

1 **Observation of SO₂ dry deposition velocity at a high elevation**
2 **flux tower over an evergreen broadleaf forest in Central Taiwan**

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Abstract

12

13 A 60-m flux tower was built on a 2100 m mountain for the measurement of the
14 air pollutant concentration and the evaluation of dry deposition velocity in Central
15 Taiwan. The tower was constructed in an evergreen broadleaf forest, which is the
16 dominant species of forest in the world. Multiple-level SO₂ concentrations and
17 meteorological variables at the site were measured from February to April 2008. The
18 results showed that the mean dry deposition velocities of SO₂ were 0.61 cm s⁻¹ during
19 daytime and 0.27 cm s⁻¹ during nighttime. A tendency was observed that the dry
20 deposition velocity increases with LAI and solar radiation from the comparison of the
21 monthly data. Furthermore, it was observed that the deposition velocity was larger
22 over wet canopy than over dry canopy, that higher deposition velocities in the wet
23 season were mainly caused by non-stomatal uptake of wet canopy. Over wet canopy,
24 the mean dry deposition velocities of SO₂ were estimated to be 0.83 cm s⁻¹ during
25 daytime and 0.47 cm s⁻¹ during nighttime; and 0.44 cm s⁻¹ during daytime and 0.19
26 cm s⁻¹ during nighttime over dry canopy. Comparisons of the results in this study and
27 the predictions of Zhang et al. (2003a) are in a good agreement. The median
28 (geometric mean) of derived r_c during daytime are 233 (266) m s⁻¹ over dry canopy
29 and 147 (146) m s⁻¹ over wet canopy. It was found that the solar radiation is the
30 critical important meteorological variable determining the stomatal resistance during
31 daytime. For non-stomatal resistance, clear dependencies were observed on the
32 friction velocity and relative humidity.

33 Keywords: East Asia; Sulfur dioxide; dry deposition; stomatal and non-stomatal
34 resistances; evergreen broadleaf forest

35 **1. Introduction**

36 Sulfur dioxide (SO₂) is not only one of the important specie resulting in acid
37 precipitation, also air pollution and is the precursor of sulfate. By means of dry
38 deposition of these species transferred by air motion to the surface, serious damage
39 was caused to the environment and the ecology and worldwide have become of
40 increasing concern. Consequently, evaluation of dry deposition for SO₂ is a critical
41 issue with respect to ecological impact, crop growing and air quality research.

42 Observations of SO₂ dry deposition have been carried out over different land use
43 types and vegetation areas in the last two decades, including desert, bare soil,
44 grassland, crops and forest (e.g. Brook et al., 1999). However, scant observation over
45 evergreen broadleaf forest has been reported. In the U.S. Geological Survey (USGS)
46 land cover classification (http://edcdaac.usgs.gov/glcc/globe_int.html), the coverage
47 of forest globally is about 15 % with 38% of that being evergreen broadleaf forest.
48 Hence, it is necessary to understand the behaviors of SO₂ such as its dry deposition
49 rate and related issues to increase knowledge of environmental degradation in this
50 area.

51 Recently, acidic deposition at high elevation has received much attention (Heuer
52 et al., 2000; Wai et al., 2008). It was found that at high elevations, forests are
53 vulnerable as they are often surrounded by acidic clouds and fog which are thought to
54 contribute to the decline of forests. In view of the unique environmental settings and
55 the extremely scarce data set that is available at high elevations, data from a flux
56 tower on a high mountain with the elevation exceeding 2000 m above sea level was
57 used to provide more valuable information about this issue.

58 In this paper, multilevel SO₂ concentrations and SO₂ dry deposition velocity (V_d)
59 measured over an evergreen broadleaf forest from the beginning of February to the

60 end of April 2008 are presented. Diurnal and monthly variations of dry deposition
61 velocity were estimated and related to the Leaf Area Index (LAI) and environmental
62 variables. The results were also compared with those studies of similar land use type.
63 The dataset shown in this paper is extremely informative for estimating the dry
64 deposition for the assessment of acid deposition and air pollutants.

65 **2. Site description and Methodology**

66 2.1. Flux tower

67 Data used in this study was gathered over an evergreen broadleaf forest with a
68 elevation of 2100 m in Huisun Forest flux station of National Chung-Hsing University
69 (24.076 °N, 121.126 °E), Nantou, Central Taiwan (Fig. 1). This flux tower site was
70 constructed with a grant from the Taiwan Ministry of Education and National
71 Chung-Hsing University. Its main purpose was established as one of the research
72 activities of the FLUXNET, which is the global network to assess the energy and trace
73 gas fluxes between the biosphere and atmosphere (Baldocchi et al., 2001). Therefore,
74 most meteorological variables and momentum, energy and CO₂ fluxes are measured
75 at this site. In addition, it is also designed to have capacity for air quality research,
76 including the measurement of air pollutant concentrations at different levels, and
77 evaluation of dry deposition velocities.

78 The study site is located at the top of a mountain where the general terrain in the
79 surrounding is slightly tilted. It slopes with an angle of 17° extended over roughly 3
80 kilometer towards 315°, approximately northwest (NW) which is the dominating
81 wind direction during study period in central Taiwan. The horizontal distances to the
82 nearest public road and populated area are 4 and 7 km in the west and south areas,
83 respectively, which imply the measurements at this site can be considered an
84 undisturbed results without human management (e.g. Eugster et al., 2006). The

85 dominant species in the native evergreen broadleaf forest are *Castanopsis carlesii*,
86 *Neolitsea konishii* and *Machilus thunbergii*. The height of the canopy top from the
87 ground is about 24 m and the tree density is dense, about 2115 trees ha⁻¹. In the
88 northwest direction, the extent of the homogeneous canopy for optimum footprint
89 conditions was measured to be exceeded 3 km. Besides, no abrupt changes in
90 vegetation roughness and canopy opening were occurred in the upwind areas.

91 The main structure at the site is a 60-m-high steel tower and a container housing
92 facility near the base of the tower (Fig. 2). The tower is firmly fixed with cement
93 foundations and several steel ropes anchored to the ground. Several platforms extend
94 from the tower at 10 m, 26 m, 46 m and 56 m to observe the profiles of
95 meteorological data and air pollutant concentrations. The SO₂ analyzer and the data
96 storage device are installed in the container house. SO₂ concentrations were measured
97 using a UV fluorescence SO₂ monitor (TECO model 43A) at each platform,
98 controlled by an electric valve system with 10-minute intervals between the upper and
99 lower levels. Intakes were set up at each platform and connected to the analyzer with
100 an 80 m Teflon tube. The attenuation of SO₂ concentration, such as the absorption of
101 tube and time lag between the intake and the analyzer were corrected by using
102 standard gas calibration of known SO₂ concentration.

103 Friction velocity and surface sensible heat flux were measured by a
104 three-dimensional sonic anemometer (CSAT3, Campbell Scientific Inc.) at a
105 frequency of 10 Hz and consequently, Monin–Obukhov length was determined by
106 using those observations (e.g., Businger et al., 1971). In addition, other
107 meteorological parameters such as four components of radiation, Photosynthetically
108 Active Radiation (PAR), specific humidity, temperatures, etc. were also observed at
109 the same location. Monthly Leaf Area Index (LAI) was obtained from satellite data of
110 Moderate Resolution Imaging Spectroradiometer (MODIS) and the values were 4.1 in

111 February and 6.2 in the other two months. Ancillary measurements were also
 112 conducted during study period. By analyzing the leaves of these species, the pH
 113 values were found to be from 4.4 to 5.9. The soil pH at our observation site had a pH
 114 value of about 4.7 in March.

115 2.2. The gradient method

116 Within the atmospheric surface layer, the vertical profile of scalar can be
 117 depicted by the similarity theory (e.g., Brutsaert, 1982). Based on the assumption that
 118 heat and mass are transported in a similar way, the flux of SO₂ can be determined
 119 using the gradient method (Baldocchi et al., 1988) as:

$$120 \quad F = -u^* c^* \quad (1)$$

$$121 \quad c^* = k\Delta c / [\ln((z_2 - d_0)/(z_1 - d_0)) - \Psi_h((z_2 - d_0)/L) + \Psi_h((z_1 - d_0)/L)] \quad (2)$$

122 where u^* is the friction velocity (m s⁻¹); c^* is the eddy concentration (μg m⁻³); Δc
 123 is the difference of SO₂ concentrations at a height between z_2 and z_1 (μg m⁻³); k is the
 124 Von Karman constant; L is the Monin–Obukhov length (m); Ψ_h is the integrated
 125 stability correction function for heat defined by Businger et al. (1971); d_0 is the
 126 zero-plane displacement height (m), which is typically 60-75% of the height of the
 127 vegetative canopy (Wesely and Hick, 2000). To determine d_0 in each month, a
 128 relationship, which predicts the ratio of d_0 to canopy height (h) to vary with LAI ,
 129 proposed by Perrier (1982) was used:

$$130 \quad \frac{d_0}{h} = 1 - \left(1 - \left[e^{\frac{-aLAI}{2}} \right] \right) \left(\frac{2}{aLAI} \right) \quad (3a)$$

$$131 \quad a = \begin{cases} 2f, & f \geq 0.5 \\ (2(1-f))^{-1}, & f < 0.5 \end{cases} \quad (3b)$$

132 where a is an adjustment factor for LAI distribution within the canopy and f is the
 133 proportion of LAI lying above $h/2$. Here the f is approximately estimated to be 0.75

134 for the native evergreen broadleaf forest in each month. Using Eq (3a) and (3b), the d_0
 135 are determined as $0.70h$ for February and $0.78h$ for March and April. Once the SO_2
 136 flux has been determined, the dry deposition velocity V_d , can be estimated by the
 137 following equation:

$$138 \quad V_d = F/C \quad (4)$$

139 where C is the SO_2 concentration at height z_2 ($\mu g\ m^{-3}$). In this study, F is positive
 140 when the flux is towards the surface and negative when flux occurs in the opposite
 141 direction.

142 In the resistance analogy models, the primary resistances to the transport of the
 143 pollutant from the atmosphere to the surface are the aerodynamic resistance (r_a), the
 144 quasi-laminar sublayer resistance (r_b) above the canopy, and the overall canopy
 145 resistance (r_c) (e.g. Wesely, 1989; Feliciano, et al., 2001; Zhang et al., 2003a).

146 Therefore, the observed r_c can be obtained, from the observed deposition velocity, the
 147 calculated r_a and r_b , as following:

$$148 \quad r_c = \frac{1}{V_d} - r_a - r_b \quad (5)$$

149 Expressions for r_a and r_b can be found in many dry deposition studies (e.g. Padro et
 150 al., 1991; Erisman et al., 1994; Massman et al., 1994) and the uncertainties in r_a and r_b
 151 in the different models are small (Massman et al., 1994). The following equation was
 152 adopted to calculate r_a and r_b (e.g. Seinfeld and Pandis, 1998):

$$153 \quad r_a = \frac{\ln(z/z_0) - \psi_h}{ku^*} \quad (6)$$

$$154 \quad r_b = \frac{5}{u^*} \left(\frac{\nu}{D_{SO_2}} \right) \quad (7)$$

155 where z is reference height (m), z_0 the roughness length (m), ν the viscosity of air
 156 and D_{SO_2} the diffusivity of SO_2 . The value of z_0 is estimated as 0.1 times as height of
 157 forest canopy (e.g. Baldocchi et al., 1998).

158 **3. Results and discussions**

159 3.1. Meteorological conditions and data screenings

160 In order to study the dry deposition of SO₂ over forest, it is first necessary to
161 understand the meteorological variables affecting the vegetation physiology. Field
162 measurements were conducted from 1 February to 30 April 2008 which is late winter
163 to early spring in the North Hemisphere. During this period, winds from north to
164 northwest occurred during 60% of the time approximately (Fig 3). As mentioned in
165 Section 2, within the directions we consider this site to be the one of those with the
166 possible footprint conditions for micrometeorological and dry deposition velocities of
167 pollutants studies. Since a truly flat terrain with reasonable horizontal extent does
168 probably not exist at this altitude range in mountainous Taiwan (e.g. Klemm et al.,
169 2006).

170 In the parameterization of canopy resistance for SO₂ dry deposition of Wesely
171 (1989), solar radiation and air temperature were consider as the key meteorological
172 variables. ~~The hourly values of measured meteorological variables that affected the~~
173 ~~SO₂ dry deposition during this period are presented in Fig. 2, including solar radiation,~~
174 ~~air temperature, wind speed, relative humidity and precipitation.~~ The mean monthly
175 values of these variables were divided into daytime (7:00–17:00) and nighttime
176 (18:00–6:00) groups and summarized in Table 1. During clear days, solar radiation
177 can be more than 1000 W m⁻² around noon; in contrast, it decreased to less than 200
178 W m⁻² during cloudy days and under foggy conditions. Other variations seen were the
179 mean daily solar radiation in March and April being larger than that in February.
180 There were obvious variations in temperature (T_a) during the study period. Daytime
181 T_a ranged from low levels (< 5 °C) in February to normal levels (~ 15 °C) near the
182 end of April. The total amount of precipitation during the study period was 251 mm

183 which is approximate 10 % of the mean annual precipitation from 2001 to 2008
184 according to a weather station 7.5 km to the southeast. In addition, due to the local
185 circulation at such high altitude, mist occurred occasionally in the afternoon.

186 A total of 2116 hourly data were screened to eliminate or minimize the influence
187 of periods associated with inadequate meteorological conditions, low precision of
188 measured concentration and other errors arising both from measurements and
189 mathematical approaches. Less reliable data were removed from the sampled datasets
190 using the following data-screening criteria (Feliciano et al., 2001; Matsuda et al.,
191 2005):

192 (1) Wind speed was less than 1 m s^{-1} ,

193 (2) SO_2 concentration was lower than 0.1 ppb,

194 (3) Absolute value of V_d exceeded potential max dry deposition velocity ($V_{d, \max}$) by

195 1.5 times (i.e., $1.5 V_{d, \max}$), where the potential max dry deposition velocity is

196 defined as $V_{d, \max} \equiv (r_a + r_b)^{-1}$.

197 After screening the data with the criteria, only 934 records were used in the following
198 analysis.

199 It is generally agreed that V_d over dry and wet canopies are different. With the
200 existence of aqueous layers on the surface, the wetness increases the SO_2 uptake.
201 Consequently, data in this study were separated into those over dry and wet canopies
202 to discuss the individual characteristics of SO_2 V_d . Canopy wetness may be caused by
203 rain, fog, mist and dewfall from the atmosphere in the study site and its duration is a
204 difficult variable to measure. Here, two criteria were adopted for the identification of
205 wet canopy. The first was the estimation of the rain period. Data recorded
206 precipitation and the one following just after precipitation took into account a
207 continuation of wet conditions by rain (e.g. Matsuda et al., 2006). The second was the

208 estimation of dew condensation. In this study we adopted the work of Sentelhas et al.
209 (2008) who pointed out that the constant relative humidity threshold method could be
210 a practical and useful tool for estimating dew condensation. Here a constant value of
211 $RH = 90\%$ was chosen as the threshold for wetness presence.

212 3.2. Concentration Profiles of SO_2

213 Composite vertical profiles of SO_2 concentration at four levels are presented in
214 Fig. 3. The variation patterns of the 3-hourly profiles can be classified into two groups,
215 the ascending (Fig. 3a) and the descending (Fig. 3b) with increasing time. During
216 daytime, the profile of SO_2 concentration showed a generally increasing trend with
217 increasing time, and reached a maximum at 15:00 (Fig. 3a); in contrast, the profile of
218 SO_2 concentration at nighttime showed a generally decreasing trend with increasing
219 time, and reached a minimum at 3:00 in the wee hours of the morning. Such diurnal
220 variation is attributed to the development of mixed layers and SO_2 pollutant transfer
221 from regional areas.

222 Above the canopy, whether daytime or nighttime, there were positive gradients
223 for all the profiles. In each profile, it can be seen that the minimum concentration of
224 SO_2 always occurred at 26 m where is close to the height of canopy. Occasionally,
225 negative gradients of SO_2 were occurred when the SO_2 concentrations at 26 m were
226 larger than those at upper levels and consequently, resulted in the $V_d < 0$. This
227 probably originated from the random measurement errors under conditions of low
228 SO_2 concentration and weak turbulence where random measurement error leading to
229 random errors in deposition velocity becomes high (e.g. Matsuda et al., 2006). By
230 means of choosing height intervals sufficiently large to yield concentration differences
231 and adequate criteria for data screening, only small number of available data obtained
232 under such situation.

233 Within the canopy, the SO₂ concentration was only measured at 10 m, where is
234 lower than the bottom of the canopy. Whether day or night, data showed that the SO₂
235 concentrations at 10 m were slightly larger than those at 26 m. The vertical profiles of
236 pollutants were influenced by meteorological conditions, canopy structures and other
237 factors (e.g. Meng et al., 2007). In this study, it is considered that this phenomenon
238 might be caused by remained SO₂ in the canopy for the weak wind speed (e.g. Lovett
239 and Lindverg, 1992; Walton et al., 1997), though no meteorological measurement was
240 conducted with the canopy during the study period. Besides, the SO₂ concentration
241 gradient could not be determined for only one level with the canopy, hence, the effect
242 of soil uptake for SO₂ from the observation is out of discussion in this study.

243 3.3. Evaluation of SO₂ dry deposition velocity

244 The hourly values of V_d obtained in this study are presented in Fig. 4a which
245 shows such a large diurnal variation that dry deposition velocities were higher during
246 daytime than at night in all periods. The values of V_d are so widely distributed that the
247 maximum value of observed V_d can reach 3 cm s⁻¹; in contrast, the minimum V_d is
248 negative. Fig. 4b showed the composite diurnal cycle of SO₂ V_d . During daytime, the
249 dry deposition velocity of SO₂ ranged from 0.28 cm s⁻¹ to 0.89 cm s⁻¹ with a mean
250 value of 0.61 cm s⁻¹; during nighttime, the V_d ranged from 0.22 cm s⁻¹ to 0.36 cm s⁻¹
251 with a mean value of 0.27 cm s⁻¹. In addition, the standard deviations of SO₂ V_d
252 during daytime were larger than those during nighttime.

253 It was found that the diurnal variation of V_d was highly dependent on incoming
254 solar radiation so that the daytime V_d increased with the solar radiation and the daily
255 maximum value of V_d always occurred around noon when the incoming solar
256 radiation was the most intense. Due to the absence of solar radiation during nighttime,
257 the V_d were usually smaller than those during daytime. The V_d also increased with u^* .

258 The larger u^* resulted in not only smaller aerodynamic resistance but also
259 non-stomatal resistance (Zhang et al., 2003a).

260 3.4. Monthly variations of SO₂ deposition velocity over dry and wet canopies

261 Fig. 5 presents the composite diurnal cycles of V_d over dry and wet canopies for
262 each month. It can be seen that both V_d show obviously diurnal variations and in both
263 the daytime and nighttime, V_d over wet canopy were higher than those over dry
264 canopy. Over dry canopy, stomatal uptake is an important sink of SO₂ when
265 vegetation is biologically active. During daytime, stomata are opening for
266 photosynthesis and, in the mean while, stomatal resistance for SO₂ deposition was
267 reduced. In the absence of solar radiation during nighttime, however, the stomata
268 would be close and resulted in huge stomatal resistance. Over wet canopy, wetness
269 would decrease stomatal uptake, but substantially increases non-stomatal uptake
270 (Feliciano et al., 2001; Zhang et al., 2003a, 2003b). In the presence of aqueous layers
271 on the surface, it is assumed that stomatal uptake is not important in light of stomata
272 blocking by water drops and the presence of weak solar radiation (Zhang et al.,
273 2003a). On the contrary, SO₂ is a reasonably soluble gas in pure water and is
274 effectively removed at higher rates under moist conditions in this study. Therefore, it
275 is considered that higher deposition velocities over wet canopy were mainly caused by
276 non-stomatal uptake.

277 The comparison of monthly diurnal variations of V_d showed that the daytime V_d
278 in February were smaller than those in the other two months. The results implied that
279 V_d tends to be larger with a larger leaf area index (LAI) in March and April. That the
280 canopy resistance, which is the major factor determining the V_d in vegetation areas,
281 has been shown to be correlated with individual leaf stomatal resistance divided by
282 the LAI is also demonstrated in other studies (Kelliher et al., 1995; Wilson et al.,

283 2002). In addition, more intense radiation also results in the increasing of V_d (Jitto et
284 al., 2007). As shown in Table 1, the mean daytime value of solar radiation in March
285 and April are 1.5 times of that in February.

286 Fig. 5 also showed that the V_d in the nighttime is similar in each month over dry
287 and wet canopies respectively. It implies that LAI is not the major variable
288 determining the V_d in the nighttime.

289 3.5. Comparisons with the observations from other studies over forest

290 A review of published results for V_d of SO_2 determined by several methods over
291 forest at different locations is presented in Table 2. The mean values of V_d range from
292 about 0.1 cm s^{-1} over deciduous forest during winter nighttime to 2.3 cm s^{-1} over
293 coniferous forest during wet daytime. It reveals general phenomena that daytime V_d is
294 higher than that in nighttime and V_d over canopy higher than that over dry canopy.
295 The daytime V_d (0.61 cm s^{-1}) during wintertime in this study is lower than (about 1 cm
296 s^{-1}) of Finkelstein et al. (2000) during summertime. Over dry canopy, the daytime V_d
297 (0.47 cm s^{-1}) over broadleaf evergreen forest in this study is higher than that (0.21 cm
298 s^{-1}) of dry season over deciduous forest in Thailand (Matsuda et al., 2006) and that
299 (0.3 cm s^{-1}) over deciduous forest in Netherlands (Erisman, 1994). This is probably
300 caused by forest conditions in seasonal differences when deciduous forest is more
301 active during summer time and defoliated during wintertime. On the other hand, over
302 wet canopy, daytime V_d over broadleaf evergreen forest in Taiwan (this study) and that
303 over deciduous forest in Thailand (Matsuda et al., 2006) are at the same level which
304 means not so large differences in wet canopy V_d between Taiwan and Thailand.
305 However, significant differences of V_d over wet canopy are also observed over
306 coniferous forest and deciduous forest in Erisman (1994). From discussions above, it
307 indicates that, in addition to the deviations caused by the random and systematic

308 instrumental errors and inaccuracies following from theoretical assumptions, V_d also
309 varies with underlying forest physiology, time of day and season (e.g. Brook et al.,
310 1999). Compared with the results listed in this Table, the observed V_d in this study are
311 reasonable.

312 3.6. Comparisons with modeled deposition velocities

313 Zhang et al. (2003a, 2003b, denoted as Z03 hereafter) proposed a resistance
314 analogy parameterization for calculating gaseous dry deposition based on study
315 results over 5 different vegetation types in the North America. The structure of Z03 in
316 the parameterization of r_c is separated into two parallel paths; one is stomatal
317 resistance (r_{st}) with its associated mesophyll resistance (r_m), and the other is
318 non-stomatal resistance (r_{ns}). In addition, look-up tables, classified according to the
319 land types, seasonal category, and canopy wetness, were also used to calculate SO_2
320 dry deposition velocity. More detail is referred to Zhang et al. (2002, 2003a). In this
321 study, Z03 was applied to evaluate SO_2 dry deposition velocity and discuss the
322 applicability to the mountain forest area.

323 Fig. 7 shows the hour values of observed and modeled V_d of Z03 over dry and
324 wet canopy. For dry canopy, Z03 seemed to predict reasonable SO_2 V_d during
325 nighttime, but underestimated SO_2 V_d during daytime. Similar result was also reported
326 by Zhang et al. (2003a) for SO_2 V_d in deciduous forest. The correlation coefficient
327 between the observed and the modeled V_d is 0.96, the bias is -0.05 cm s^{-1} and the
328 RMSE is 0.07 cm s^{-1} . For wet canopy, Z03 slightly overestimated the V_d not only
329 during daytime but also in the nocturnal. The correlation coefficient between the
330 observed and the modeled V_d is 0.9, the bias is 0.06 cm s^{-1} and the RMSE is 0.12 cm
331 s^{-1} . In general, Z03 can estimate SO_2 V_d for dry and wet canopy approximately, not
332 only the magnitude but also the diurnal pattern of V_d .

333 Z03 also provided a lookup table for an indication of the typical V_d of SO_2 under
334 different meteorological conditions with the dominant values of meteorological
335 variables in each category of land use and the predictions are consistent with the
336 published measurements. Therefore the results in this study may be reliably compared
337 with the predicted values in Zhang et al. (2003a). It is noticed that this prediction was
338 designed for summer season. For the location of Taiwan, however, is in the
339 subtropical area where the climate is hotter and wetter, comparison of the result and
340 the prediction is with reference value for the scientists in this community. In this
341 prediction, u^* values of the V_d over evergreen broadleaf trees are prescribed as 0.7 for
342 dry or rainy days, 0.35 for dry or rainy nights and 0.2 for dewy nights. The observed
343 u^* in this study were similar with those in Z03 except the value for dry or rainy days
344 where 0.51 for dry or rainy days, 0.31 for dry or rainy nights and 0.22 for dewy nights,
345 and more 0.29 for dewy days. Moreover, the V_d were 0.88 (0.22) cm s^{-1} under dry day
346 (night) and 2.47 (1.12) cm s^{-1} under rainy day (night). Moreover, the predicted
347 maximum V_d was also presented such that the maximum V_d over dry canopy is 1.7 cm
348 s^{-1} and 3.9 cm s^{-1} over wet canopy. Overall, the observed V_d in this study are close to
349 the predictions of Zhang et al. (2003a). Though the observed V_d over wet canopy is
350 smaller than the prediction, it is noted that the prediction of Zhang et al. (2003a)
351 excluded the results under dew when the V_d is small with stable atmospheric
352 conditions (Zhang et al., 2003b). In addition, all the values of observed V_d are smaller
353 than the predictions.

354 3.7. Evaluation of canopy resistance of evergreen broadleaf forest

355 Fig. 8 presents the composite observed $r_a + r_b$ during the study period calculated
356 from Eqs. (5) and (6). A typical “U” shaped diurnal variation was observed with the
357 minimum mean value occurring at 32 s m^{-1} approximately around noon and the mean

358 values being larger than 65 s m^{-1} during nighttime. The values of $r_a + r_b$ were larger
359 during nighttime due to smaller u^* , when the atmosphere was under stable conditions
360 compared to those during daytime. In addition, it was also found that $r_a + r_b$ were
361 larger under dew conditions compared to rain conditions for the smaller u^* (Zhang et
362 al., 2003b).

363 Fig. 8 also presents the composite observed r_c over dry and wet canopies during
364 the study period, calculated from Eq. (4) with the observed values of V_d , r_a and r_b . The
365 diurnal pattern of r_c is similar to that of $r_a + r_b$ in that the minimum occurred around
366 noon. The values of r_c over day (wet) canopy were about $225 (112) \text{ s m}^{-1}$ in the
367 daytime and $735 (366) \text{ s m}^{-1}$ in the nighttime. Compared to the results of Zhang et al.
368 (2003b) observed at mixed forest and deciduous forest sites for a full growing season,
369 the estimated r_c in this study are relatively large. Note that the observation in this
370 study was conducted during the transition between late winter and early spring, when
371 the environmental conditions, such as solar radiation and air temperature, were not the
372 most suitable for vegetative activity. Therefore, it is deduced that the representative
373 value of r_c for evergreen broadleaf forest in growing season should be smaller than
374 that in this study.

375 In studying dry deposition of air quality models over areas covered by vegetation
376 (e.g. Wesely, 1989; Zhang et al., 2003a, 2003b) canopy resistance is often partitioned
377 into two resistances, the stomatal resistance (r_{st}) and the non-stomatal resistance (r_{nst}).
378 Separation of these processes is important in estimating the accurate representation of
379 diurnal variations of dry deposition because stomatal uptake usually occurs during the
380 daytime, during which time it predominates over non-stomatal uptake for many
381 chemical species; in contrast, during nighttime, when the stomata are closed, r_c over
382 dry canopy was mainly controlled by r_{nst} (Zhang et al., 2003a). In this study, it was
383 found that the daytime r_c were mainly determined by the solar radiation, which is

384 consistent with those in Wesely (1989) and Zhang et al., (2003a). Because the
385 vegetation obtains the required nutrients for living through photosynthesis which acts
386 with the Photosynthetically Active Radiation of solar radiation, it is expected that the
387 variation of daytime r_c will follow the intensity of the solar radiation. During the study
388 period, the air temperatures were lower than 20 °C which is relative low compared to
389 the optimal air temperature of evergreen broadleaf forest, 30 °C, assigned in Zhang et
390 al. (2003a). This situation limited the vegetative growth and showed that r_c is only
391 slightly dependent on air temperature. A similar phenomenon was also observed for r_c
392 dependency on vapor pressure deficit. Lower air temperatures result in a lower vapor
393 pressure deficit so that during the study period the values of the vapor pressure deficit
394 were lower than 900 pa.

395 During nighttime, r_c was considered to be equal to r_{nst} when the r_{st} is less
396 important and can be ignored. Fig. 9 shows canopy resistance versus friction velocity
397 during nighttime over dry canopy where r_c is limited to within 5000 s m⁻¹ and u^*
398 ranges from 0.1 to 1 m s⁻¹. It shows that r_c values decreased as u^* increased, which is
399 consistent with other studies (Zhang et al., 2003b; Matsuda et al., 2005). Large u^*
400 values, which represent strong turbulence, can facilitate the transportation of SO₂
401 between the atmosphere and the underlying canopy (Hicks et al., 1989), thus
402 enhancing non-stomatal uptake. It is also found that nighttime r_c versus RH over dry
403 canopy showed that r_c only slightly depends on RH. There was also a clear tendency
404 towards a smaller and less variable r_c for RH > 90% (Fig. 8) while, for RH < 90%, r_c
405 was highly variable. The relationship between r_c and RH for SO₂ were similar to the
406 relationships found by Feliciano et al. (2001) and Zhang et al. (2003b).

407 **4. Conclusion**

408 A study on SO₂ dry deposition was performed in an evergreen broadleaf forest in

409 central Taiwan. The study period was from the beginning of February to the end of
410 April 2008 which was the drier and colder season in this area. To evaluate the SO₂ dry
411 deposition velocity with the gradient method, the multilevel SO₂ concentrations,
412 meteorological variables and energy fluxes were measured by an eddy covariance
413 system and other instruments during the same period. The results show that mean SO₂
414 dry deposition velocity in the daytime is 0.61 cm s⁻¹ and 0.27 cm s⁻¹ in the nighttime.
415 Compared with those results in the literature that were carried out over forest areas
416 using the observations and resistance methods, the results in this study are reasonable.

417 It was observed that the deposition velocity was larger over wet canopy than
418 over dry canopy. Over wet canopy, the mean dry deposition velocities of SO₂ were
419 estimated to be 0.83 cm s⁻¹ during daytime and 0.47 cm s⁻¹ during nighttime; and 0.44
420 cm s⁻¹ during daytime and 0.19 cm s⁻¹ during nighttime over dry canopy. It is clear
421 that higher deposition velocities were mainly caused by non-stomatal uptake of wet
422 canopy, especially during nighttime. A tendency was also observed that dry deposition
423 velocity increases with LAI and solar radiation from the comparison of the monthly
424 data.

425 Compared with the predictions of Zhang et al., (2003), the results showed that
426 the observed V_d are in agreements with the predictions over dry and wet canopies. The
427 observed u^* in this study were similar with those in Z03 except the value for dry or
428 rainy days where 0.51 for dry or rainy days, 0.31 for dry or rainy nights and 0.22 for
429 dewy nights, and more 0.29 for dewy days.

430 The patterns of diurnal variation for $r_a + r_b$ and r_c showed obvious diurnal
431 variations so that the median (geometric mean) of derived r_c during daytime are 233
432 (266) m s⁻¹ over dry canopy and 147 (146) m s⁻¹ over wet canopy. During daytime,
433 solar radiation is the major meteorological variable determining the SO₂ uptake so
434 that the values of canopy resistance were minimized around noon and increased as

435 time became close to the early morning and late afternoon. It was found that wetness
436 and high humidity enhanced canopy SO₂ uptake and thus decreased non-stomatal
437 resistance. For non-stomatal resistance, evident dependencies were observed in the
438 friction velocity and relative humidity.

439 This study provides valuable information regarding dry deposition velocity for
440 SO₂ in evergreen broadleaf forest, being the dominant species of forest worldwide but
441 with relatively few studies focused on this area. The knowledge of dry deposition
442 velocity for SO₂ obtained in this study, such as the diurnal and monthly variations,
443 effects resulting from meteorological variables and vegetation physiology, not only
444 helps to extend the observation to longer periods and to different locations, but is also
445 useful in the examination and evaluation of parameterization.

446

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447

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451

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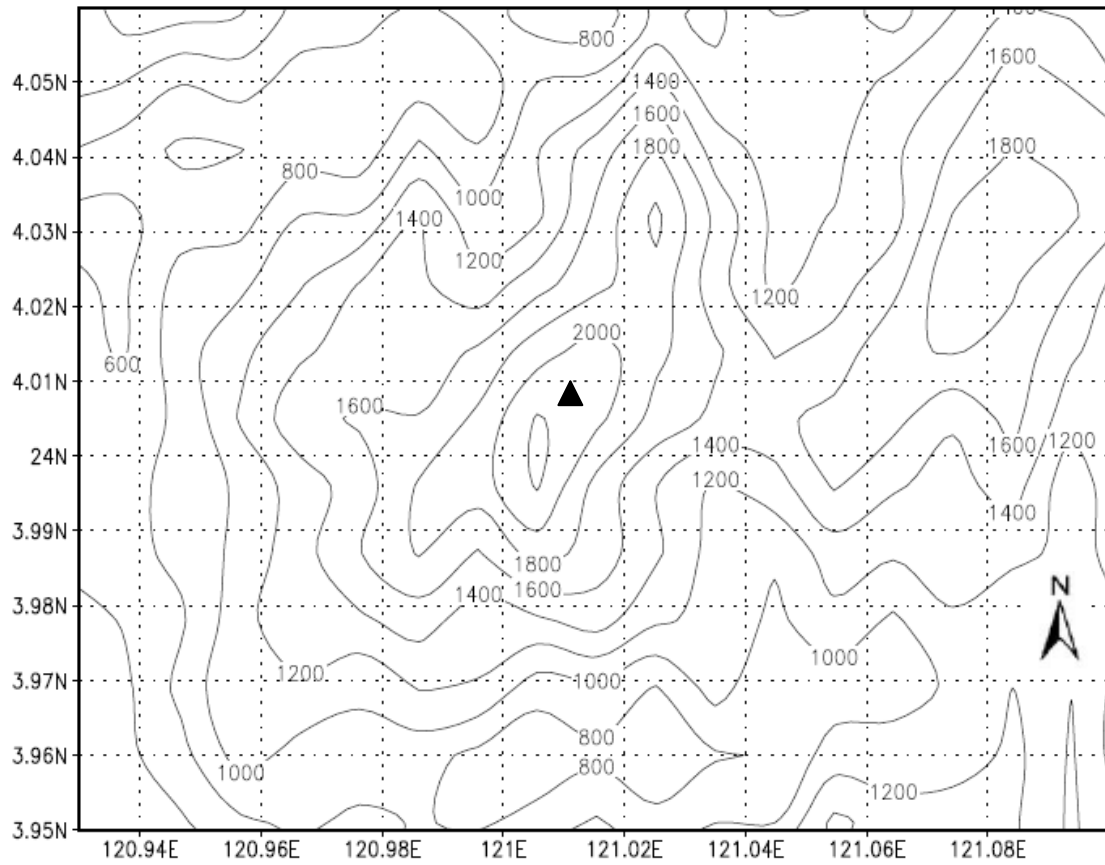


Fig. 1. Topographic map of the flux site with the position of the eddy covariance tower shown by a solid triangle. Contour interval is 200 m.

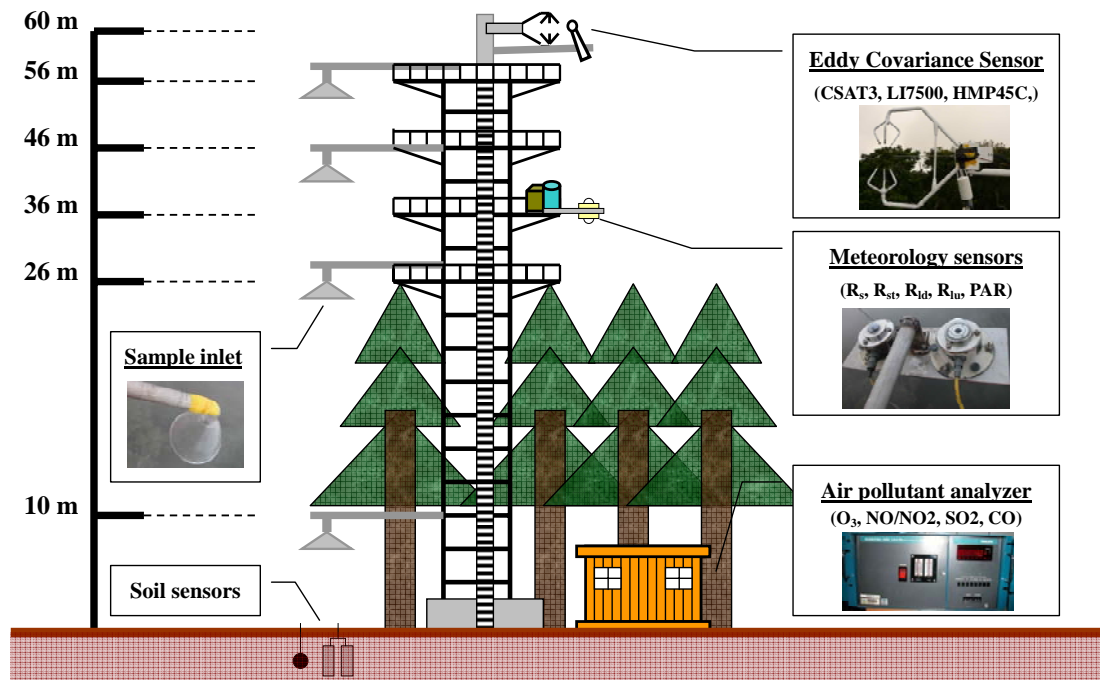


Fig. 2. The sketch of Huisun Forest flux tower in Taiwan.

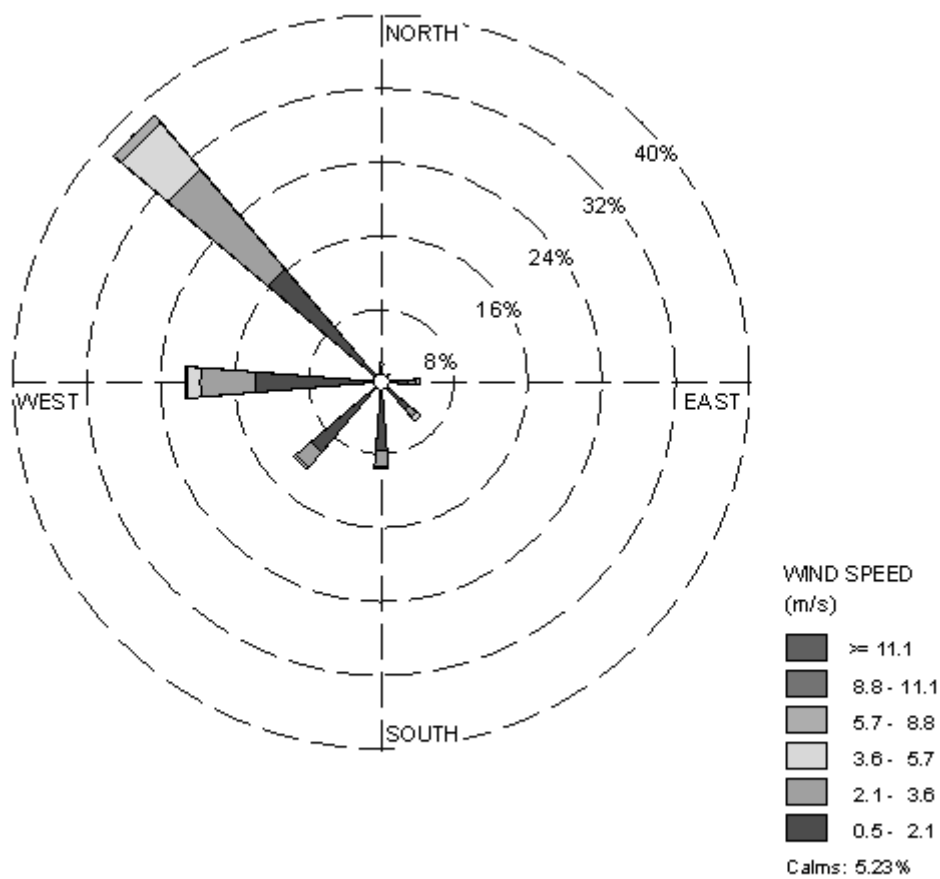
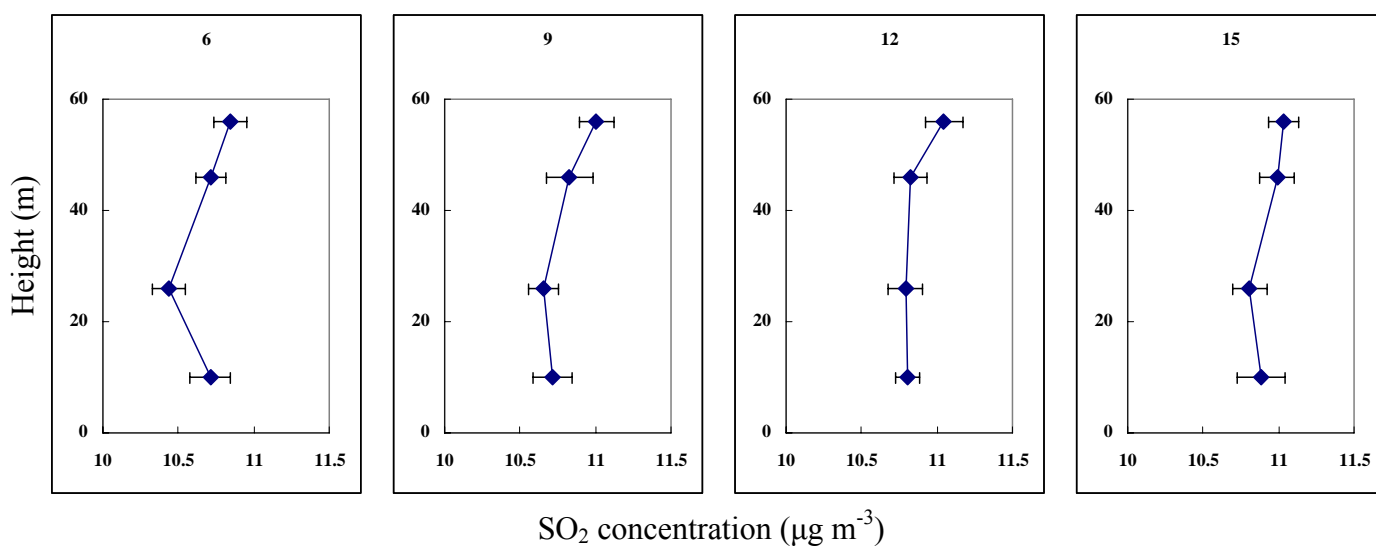


Fig. 3. Relative distribution of the wind speeds and directions for the time period February 1–April 30 2008 at the study site.

a) Ascending



b) Descending

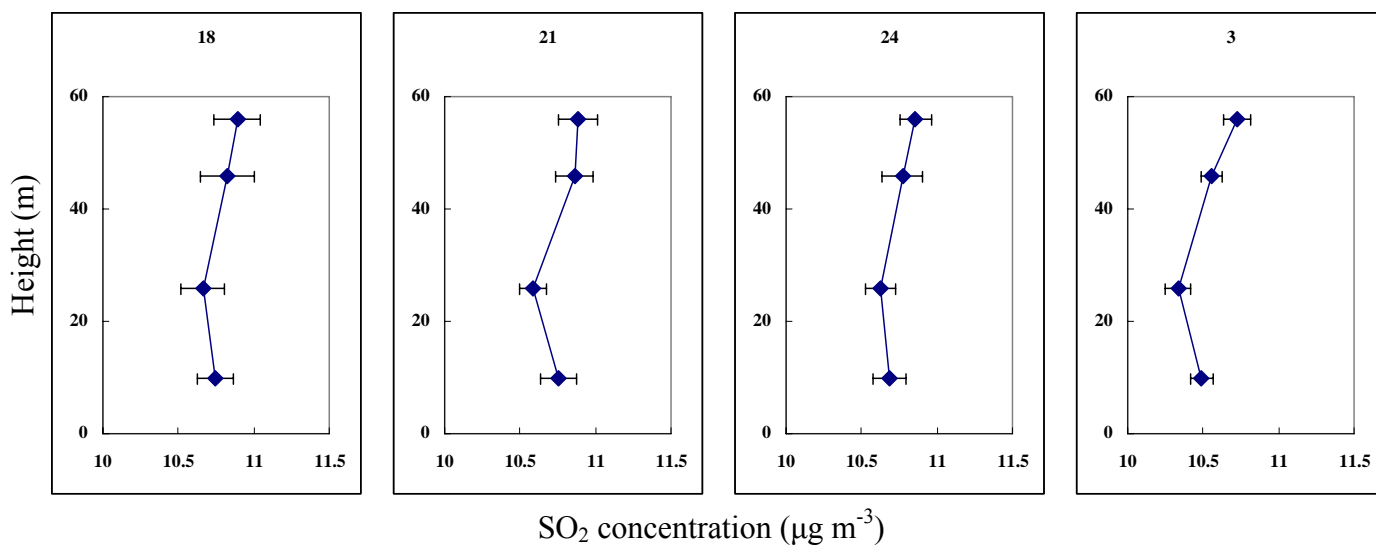
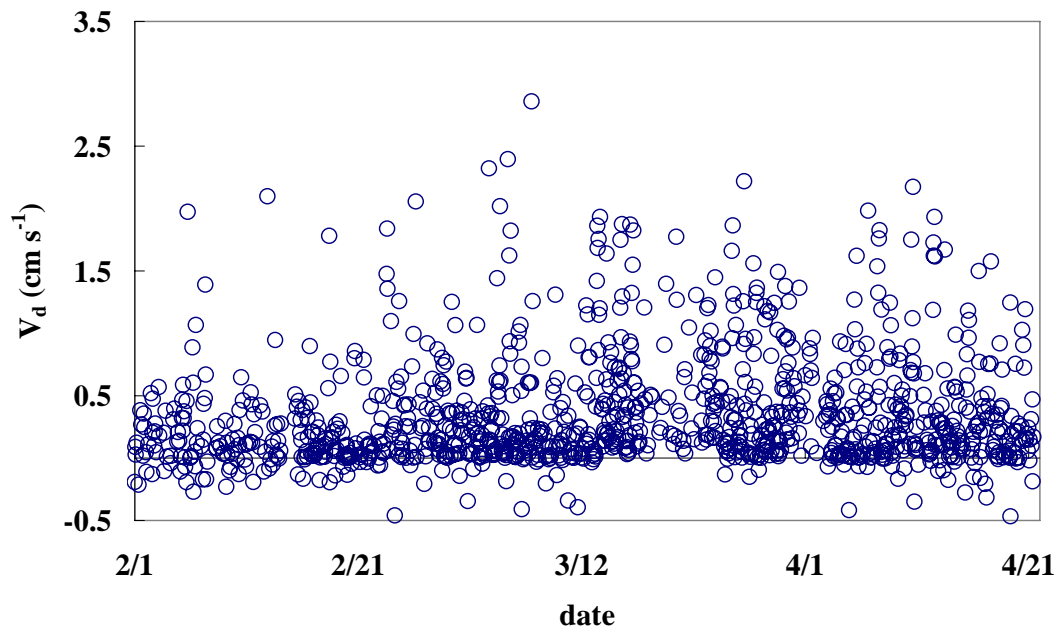


Fig. 4. The composite vertical profiles of SO₂ concentration measured at 10 m, 26 m, 46 m and 56 m with 3-hours bin.

a)



b)

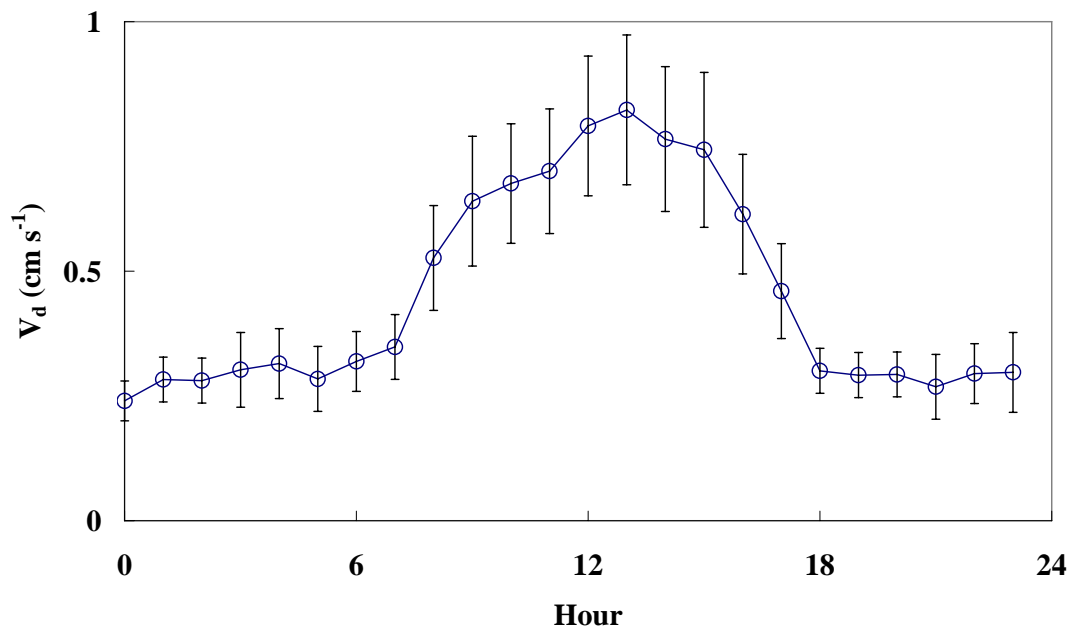


Fig. 5. The time series and composite diurnal cycle of SO₂ dry deposition velocity from observations, where the high–low bars denote standard deviations of observations.

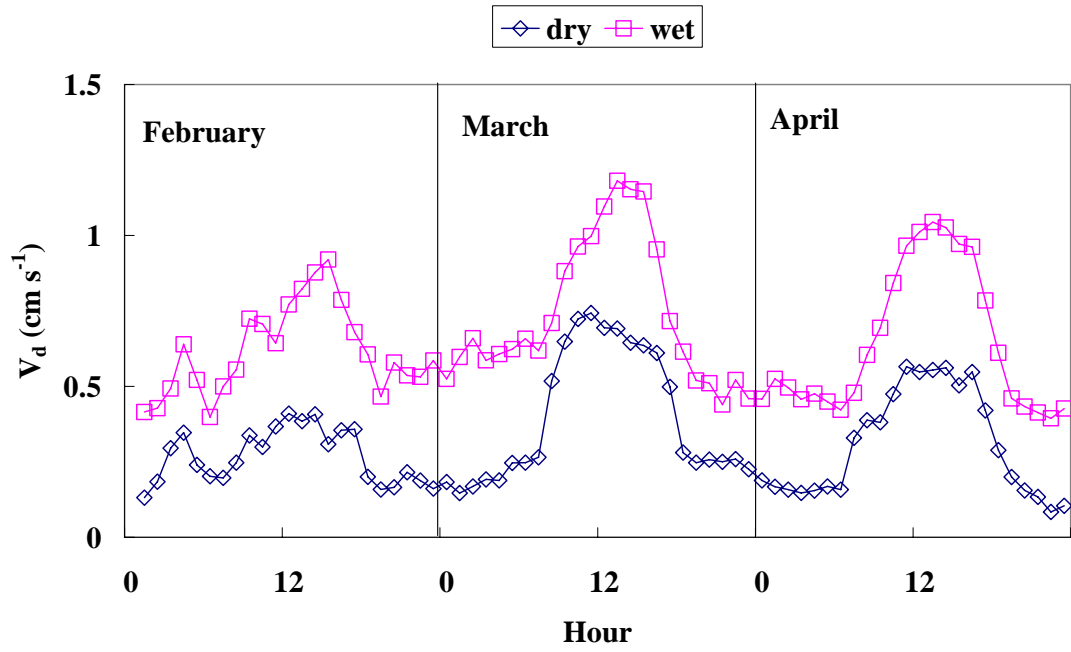


Fig. 6. Composite diurnal variations of SO₂ dry deposition velocity over dry and wet canopies at each month.

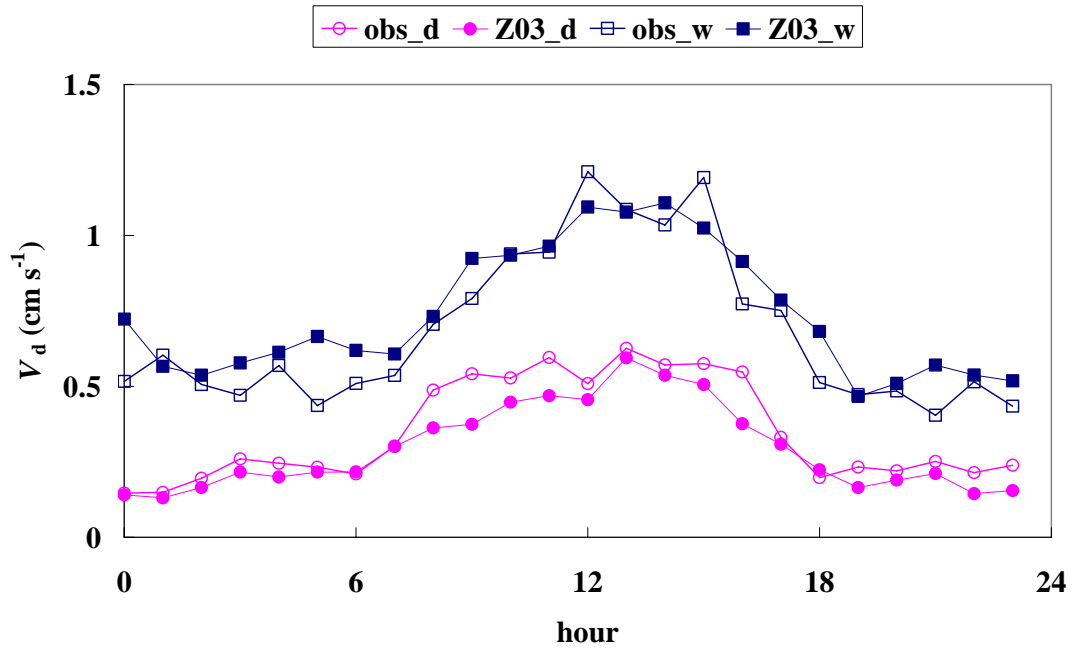
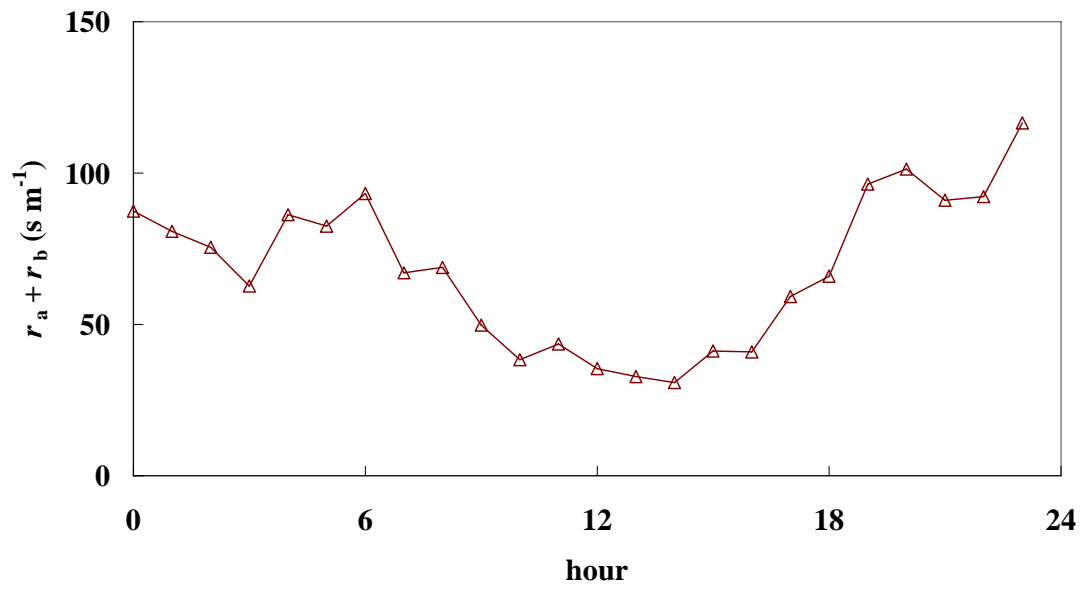


Fig. 7. The composite hour values of predicted V_d of Z03 versus the observations over dry and wet canopies.

a)



b)

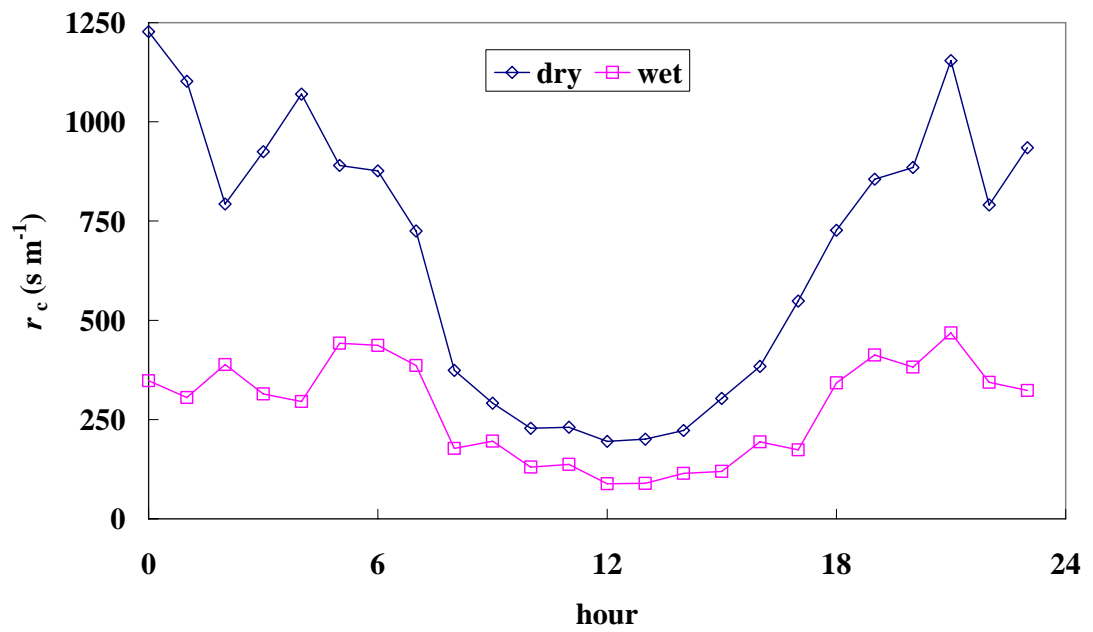


Fig. 8. Composite diurnal cycles of observed a) aerodynamic and quasi-laminar layer resistances and b) canopy resistances over dry and wet canopies.

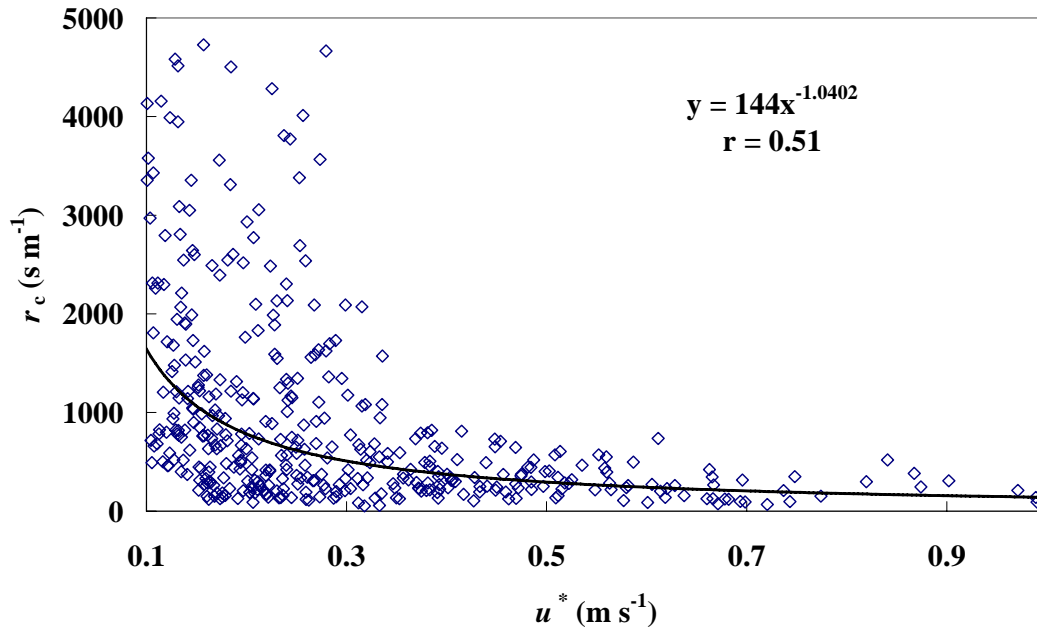


Fig. 9. The observed canopy resistance versus friction velocity over dry canopy during nighttime.

Table 1. Mean values of meteorological observation at flux tower during the study period in 2008.

	Daytime			Nighttime		
	Feb.	Mar.	Apr.	Feb.	Mar.	Apr.
Wind speed (m s^{-1})	2.3	2.1	1.8	2.4	1.8	1.8
Friction velocity (m s^{-1})	0.41	0.35	0.33	0.36	0.26	0.28
Solar radiation (W m^{-2})	181	261	254	-	-	-
Temperature ($^{\circ}\text{C}$)	7	9.2	14.5	6.7	7.8	13.6
Relative humidity (%)	79	82	94	75	74	91
Total precipitation (mm)	16.6	8.6	6	10.6	18.6	7

Table 2. Comparisons of dry deposition velocity for SO₂ in this study and those of other studies over forest areas.

Forest types	Location	Condition	methods	V _d (cm s ⁻¹)	Reference
Broadleaf evergreen forest	Taiwan	Early spring (Feb.-Apr.)	Gradient method	Day 0.61; Night 0.27 Dry canopy Day 0.44; Night 0.19 Wet canopy Day 0.83; Night 0.47	This study
Deciduous forest (teak)	Thailand	Dry season (Jan. – Apr.) Wet season (May – Aug.)	Gradient method	Dry season Day 0.21; Night 0.09 Wet season Day 1.17; Night 0.34	Matsuda et al., 2006
Coniferous forest (pine)	Japan	Sep. – Nov.	Bowen ratio	Day 0.9	Matsuda et al., 2002
Coniferous forest (pine)	Sweden	Summer and winter	Enclosure technique	0.33	Granat and Richter, 1995
Coniferous forest (pine)	Japan	Jul. – Dec.	Resistance method	0.4	Matsuda et al., 2001
Deciduous forest	Japan	Jul. – Dec.	Resistance method	0.36	Matsuda et al., 2001
Deciduous forest	USA	Apr. – Oct.	Resistance method	Day 1.04 Night 0.3	Finkelstein et al., 2000
Mixed Coniferous-deciduous forest	USA	May – Oct.	Resistance method	Day 1.01 Night 0.27	Finkelstein et al., 2000
Mixed Coniferous-deciduous forest	China	Feb., May, Aug. and Dec.	Resistance method	Summer Day 0.4 Winter Day 0.15	Xu and Carmicheal, 1998
Coniferous forest	Netherlands	Yearly	Resistance method	Dry Day 0.7; Night 0.7 Wet Day 2.3; Night 2.5	Erisman, 1994
Deciduous forest	Netherlands	Winter	Resistance method	Dry Day 0.3 Wet day 0.6	Erisman, 1994