan hujan. Halaju geseran sangat bergantung kepada kekasaran permukaan kanopi hutan. V<sub>d</sub> SO<sub>2</sub> dinilai melalui kecerunan aerodinamik dibandingkan dengan V<sub>d</sub> yang dikira melalui model rintangan. Didapati bahawa penilaian V<sub>d</sub> hampir sama dengan ramalan model di siang hari dalam semua keadaan, iaitu basah, kering dan purata tahunan. Purata tahunan V<sub>d</sub> yang ditentukan melalui kecerunan aerodinamik adalah pada 0.43±0.06 cm s<sup>-1</sup>.

*Kata kunci:* Halaju pemendapan; kecerunan aerodinamik; model rintangan; sulfur dioksida

### INTRODUCTION

Due to its effects on humans and on ecological systems, SO<sub>2</sub> is the most extensively measured gas in many countries and regions. SO<sub>2</sub> is one of the three major players in the atmospheric sulfur cycle composed of dimethyl sulfide (DMS), SO<sub>2</sub> and SO<sub>4</sub><sup>2-</sup>. When SO<sub>2</sub> is released, it undergoes gas-to-particle conversion processes by reacting chemically with atmospheric radicals, dust particles and water vapor in the presence of sunlight (Cooper et al. 1993; Thornton et al. 1996). The acidic sulfate aerosol (SO<sub>4</sub><sup>2-</sup>) is the main product of the process. SO<sub>2</sub> is considered to be both a source and sink in transportation, transformation (chemical reaction) and removal processes from the atmosphere or surface exchange (Delmas & Servant 1983; Erisaman & Draaijers 1995; Rodhe 1978).

In recent years, many measurements of SO<sub>2</sub> dry deposition fluxes and velocity have been made by various means and in several areas such as in urban (polluted areas), rural and remote areas. SO<sub>2</sub> can be distinguished

as being either exported from or imported into a local area or surrounding region depending on their SO<sub>2</sub> balance (Fellenberg 2000). Fowler et al. (2009) developed a monitoring system and continuous determination of dry deposition fluxes and velocities of acidifying gases, as well as gas absorbed as tiny aerosol particles, over a forest canopy. The dry deposition velocity of SO<sub>2</sub> over an evergreen broadleaf forest in Central Taiwan showed that the deposition rate was larger over a wet canopy and in the daytime. It indicated that the dry deposition rate increased with surface wetness (non-stomatal resistance) during the wet season.

In the daytime, the dry deposition rate depended strongly on the leaf area index (LAI), the stomata uptake and the amount of solar radiation (Tsai et al. 2010). The vegetation characteristics of the canopy also greatly impacted dry deposition velocities of gases, such as SO<sub>2</sub> and O<sub>3</sub>, aside from the atmospheric turbulent state and molecular exchange (Padro 1993). The dry deposition

velocities of SO<sub>2</sub> were estimated in several different surfaces. It was found that the dry deposition rate of SO<sub>2</sub> was strongly dependent upon the seasonal and diurnal variations. In the daytime, during the summer, the rate was observed to be 0.4, 0.2, 0.5 and 0.8 cm s<sup>-1</sup> for the forest, cultivated land, grassland and ocean, respectively (Xu & Carmichael 1998a).

The deposition velocity of SO<sub>2</sub> was measured over red pine forest in Japan, and its value was observed to be 0.9 cm s<sup>-1</sup> (Matsuda et al. 2002). Moreover, the deposition velocity of SO<sub>2</sub> over a tropical teak forest in Northern Thailand was measured using the aerodynamic gradient method for estimation. It was found that higher deposition velocities occurred during the wet season due to non-stomatal uptake. The deposition velocity was higher in the daytime than in the nighttime for both dry and wet seasons and increased with relative humidity and surface wetness (Matsuda et al. 2006). The SO<sub>2</sub> dry deposition was found to increase with high relative humidity, dew and rain (Zhang et al. 2003). This finding is in agreement with other studies reported in the literature. In Central Thailand, the dry deposition of SO<sub>2</sub> was measured over a rice paddy field using the Bowen ratio technique. The measurements showed that the deposition velocity varied with solar radiation, with the time of day and with climates (Jitto et al. 2007). The dry deposition of SO<sub>2</sub> was also measured over grass fields (bahiagrass and bermudagrass) along with other key atmospheric species using the relaxed eddy accumulation method. Acid deposition affected our ecosystem by reducing the growth rate, amount and type of forests. Thus, more recent studies of acid deposition are concentrated in remote areas such as the highlands, mountains and areas over natural forests.

This investigation focused on SO<sub>2</sub> deposition over the dry dipterocarp forest (DDF) in tropical climates in northeast Thailand. The micro-meteorological parameters were measured continuously, along with the ambient SO<sub>2</sub> concentration. The deposition flux of SO<sub>2</sub> was evaluated using the gradient method. Once the flux of SO<sub>2</sub> was obtained, the deposition velocity could be determined. The deposition velocity of SO<sub>2</sub> evaluated for this study was then compared with the resistance model in order to see the model applicability in this tropical climate.

## EXPERIMENTAL DETAILS

### EXPERIMENTAL SITE DESCRIPTION

The experimental tower used in this study was located in dry dipterocarp forest (DDF) at Sakaerat Environmental Research Station (SERS; 14°30'13.68"N, 101°57'8.67"E) at 300 m above sea level in Nakorn Ratchasima province, northeast Thailand. That area is surrounded by a mountain range with heights from 200 to 272 m.

### EXPERIMENTAL SET UP

The SO<sub>2</sub> concentrations were measured on the experimental tower at two levels, 36 and 27 m above the ground, in

order to measure the sample concentration differences. The ambient airflow to the system was induced constant flow rate of 10 L min<sup>-1</sup>. The concentration of SO<sub>2</sub> from the two air samplers was determined by ultraviolet fluorescence (UVF-100E). The wind velocity was monitored by a 3D ultrasonic anemometer (YOUNG, 81000). The field measurements were conducted for one year from August 2011 to August 2012. The period from November to April was classified as the dry season for this tropical region. The period from May to October was classified as the wet season.

### AERODYNAMIC GRADIENT

The gradient method was applied to estimate the dry deposition fluxes of SO<sub>2</sub>. The mass flux of SO<sub>2</sub> gas can be calculated using (1):

$$F = -u_*c^*, \quad (1)$$

where  $u_*$  is the friction velocity and  $c^*$  is the eddy concentration. The latter is expressed in (2) (Feliciano et al. 2001; Matsuda et al. 2012),

$$c^* = k\Delta c / [\ln(z_2-d/z_1-d) - \Psi_h(z_2-d/L) + \Psi_h(z_1-d/L)], \quad (2)$$

where  $\Delta c$  is the difference between SO<sub>2</sub> concentrations at heights between  $z_1$  and  $z_2$ ;  $d$  is the displacement height ( $d = 10, 12$  and  $14$  m) (different values of  $d$  were selected, depending on the season) (Matsuda et al. 2012);  $k$  is the Von Kaman constant (0.4);  $L$  is the Monin-Obukhov length; and  $\Psi_h$  is the integrated stability correction function for heat. In this experiment, the sampling points were located on the tower at heights of 27 and 36 m for the lower height ( $z_1$ ) and upper height ( $z_2$ ), respectively. From (1) and (2), the mass flux  $F$ , can be defined in (3):

$$F = -ku_* \Delta c / [\ln(z_2-d)/(z_1-d) - \Psi_h(z_2-d/L) + \Psi_h(z_1-d/L)], \quad (3)$$

values for  $u_*$  and  $L$  were averaged every 10 min. The transfer velocity,  $D$  (m s<sup>-1</sup>), which was a part of the flux calculation was computed using (4):

$$D = ku_* / [\ln(z_2-d/z_1-d) - \Psi_h(z_2-d/L) + \Psi_h(z_1-d/L)]. \quad (4)$$

In this study, the value of  $D$  was averaged over 1 h period. The value was then multiplied by the difference in the concentrations between the two heights ( $\Delta c$ ). From the previous study, this method was used to estimate both aerosol and trace gas fluxes (Hayashi et al. 2011; Matsuda et al. 2010, 2005). The deposition velocity,  $V_d$ , is determined using (5):

$$V_d = -F/C, \quad (5)$$

where  $F$  is the dry mass deposition flux and  $C$  is the concentration at the reference point.

## RESISTANCE MODELING

The resistance estimation of atmospheric gases (Wesley & Hicks 2000),  $SO_2$  is expressed in (6):

$$V_d = (R_a + R_b + R_c)^{-1}, \quad (6)$$

where  $R_a$  is the aerodynamic resistance;  $R_b$  is the quasi-laminar boundary layer resistance and  $R_c$  is the surface resistance.  $R_a$  and  $R_b$  were determined as a function of meteorological conditions each day in each season, following (7-8) (Hicks et al. 1987; Myles et al. 2007):

$$R_a = \bar{u}(z)u^{*-2}, \quad (7)$$

$$R_b = 5(Sc^{0.67} u^{*-1}), \quad (8)$$

where  $\bar{u}$  is the wind speed at the top of the canopy;  $u^*$  is the friction velocity; and  $Sc$  is the Schmidt number (defined as the ratio of the kinematic viscosity of air to the molecular diffusivity coefficient).  $R_c$  was expressed by Wesley resistance model (Erisman & Baldocchi 1994; Matsuda et al. 2001; Zhang et al. 2003): The important parameter,  $u^*$  was expressed in terms of  $R_a$  and  $R_b$ .

## RESULTS AND DISCUSSION

## METEOROLOGICAL PARAMETER MEASUREMENT

Figure 1 shows the variations of meteorological parameters, including wind speed, rainfall levels, temperature, relative humidity, solar radiation and net radiation during the wet and the dry seasons. Rainfall levels during the wet season (May to October) ranged from 75 to 246 mm, whereas the measured precipitation ranged in value from 0 to 34 mm during the dry season (November to April). The wind speed showed some fluctuation during the transitional period from wet to dry weather (September–November) and then increased continuously from the dry ambient (December 2011) towards the wet ambient environment (April 2012) as shown in Figure 1(a). The ambient temperature in the tropical climate does not vary substantially (Figure 1(b)). It was in the range of 22–27°C. The average temperature throughout the year was 26°C. The relative humidity was found to be somewhat lower in the dry season (58%–65%) than in the wet season (70%–80%), Figure 1(b). Normally, the temperature in the dry season (December and January) would be around 3–4°C lower than the ambient average. The weather during the dry season was characterized by lower average temperature and humidity and no precipitation (November–April). The weather during the wet season was characterized by higher average temperature and humidity with large amount of precipitation (August–October). The solar radiation (SR) and the net radiation (Rn) were found to be in the ranges of 0.12–0.2 and 0.08–0.14 kW/m<sup>2</sup>, respectively. The intensity of solar energy was influenced by the amount of cloud cover in the daytime (Figure 1(c)).

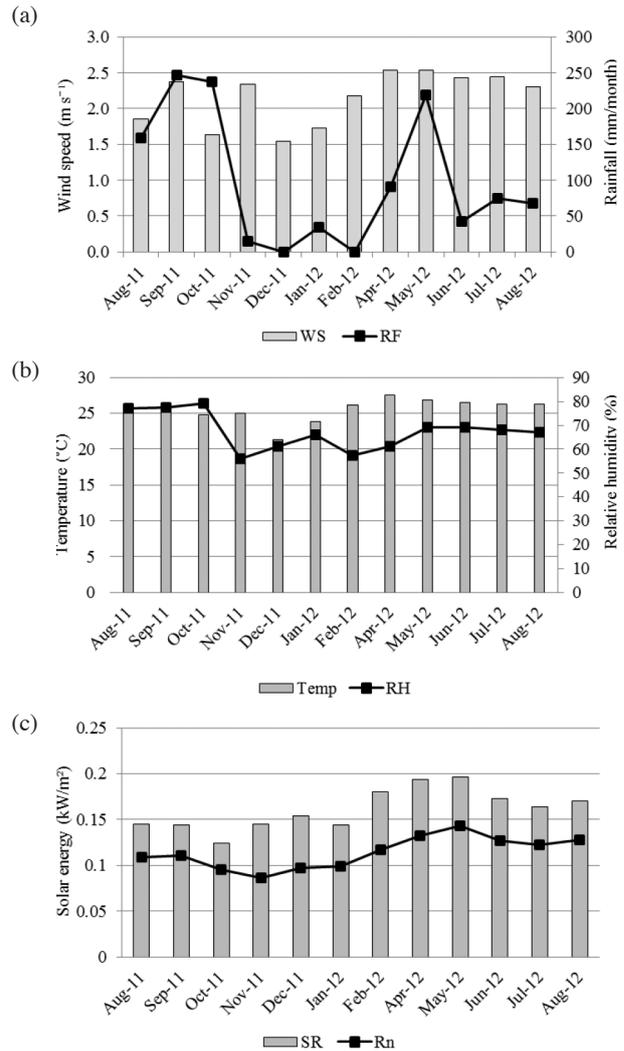


FIGURE 1. Meteorological parameters measured during August 2011–August 2012

## EVALUATION OF DRY DEPOSITION FLUX

The dry deposition flux,  $F$ , of  $SO_2$  was estimated using (1) and the median values were shown in Figure 2. The study showed a seasonal effect on deposition fluxes. The flux observed during the wet season ( $20 \pm 7.58 \text{ ng m}^{-2} \text{ s}^{-1}$ ) was two times higher than the value observed during the dry season ( $10 \pm 11.05 \text{ ng m}^{-2} \text{ s}^{-1}$ ). The important meteorological parameters that affected the  $SO_2$  deposition flux were found to be the friction velocity,  $u^*$ , and the humidity. The value of  $u^*$  that was calculated in a wet environment was  $0.48 \pm 0.06 \text{ cm s}^{-1}$ . This was also two times higher than the value that was calculated in a dry environment ( $0.26 \pm 0.02 \text{ cm s}^{-1}$ ). The monthly average value of the humidity, as shown in Figure 1(b), was also in line with the deposition flux. Other meteorological parameters, i.e. temperature and solar intensity, showed less effect on the deposition flux. The annual average  $F$  was determined to be  $16.76 \pm 5.82 \text{ ng m}^{-2} \text{ s}^{-1}$ .

The leaf area index (LAI) has been studied for its influence in increasing the rate of  $SO_2$  deposition. In this

experiment, the LAI was measured to be 5.0-6.0 during a leafy-transitional period (wet season) and 2.5-3.5 during transitional-leafless period (dry season).

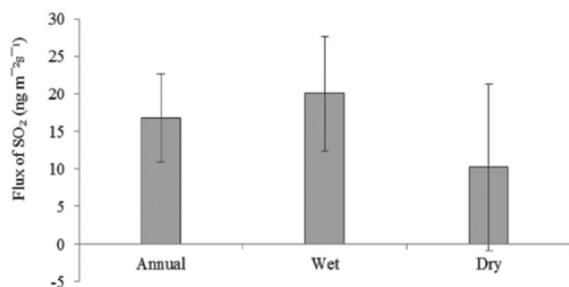


FIGURE 2. Dry deposition flux of SO<sub>2</sub> evaluated using the aerodynamic gradient method

#### EVALUATION OF DRY DEPOSITION VELOCITY

The dry deposition velocity,  $V_d$ , of SO<sub>2</sub> was estimated using (5) and shown in Figure 3. The median value of  $V_d$  in daytime was determined to be  $0.50 \pm 0.06$  and  $0.45 \pm 0.02$  cm s<sup>-1</sup> during the wet and the dry season, respectively. The annual median value of  $V_d$  in daytime was  $0.43 \pm 0.06$  cm s<sup>-1</sup>. For nighttime,  $V_d$  was estimated to be  $0.69 \pm 0.07$  cm s<sup>-1</sup> in the wet season and nearly zero in the dry season. Daytime and nighttime  $V_d$  values determined during the wet season were close due to the significant role of the high humidity present during both day and night. Dissolution of SO<sub>2</sub> in the wet environment occurred while descending downward. The nearly zero value of  $V_d$ , observed during the dry season and in the nighttime, was commonly found by previous studies (Chimjan & Khummongkol 2012; Jitto et al. 2007). This indicated the effect that, under low humidity conditions, it causes upward motion, due to buoyancy of air during the night. The higher heat capacity of soil caused the soil surface temperature to remain high during the night while the atmospheric air temperature was decreasing.

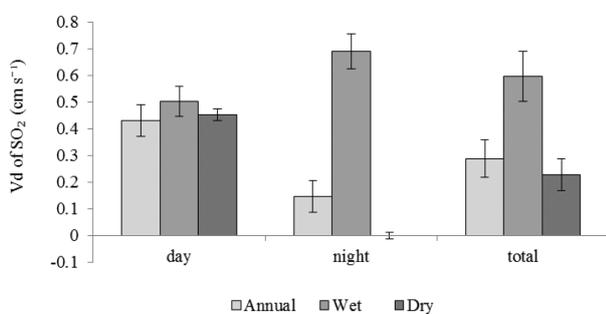


FIGURE 3. Dry deposition velocity of SO<sub>2</sub> evaluated using the aerodynamic gradient method

#### DRY DEPOSITION VELOCITY ESTIMATED BY A RESISTANCE MODEL

For this study, the average  $u^*$  values used in (7) and (8) were estimated to be  $0.26 \pm 0.02$  and  $0.48 \pm 0.06$  m s<sup>-1</sup>, for the dry and the wet seasons, respectively. The annual mean value of  $u^*$  was determined to be  $0.42 \pm 0.06$  m s<sup>-1</sup> (Figure 4). The friction velocity was very much dependent upon the surface roughness of the canopy of the forest. Naturally, the value of the leaf area index (LAI) would be high in the wet season and low in the dry season. In addition, the seasonal forest canopy at the experimental site changed from a leafy condition (April-October) to a leafless condition (November-March). The LAI values measured at the site were in the range of 5-6 during the leafy-transitional period (wet season) and 2.5-3.5 during the transitional-leafless period (dry season). The averaged  $u^*$  value observed in this study increased with LAI and vice versa.

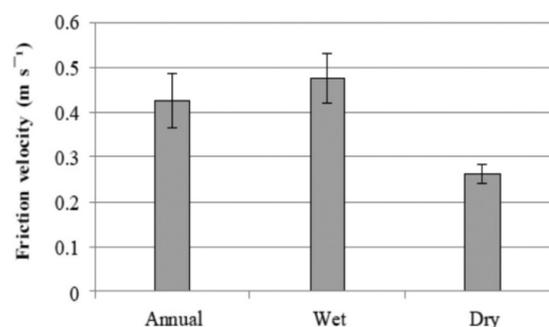


FIGURE 4. Seasonal and annual variation of friction velocity

From the field data, the amount of rainfall in the wet season greater than 1 mm day<sup>-1</sup> was regarded as wet canopy, which was recommended by the previous experiment (Matsuda et al. 2006). The  $R_c$  in this study was applied from the previous report for dry conditions, having values of 115 and 437 s m<sup>-1</sup> for the daytime and nighttime, respectively; and for wet conditions, having values of 69 and 211 s m<sup>-1</sup> for the daytime and nighttime, respectively (Zhang et al. 2003). This indicated that seasonal variation affected the  $R_c$  value.

Figure 5 shows a comparison of the observed  $V_d$  of SO<sub>2</sub>, determined by the aerodynamic gradient method with that determined using the resistance model. The observed  $V_d$  was found to be in close agreement with the model prediction in the daytime in all conditions, i.e., wet, dry and annual average. The observed and the predicted values of the annual average  $V_d$  were determined to be  $0.43 \pm 0.06$  and  $0.53 \pm 0.07$  cm s<sup>-1</sup>, respectively.

Table 1 summarizes the results of dry deposition velocity range to difference surface and several experimental investigation of SO<sub>2</sub> deposition from the previous studies.

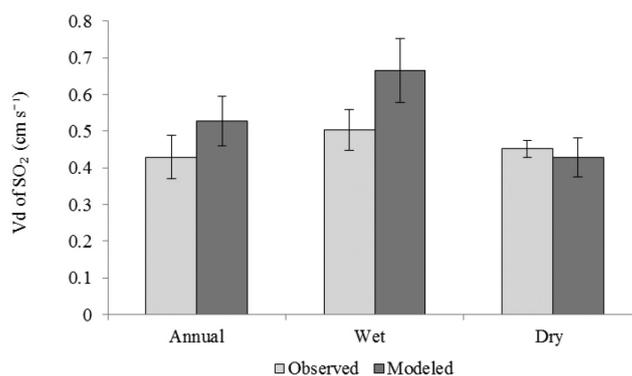


FIGURE 5. Comparison of the dry deposition velocity of SO<sub>2</sub> determined by the aerodynamic gradient method with that determined using the resistance model

TABLE 1. Deposition velocities value of SO<sub>2</sub> compared with other studies

Surface type	Location	Condition	Method	Vd (cm s <sup>-1</sup> )	References
Deciduous forest	Netherland	Winter day	Resistance method	0.3 dry 0.6 wet	Erisman et al. 1993
Coniferous forest	Sweden	Summer and winter	Enclosure technique	0.33	Granat & Richter 1995
Speulder forest	Netherland	3 years	Gradient method	1.5	Erisman et al. 1999
Low grassland	USA	May 2002	REA	0.45±0.98	Myles et al. 2007
Grassland*	Netherland	3 years	Gradient method	1.2±0.3*	Erisman et al. 1993
Healhlnd**				0.8±0.4**	
Forest (mixed)	Asia	Summer	Resistance method	0.4 day ≤0.2 night	Sorimachi et al. 2003; Xu & Carmichael 1998b
		Winter		0.1-0.2	
Deciduous forest	Japan	Jul.-Dec.	Resistance method	0.36	Wesley & Hicks 2000
Short vegetation	Germany	September	Gradient method	0.19-0.20	Horvath et al. 1998
Broadleaf evergreen forest	Taiwan	Early spring	Gradient method	0.61 day 0.27 night Dry canopy; 0.44 day 0.19 night Wet canopy; 0.83 day 0.47 night	Tsai et al. 2010
Deciduous forest	Thailand	Yearly	Gradient method	0.43±0.06 annual 0.45±0.02 dry 0.50±0.06 wet	This study
			Resistance method	0.53±0.07 annual 0.43±0.06 dry 0.67±0.09 wet	

## CONCLUSION

The flux of SO<sub>2</sub> was evaluated in the wet and dry season using the aerodynamic gradient method. The value of the deposition flux observed in the wet season was higher than the value observed in the dry season by a factor of 2 (20±7.58 and 10±11.05 ng m<sup>-2</sup> s<sup>-1</sup>, respectively). The meteorological parameters that most significantly affected SO<sub>2</sub> deposition flux were the friction velocity and the

humidity. The leaf area index (LAI) was also found to impact the rate of SO<sub>2</sub> deposition. Naturally, the value of LAI would be high in the wet season and low in the dry season. In addition, the seasonal forest canopy at the experimental site changed from a leafy condition (April-October) to a leafless condition (November-March). The value of the LAI measured at the site ranged from 5 to 6 during the leafy-transitional period (wet season) and 2.5 to

3.5 during the transitional-leafless period (dry season). The average  $u^*$  value observed in this study also increased with LAI and vice versa. A comparison was made of the observed  $V_d$  of  $\text{SO}_2$ , determined by the aerodynamic gradient method, with the  $V_d$  determined using the resistance model. The observed  $V_d$  was in close proximity with the model prediction for the daytime in all conditions, i.e., wet, dry and annual average. The observed and the predicted values for the annual average  $V_d$  were determined to be  $0.43 \pm 0.06 \text{ cm s}^{-1}$  and  $0.53 \pm 0.07 \text{ cm s}^{-1}$ , respectively.

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Phuvasa Chanonmuang\*  
 Environmental Technology Division  
 The Joint Graduate School of Energy and Environment  
 King Mongkut's University of Technology Thonburi  
 126 Pracha U-thit Rd., Bang-Mod  
 Bangkok 10140  
 Thailand

Phuvasa Chanonmuang\*  
 Thailand Institute of Scientific and Technological Research  
 35 Moo 3 Technopolis, Rungsit Nakornayok Rd.  
 Klong 5, Klong-Luang  
 Pathumthani 12120  
 Thailand

Pojanie Khumongkol  
 Environmental Technology Division  
 School of Energy Environment and Materials  
 King Mongkut's University of Technology Thonburi  
 126 Pracha U-thit Rd., Bang-Mod  
 Bangkok 10140  
 Thailand

Kazuhide Matsuda  
 Faculty of Agriculture Field Science Center  
 Tokyo University of Agriculture and Technology  
 3-5-8 Saiwai-cho, Fuchu, Tokyo  
 Japan

\*Corresponding author; email: phuvasa@tistr.or.th

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