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# 1 Soil ozone deposition: dependence of soil resistance to soil texture

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3 P. Stella<sup>1,\*</sup>, B. Loubet<sup>2</sup>, C. de Berranger<sup>3</sup>, X. Charrier<sup>3</sup>, E. Ceschia<sup>4</sup>, G. Gerosa<sup>5</sup>, A.  
4 Finco<sup>5</sup>, E. Lamaud<sup>6</sup>, D. Serça<sup>7</sup>, C. George<sup>8</sup>, R. Ciuraru<sup>2</sup>

5  
6 [1] UMR SAD-APT, AgroParisTech, INRA, Université Paris-Saclay, 75005, Paris, France

7 [2] UMR1402 ECOSYS, INRA-AgroParisTech, Université Paris-Saclay, 78850 Thiverval-  
8 Grignon, France

9 [3] INRA UE FERLUS, Les Verrines – CS 80006, 86600 Lusignan, France

10 [4] CESBIO, UMR 5126 – CNES-CNRS-UPS-IRD – 18 avenue Edouard Belin 31401  
11 Toulouse cedex 9, France

12 [5] Dipartimento di Matematica e Fisica, Università Cattolica del Sacro Cuore, via Musei 41,  
13 25121 Brescia, Italia

14 [6] UMR 1391 ISPA, INRA-Bordeaux Sciences Agro, F-33140 Villenave d'Ornon, France

15 [7] Laboratoire d'Aérodologie, Université de Toulouse, CNRS, UPS, France

16 [8] Université de Lyon 1 CNRS, UMR5256, IRCELYON, Institut de Recherches sur la  
17 Catalyse et l'Environnement de Lyon, F-69626 Villeurbanne, France

18 [\*] Correspondence to: P. Stella ([patrick.stella@agroparistech.fr](mailto:patrick.stella@agroparistech.fr))

## 19 20 Abstract

21 Soil deposition is an essential pathway for tropospheric ozone ( $O_3$ ) removal, but its  
22 controlling factors remain unclear. Here, we explored the variability of soil  $O_3$  resistance in  
23 response to soil texture. To this aim, data of  $O_3$  deposition over bare soil obtained from  
24 micrometeorological measurements under contrasted meteorological conditions for five sites  
25 were used. The results obtained are twofold: (i) soil resistance ( $R_{soil}$ ) increased with soil  
26 surface relative humidity ( $RH_{surf}$ ), but (ii) this relationship exhibited large site-by-site  
27 variability. Further analysis showed that the minimum soil resistance (corresponding to  
28 completely dry soil surface or  $RH_{surf} = 0\%$ ) and the increase of  $R_{soil}$  with  $RH_{surf}$  are both  
29 linked to soil clay content. These results can be explained by (i) the soil surface available for  
30  $O_3$  deposition at a microscopic scale which is a function of the soil specific surface area, and  
31 (ii) the capacity of a soil to adsorb water according to its clay content and therefore to reduce  
32 the surface active for  $O_3$  deposition. From these results, a new parameterization has been  
33 established to estimate  $R_{soil}$  as a function of  $RH_{surf}$  and soil clay fraction.

34

35 **Keywords:** Ozone; soil resistance; clay content; relative humidity.

36

## 37 1 – INTRODUCTION

38 Since the pre-industrial era, concentrations of tropospheric ozone (O<sub>3</sub>) have sharply increased  
39 in the atmosphere. It is a well-known greenhouse gas responsible for a positive radiative  
40 forcing of 0.40 W m<sup>-2</sup> i.e., around 20% of the total radiative forcing attributed to human  
41 activities, and the largest contributor to radiative forcing after long-lived trace gases (CO<sub>2</sub>,  
42 N<sub>2</sub>O, CH<sub>4</sub>, and halocarbons) (IPCC, 2013). Yet, due to its oxidative capacity, O<sub>3</sub> is also a key  
43 compound in atmospheric chemistry (Monks, 2005) and a widespread secondary pollutant. It  
44 is responsible for the oxidation of numerous compounds (e.g., Lee et al., 1996; Ahmad et al.,  
45 2000) and for negative impacts on human health (Ito et al., 2005; Hazucha and Lefohn, 2007;  
46 Doherty et al., 2017). On terrestrial ecosystems, O<sub>3</sub> penetrates through plant stomata and  
47 induces a range of metabolic changes such as a decrease of photosynthetic capacity, alteration  
48 of plant biomass and structure, stomatal closure, and acceleration of senescence (e.g.,  
49 Karnosky et al., 2003; Paoletti, 2005; Felzer et al., 2007; Dizengremel et al., 2008; Booker et  
50 al., 2009; Wittig et al., 2009; Ainsworth et al. 2012, Lombardozzi et al., 2013). All these  
51 alterations lead to the decrease of ecosystem productivity and crop yield losses (e.g.,  
52 Ainsworth et al., 2012; Lombardozzi et al., 2015; Franz et al., 2017), which in turn could  
53 contribute indirectly to global warming due to the alteration of global carbon cycle (Felzer et  
54 al., 2007; Sitch et al., 2007).

55 Since O<sub>3</sub> is weakly soluble in water, it is mainly deposited through dry deposition on  
56 terrestrial ecosystems (Fowler et al., 2009), which is the only net removal pathway of O<sub>3</sub> from  
57 the atmosphere and therefore an important process governing the tropospheric O<sub>3</sub> budget  
58 (Stevenson et al., 2006; Wild, 2007). Many studies have been carried out over natural  
59 ecosystems and agroecosystems in order to (i) understand the processes governing O<sub>3</sub> dry  
60 deposition, (ii) establish parameterizations for O<sub>3</sub> deposition, and (iii) quantify the terrestrial  
61 O<sub>3</sub> sink (e.g., Zhang et al., 2006; Coyle et al., 2009; Lamaud et al., 2009; Stella et al., 2011a,  
62 2013; Fares et al., 2012; Launiainen et al., 2013; Clifton et al., 2017; Freire et al., 2017).  
63 Deposition occurs through stomatal and non-stomatal (i.e., soil and cuticular) pathways. Due  
64 to their dependence on leaf area index (LAI), O<sub>3</sub> deposition mainly occurs through stomatal  
65 and cuticular pathways on fully developed canopies, while transfer from the atmosphere  
66 toward the ground is reduced when the canopy height and LAI increase (van Pul and Jacobs,  
67 1994; Zhang et al., 2002; Massman, 2004; Tuovinen et al., 2004; Stella et al., 2011a, 2013).  
68 Hence, strong efforts have been done to understand the processes governing stomatal (e.g.,  
69 Emberson et al., 2000) and cuticular deposition (e.g., Zhang et al., 2002; Altimir et al., 2004,

70 2006; Coyle et al., 2009; Cape et al., 2009; Lamaud et al., 2009; Potier et al., 2015), although  
71 these processes are still not well described and no consensual parameterization exists.

72 Over fully developed canopies, soil deposition is the smallest contributor to total deposition  
73 and processes governing this deposition pathway received little attention. Nevertheless, it  
74 cannot be neglected for short or sparse canopies, and of course during bare soil periods. Yet,  
75 Stella et al. (2013) reported that soil O<sub>3</sub> deposition was the main deposition pathway and  
76 represented 55% of the total O<sub>3</sub> deposition over an agricultural field for a 2 year period.

77 Few studies have investigated the processes governing soil deposition. Some authors  
78 associated soil O<sub>3</sub> resistance ( $R_{\text{soil}}$ ) to soil water content (e.g., Bassin et al., 2004; Massman,  
79 2004; Meszaros et al., 2009), but the suggested parameterizations are not able to correctly  
80 estimate O<sub>3</sub> deposition during bare soil or growing season periods (Stella et al., 2011b, 2013).  
81 From measurements carried out over bare soil, Stella et al. (2011b) showed that  $R_{\text{soil}}$  was  
82 linked to soil surface relative humidity ( $\text{RH}_{\text{surf}}$ ), but the relationships proposed seems to be  
83 site-dependent (Stella et al., 2011a).

84 This study aims now to explore the relationships between  $R_{\text{soil}}$  and  $\text{RH}_{\text{surf}}$  and its dependence  
85 on soil texture, with data obtained only over bare soil are used. A new parameterization of  
86  $R_{\text{soil}}$  accounting for soil texture is proposed.

## 87 **2 – MATERIAL AND METHODS**

### 88 **2.1 – Site descriptions, datasets, and measurements**

89 Standard meteorological variables (i.e., global radiation ( $R_g$ ), wind speed ( $u$ ), air temperature  
90 ( $T_a$ ), air relative humidity ( $\text{RH}_a$ ) as well as turbulent fluxes (sensible ( $H$ ) and latent (LE) heat  
91 fluxes, and momentum ( $\tau$ ) flux from which is deduced friction velocity ( $u_*$ )) and O<sub>3</sub>  
92 deposition velocities ( $V_d$ ) measured by the eddy-covariance method were collected over five  
93 different sites during bare soil periods. Site, experimental set-up, and data processing were  
94 already described in previous studies for La Crau (Michou et al., 2005), Lamasquère (Béziat  
95 et al., 2009; Stella et al., 2011a), and La Cape Sud (Stella et al., 2009, 2011a).

96 The first dataset was collected between 20 April and 31 May 2001 in the semi-arid part of the  
97 La Crau plain, France (43°34'N, 4°49'E). The site consisted of an almost bare soil with  
98 mainly pebbles. Ozone fluxes were measured by eddy-covariance with a fast-response O<sub>3</sub>  
99 chemiluminescent analyzer (OS-G-2, Güsten, 1992). Its calibration was continuously checked  
100 against a slow-response O<sub>3</sub> monitor (O<sub>3</sub> 41M, Environnement SA, FR).

101 The second dataset corresponds to measurements performed during four bare soil periods (24  
102 April 2008 to 26 May 2008, 20 November 2008 to 18 December 2008, 14 November 2009 to  
103 12 May 2010, 29 September 2010 to 9 November 2010) over an agricultural field located at  
104 Lamasquère, 20 km south-west of Toulouse, France (43°49'N, 1°23'E). Ozone deposition  
105 was assessed by eddy-covariance using a fast-response O<sub>3</sub> chemiluminescent analyzer  
106 (ATDD, NOAA, USA). Owing to the very small and constant offset of the O<sub>3</sub> analyzer, direct  
107 measurement of  $V_d$  was provided following the ratio method described in Müller et al. (2010).

108 The third dataset concerns measurements carried out over an agricultural field during bare soil  
109 period at La Cape Sud, 60 km south of Bordeaux, France (44°24'N, 0°38'W), from 19  
110 October 2007 to 4 March 2008. As for Lamasquère, O<sub>3</sub> deposition velocity was measured by  
111 eddy-covariance following the ratio method (Müller et al., 2010) by using a fast-response O<sub>3</sub>  
112 chemiluminescent analyzer (ATDD, NOAA, USA).

113 The fourth dataset was collected from 17 March to 5 May 2011 over an agricultural field  
114 during bare soil period before maize sowing at Lusignan site, 30 km south of Poitiers, France  
115 (46°24'N, 0°07'E). Standard meteorological conditions were measured at 1.86 m above  
116 ground level (a.g.l.) including net, incident and reflected shortwave, and incident and reflected  
117 longwave radiations (CNR1, Kipp & Zonnen, NL), air temperature and relative humidity  
118 (HMP45C, Vaisala, FI), wind speed (A100R, Campbell Scientific, USA), wind direction  
119 (W200P, Campbell Scientific, USA), and rainfall (SBS500, Campbell Scientific, USA). Soil  
120 temperatures were measured at 0.05, 0.10, 0.20, 0.30, 0.60, 0.80, and 1m depth using PT100  
121 sensors (Mesurex, FR), as well as soil water content at 0.10, 0.20, 0.30, 0.60, 0.80 and 1m  
122 depth with CS616 probes (Campbell Scientific, USA). Soil heat flux was measured with two  
123 flux plates (HFP01, Hukseflux, NL). All microclimatic data were sampled every 30 s on data  
124 logger (CR1000, Campbell Scientific, USA) and averaged every 30 min. Turbulent fluxes of  
125 momentum, sensible heat, water vapor, CO<sub>2</sub> and O<sub>3</sub> were measured at 1.86 m a.g.l. by eddy-  
126 covariance (EC). The EC system consisted in a 3D sonic anemometer (R3-50, GILL  
127 Instruments, UK) coupled with CO<sub>2</sub>/H<sub>2</sub>O Infrared Gas Analyzer (LI-7500, LICOR, USA), and  
128 a fast-response O<sub>3</sub> chemiluminescent analyzer (ATDD, NOAA, USA). The coumarin dye of  
129 the fast-response O<sub>3</sub> analyzer was changed once per week. Data were sampled and recorded at  
130 20 Hz on a computer using Edisol software (University of Edinburgh, UK), and flux  
131 integration was performed over 30 min time spans. Flux calculation was assessed following  
132 the CarboEurope methodology (Aubinet et al., 2000). In the case of O<sub>3</sub>, the ratio method  
133 providing deposition velocity (Müller et al., 2010) was applied.

134 The last dataset corresponds to measurements performed between 24 March to 14 April 2014  
 135 at Turro (PC) Italy, (44°59'N, 9°42'E) (Scalvenzi, 2015). The site consisted in an agricultural  
 136 field with bare, ploughed and smoothed soil. Standard meteorological conditions were  
 137 measured 2.2 m a.g.l.: net radiation (NR-LITE, Kipp & Zonen, NL), incident solar radiation  
 138 (LI 200 SZ, LI-COR, USA), and air temperature and humidity (HD9000, Deltaohm, I). Soil  
 139 heat fluxes (HFP01, Hukseflux, NL) were measured at 0.05 m depth while soil water content  
 140 (CS616, Campbell Scientific, USA) was measured with a reflectometer averaging the soil  
 141 water content in the first 30 cm of soil below ground. All the data from these probes were  
 142 averaged each half an hour and collected on a data logger (CR10x, Campbell Scientific,  
 143 USA). An additional mast was set up for wind speed, wind direction and eddy covariance flux  
 144 measurements at 2.2 m a.g.l., and included an ultrasonic anemometer (USA-1, Metek, D), a  
 145 krypton hygrometer (KH2O, Campbell Scientific, USA), and a fast-response  
 146 chemiluminescent O<sub>3</sub> analyser (COFA, Ecometrics, I) based on the reaction between ozone  
 147 and coumarin for which the dye was changed typically every 5 days. An additional slow  
 148 response photometric O<sub>3</sub> analyser (1308, SIR, E) was used to calibrate the fast-response O<sub>3</sub>  
 149 analyser. Eddy covariance data were recorded at 10 Hz and collected on a personal computer  
 150 and stored in half an hour files. Flux calculation was assessed following the CarboEurope  
 151 methodology (Aubinet et al., 2000) and included raw data despiking following the procedure  
 152 proposed by Vickers and Mahrt (1997), linearly gap-filling, and a double rotation of the  
 153 reference system of the wind components (Wilczak et al., 2001).

## 154 2.2 – Calculation of ozone soil resistance and surface relative humidity

155 Following the resistance analogy (Wesely and Hicks, 2000),  $V_d$  (in  $\text{m s}^{-1}$ ) to bare soil is  
 156 expressed as:

$$157 \quad V_d = \frac{1}{R_a(z) + R_{bO_3} + R_{soil}} \quad (1)$$

158 where  $R_{soil}$  ( $\text{s m}^{-1}$ ) is the soil resistance, and  $R_a(z)$  and  $R_{bO_3}$  ( $\text{s m}^{-1}$ ) are the aerodynamic and  
 159 the quasi-laminar boundary layer resistances, respectively, calculated following Garland  
 160 (1977). From Eq. (1),  $R_{soil}$  is expressed as:

$$161 \quad R_{soil} = V_d^{-1} - R_a(z) - R_{bO_3} \quad (2)$$

162 According to Stella et al. (2011b),  $R_{soil}$  depends on surface air relative humidity ( $RH_{surf}$  in %)  
 163 at  $z_0$  (soil roughness height for scalar):

$$164 \quad R_{soil} = R_{soil\ min} \times \exp(k \times RH_{surf}) \quad (3)$$

165 where  $R_{\text{soil min}}$  ( $\text{s m}^{-1}$ ) is the soil resistance without water adsorbed at the surface (i.e. at  
 166  $\text{RH}_{\text{surf}} = 0\%$ ) and  $k$  is an empirical coefficient of the exponential function.

167 Following Stella et al. (2011b),  $\text{RH}_{\text{surf}}$  is retrieved from  $H$  and  $\text{LE}$  by using the resistance  
 168 analogy:

$$169 \quad T_{\text{surf}} = \frac{H(R_a(z) + R_b)}{\rho C_p} + T_a \quad (4)$$

$$170 \quad \chi_{\text{H}_2\text{O surf}} = E(R_a(z) + R_{b\text{H}_2\text{O}}) + \chi_{\text{H}_2\text{O a}} \quad (5)$$

$$171 \quad P_{\text{vapsurf}} = \frac{\chi_{\text{H}_2\text{O surf}} R(T_{\text{surf}} + 273.15)}{M_{\text{H}_2\text{O}}} \quad (6)$$

$$172 \quad P_{\text{sat}}(T_{\text{surf}}) = p \exp \left[ \frac{M_{\text{H}_2\text{O}} 10^{-3} L}{R} \left( \frac{1}{T_0 + 273.15} - \frac{1}{T_{\text{surf}} + 273.15} \right) \right] \quad (7)$$

$$173 \quad \text{RH}_{\text{surf}} = \frac{P_{\text{vapsurf}}}{P_{\text{sat}}(T_{\text{surf}})} \times 100 \quad (8)$$

174 with  $T_{\text{surf}}$  the surface temperature ( $^{\circ}\text{C}$ ),  $\rho$  the air density ( $\text{kg m}^{-3}$ ),  $C_p$  the air specific heat  
 175 ( $\text{J kg}^{-1} \text{K}^{-1}$ ),  $\chi_{\text{H}_2\text{O surf}}$  and  $\chi_{\text{H}_2\text{O a}}$  the air concentration of water ( $\text{g m}^{-3}$ ) at  $z_0$  and reference height,  
 176 respectively,  $E$  the water vapor flux ( $\text{kg m}^{-2} \text{s}^{-1}$ ),  $P_{\text{vapsurf}}$  the water vapor pressure at  $z_0$  (Pa),  $R$   
 177 the universal gas constant ( $\text{J mol}^{-1} \text{K}^{-1}$ ),  $M_{\text{H}_2\text{O}}$  the molecular weight of water ( $\text{g mol}^{-1}$ ),  
 178  $P_{\text{sat}}(T_{\text{surf}})$  the saturation vapor pressure at  $T_{\text{surf}}$  (Pa),  $p$  the atmospheric pressure (Pa),  $L$  the  
 179 latent heat of vaporization of water ( $\text{J kg}^{-1}$ ), and  $T_0$  the boiling temperature of water ( $^{\circ}\text{C}$ ).

## 180 3 – RESULTS AND DISCUSSION

### 181 3.1 – Weather, pedoclimate and ozone deposition

182 Meteorological variables followed typical diurnal trends at each sites. Global radiation and  $u_*$   
 183 increased in early morning to reach their maximum at around noon and then decreased to their  
 184 minimum in late afternoon (Figures 1a and 1d). Air temperature and  $\text{RH}_a$  exhibited opposite  
 185 trends: while the former increased in early morning to reach its maximum in early afternoon  
 186 and then decreased (Figure 1b), the latter decreased until early afternoon to its minimum value  
 187 before increasing to its maximum occurring during nighttime (Figure 1c). Soil water content  
 188 did not show marked diurnal trend for the three sites where it was measured (i.e., Lamasquère,  
 189 Lusignan, and Turro) (Figure 1e).

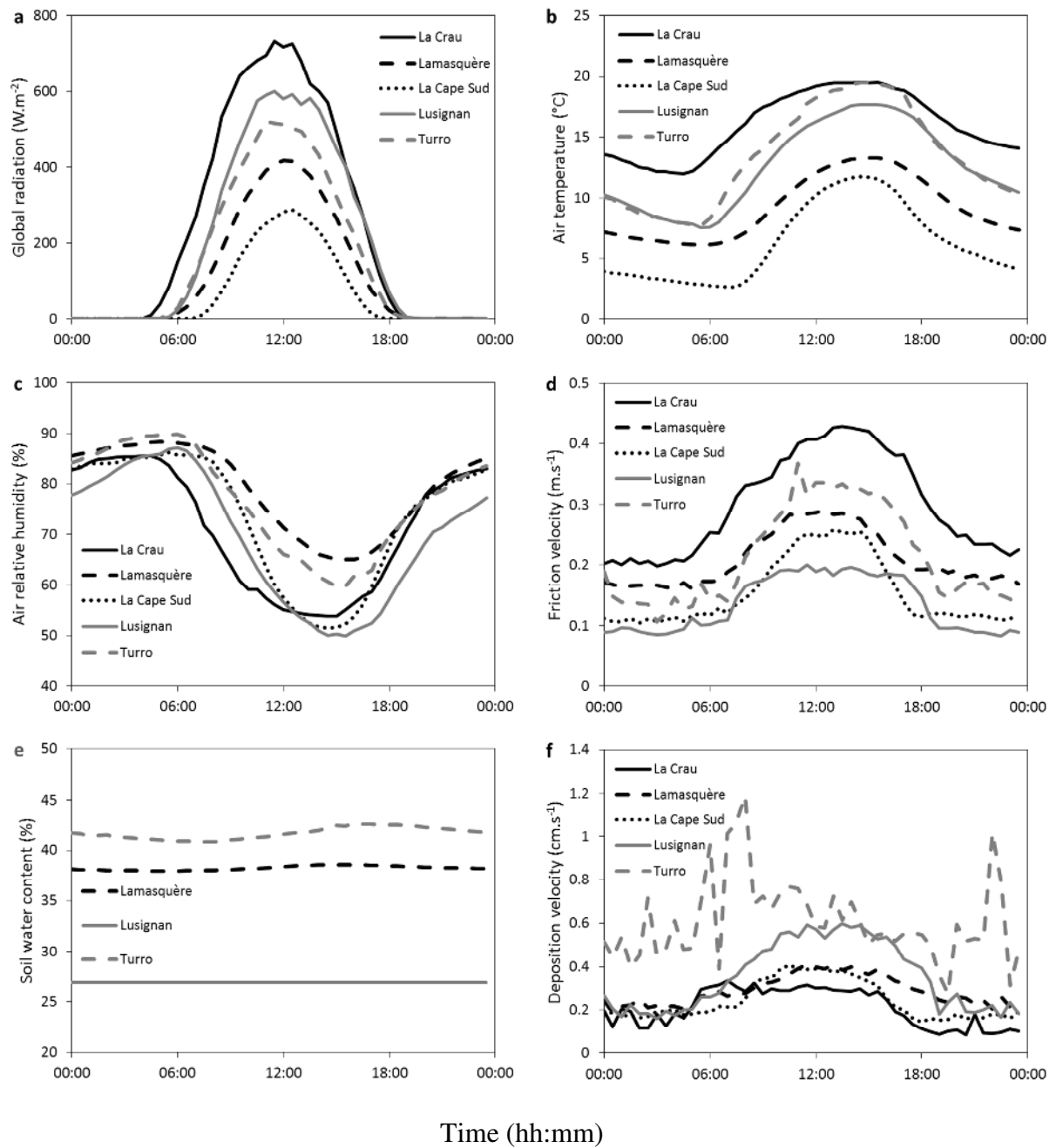
190 Although diurnal trends of meteorological variables were similar, pedoclimatic conditions  
 191 were contrasted at each site. La Crau exhibited the sunniest and warmest meteorological  
 192 conditions with mean  $R_g = 232 \text{ W m}^{-2}$  over the whole experimental period (Table 1), and



193 maximum half hourly  $R_g$  around  $730 \text{ W m}^{-2}$  (Figure 1a). Air temperature and relative  
194 humidity were on average at  $15.8^\circ\text{C}$  and  $72\%$  (Table 1) with maximum and minimum half-  
195 hourly means around  $19^\circ\text{C} / 85\%$  and  $12^\circ\text{C} / 54\%$ , respectively (Figure 1b and 1c). The  
196 highest  $u_*$ , due to windy conditions typical of Mistral in this region, was also recorded on this  
197 site:  $0.30 \text{ m s}^{-1}$  on average (Table 1) and varied between  $0.2 \text{ m s}^{-1}$  during nighttime and  
198  $0.43 \text{ m s}^{-1}$  during daytime (Figure 1d). The soil type was loam (22% clay, 36% silt, 38% sand)  
199 but the soil surface was mainly composed of pebbles. Rainfall and SWC were not recorded on  
200 this site. At Lamasquère,  $R_g$  averaged  $281 \text{ W m}^{-2}$  for the whole dataset (Table 1) and its mean  
201 half hourly value peaked at  $415 \text{ W m}^{-2}$  (Figure 1a). Half hourly means of  $T_a$  and  $\text{RH}_a$  ranged  
202 between  $6^\circ\text{C}$  and  $90\%$  during nighttime and  $13^\circ\text{C}$  and  $65\%$  during daytime (Figure 1b and  
203 1c), and averaged  $9.2^\circ\text{C}$  and  $79\%$  over the whole dataset (Table 1), respectively. Mean  $u_*$  was  
204 at  $0.21 \text{ m s}^{-1}$  (Table 1) over the whole dataset and ranged from  $0.16 \text{ m s}^{-1}$  during nighttime  
205 and  $0.29 \text{ m s}^{-1}$  during daytime (Figure 1d). During the 281 days of the dataset, 833 half hourly  
206 rainfall events were recorded, representing 544 mm cumulated and on average 0.65 mm per  
207 rainfall event. The soil type was clay (54% clay, 34% silt, 12% sand) with a mean SWC of  
208 38% (Table 1). Weather at La Cape Sud was characterized by cloudy and cold conditions:  $R_g$   
209 and  $T_a$  were the lowest of the five datasets, with averages over the whole dataset at  $68 \text{ W m}^{-2}$   
210 and  $6.2^\circ\text{C}$  (Table 1), and half hourly means ranging from 0 to  $273 \text{ W m}^{-2}$  and from  $2.5$  to  
211  $12^\circ\text{C}$  (Figure 1a and 1b), respectively. Dry atmospheric conditions were measured: although  
212  $\text{RH}_a$  averaged 73% (Table 1), it decreased to 50% during daytime and reached 85% during  
213 nighttime (Figure 1c). Friction velocity ranged between 0.1 and  $0.25 \text{ m s}^{-1}$  (Figure 1d), with  
214 mean  $u_*$  of  $0.15 \text{ m s}^{-1}$  (Table 1). Cumulated rainfall during the 137 days of the dataset was at  
215 177 mm, and 365 half hourly events of 0.48 mm on average occurred. The soil type at this site  
216 was loamy sand (6% clay, 9% silt, 77% sand) (Table 1). At Lusignan, half hourly mean  $R_g$   
217 peaked at  $600 \text{ W m}^{-2}$  (Figure 1a) and averaged  $195 \text{ W m}^{-2}$  over the whole dataset (Table 1).  
218 Means of  $T_a$  and  $\text{RH}_a$  were at  $12.6^\circ\text{C}$  and 69% (Table 1) and their half hourly means ranged  
219 from  $7.5^\circ\text{C}$  to  $17.7^\circ\text{C}$  (Figure 1b) and from 87% to 50% (Figure 1c), respectively. This  
220 dataset exhibited the weakest  $u_*$  and SWC: they were on average at  $0.13 \text{ m s}^{-1}$  and 27%,  
221 respectively (Table 1). Half hourly means of  $u_*$  only varied from  $0.09 \text{ m s}^{-1}$  during nighttime  
222 to  $0.20 \text{ m s}^{-1}$  during daytime (Figure 1d), while half hourly means SWC did not show diurnal  
223 variations (Figure 1e). Cumulated rainfall during the 49 days of the dataset was at 36 mm, and  
224 63 half hourly events of 0.59 mm on average occurred. The soil type at this site was silt loam  
225 (17% clay, 60% silt, 23% sand) (Table 1). The last site, Turro, presented intermediate  $R_g$ : it  
226 was on average over the whole dataset  $155 \text{ W m}^{-2}$  (Table 1) and its half hourly mean peaked

227 to  $516 \text{ W m}^{-2}$  (Figure 1a). Half hourly means of  $T_a$  were similar to those recorded at Lusignan  
228 during nighttime and La Crau during daytime, i.e., ranged from  $7.5^\circ\text{C}$  to  $19^\circ\text{C}$  (Figure 1b).  
229 Over the whole dataset,  $T_a$  was on average at  $13.3^\circ\text{C}$  (Table 1). For  $\text{RH}_a$ , half hourly means  
230 varied between 90% and 60% and was 77% over the whole dataset (Figure 1c and Table 1).  
231 The friction velocity exhibited quite large diurnal variation: its half hourly means ranged from  
232  $0.1 \text{ m s}^{-1}$  during nighttime to  $0.37 \text{ m s}^{-1}$  during daytime (Figure 1d) while it was on average at  
233  $0.21 \text{ m s}^{-1}$  during the whole measurement period (Table 1). This site exhibited the largest  
234 SWC, 42% (Table 1). During the 21 days of the dataset, 55 half hourly rainfall events were  
235 recorded, representing 35.6 mm cumulated and on average 0.65 mm per rainfall event. The  
236 soil type was silty clay loam (30% clay, 52% silt, 18% sand) (Table 1).

237 Half hourly means of  $V_d$  are presented in Figure 1f. Excepted at Turro,  $V_d$  measured during  
238 nighttime was similar at each site, between  $0.15\text{-}0.25 \text{ cm s}^{-1}$ . They then increased during early  
239 morning to reach their maximum, around  $0.30 \text{ cm s}^{-1}$  at La Crau,  $0.40 \text{ cm s}^{-1}$  at Lamasquère  
240 and La Cape Sud, and  $0.60 \text{ cm s}^{-1}$  at Lusignan. At Turro,  $V_d$  did not follow typical diurnal  
241 dynamics and exhibited important half-hourly variations, with half hourly  $V_d$  oscillating  
242 around  $0.5\text{-}0.6 \text{ cm s}^{-1}$  during both nighttime and daytime, and exhibiting two peaks (at around  
243  $1\text{-}1.2 \text{ cm s}^{-1}$ ) during early morning and late evening. As a consequence, with  
244  $0.61 \text{ cm s}^{-1}$ ,  $V_d$  was on average over the whole dataset the largest at this site. For the other  
245 sites, mean  $V_d$  was  $0.21 \text{ cm s}^{-1}$  for La Crau,  $0.29 \text{ cm s}^{-1}$  for Lamasquère,  $0.26 \text{ cm s}^{-1}$  for La  
246 Cape Sud, and  $0.36 \text{ cm s}^{-1}$  for Lusignan (Table 1).



247

248

249 **Figure 1:** Half hourly arithmetic means of (a) global radiation, (b) air temperature, (c) air  
 250 relative humidity, (d) friction velocity, (e) soil water content, and (f) deposition velocity for  
 251 La Crau (black line), Lamasquère (dashed black line), La Cape Sud (dotted black line),  
 252 Lusignan (grey line), and Turro (dashed grey line) sites.

253

254 **Table 1:** Arithmetic means ( $\pm$  standard deviations) of global radiation ( $R_g$ ), air temperature  
 255 ( $T_a$ ), air relative humidity ( $RH_a$ ), friction velocity ( $u_*$ ), rainfall, soil water content (SWC), and  
 256 deposition velocity ( $V_d$ ) during the measurement periods for each site. Are also indicated the  
 257 dataset duration, cumulated rainfall, number of half hourly rainfall events (n), and soil texture.

	Dataset duration	$R_g$	$T_a$	$RH_a$	$u_*$	Rainfall		SWC	Soil texture			$V_d$
						Sum	n		%Clay	%Silt	%Sand	
	Days	W m <sup>-2</sup>	°C	%	m s <sup>-1</sup>	mm		%			cm s <sup>-1</sup>	
		Mean $\pm$ SD	Mean $\pm$ SD	Mean $\pm$ SD	Mean $\pm$ SD	Mean $\pm$ SD	Sum	n	Mean $\pm$ SD		Mean $\pm$ SD	
<b>La Crau</b>	41	232 $\pm$ 298	15.8 $\pm$ 4.5	72 $\pm$ 19	0.30 $\pm$ 0.20	-	-	-	-	22* 36* 38*	0.21 $\pm$ 0.21	
<b>Lamasquère</b>	281	118 $\pm$ 207	9.2 $\pm$ 6.5	79 $\pm$ 15	0.21 $\pm$ 0.16	0.65 $\pm$ 0.86	544	833	38 $\pm$ 4	54 34 12	0.29 $\pm$ 0.33	
<b>La Cape Sud</b>	137	68 $\pm$ 118	6.2 $\pm$ 5.3	73 $\pm$ 19	0.15 $\pm$ 0.12	0.48 $\pm$ 0.49	177	365	-	6* 9* 77*	0.26 $\pm$ 0.20	
<b>Lusignan</b>	49	195 $\pm$ 261	12.6 $\pm$ 4.9	69 $\pm$ 20	0.13 $\pm$ 0.09	0.59 $\pm$ 0.74	36	63	27 $\pm$ 1	17 60 23	0.36 $\pm$ 0.28	
<b>Turro</b>	21	155 $\pm$ 209	13.3 $\pm$ 4.5	77 $\pm$ 13	0.21 $\pm$ 0.11	0.65 $\pm$ 0.83	35.6	55	42 $\pm$ 3	30 52 18	0.61 $\pm$ 0.48	

258 \*: Not measured on site, obtained from Geosol Database

259 (<http://estrada.oreans.inra.fr/geosol/>).

### 260 3.2 – Relationships between $R_{soil}$ and $RH_{surf}$

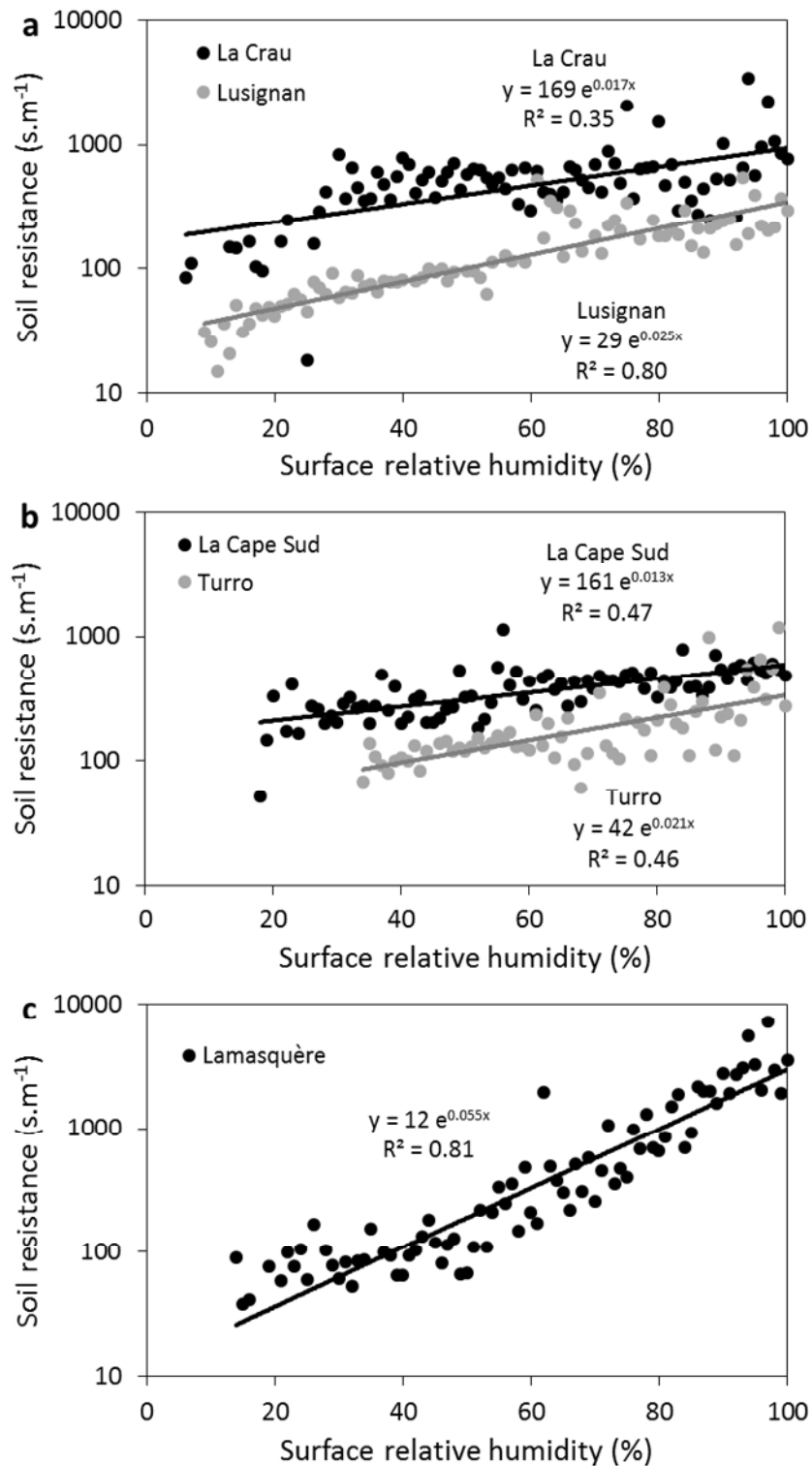
261 Several authors related  $R_{soil}$  to the amount of water in soil, and showed that the drier the soil  
 262 is, the weaker  $R_{soil}$  is (e.g., Bassin et al., 2004; Massman, 2004; Meszaros et al., 2009). More  
 263 recently, Stella et al. (2011b) showed that (i) the estimate of  $R_{soil}$  as a function of soil water  
 264 content does not provide an accurate estimate of  $V_d$  over bare soil, and (ii)  $R_{soil}$  depends on  
 265 and increases exponentially with  $RH_{surf}$ . Therefore, the relationships between  $R_{soil}$  and  $RH_{surf}$   
 266 were determined for each site and are presented in Figure 2. From these relationships, the two  
 267 parameters of the exponential function controlling  $R_{soil}$  i.e.,  $R_{soil\ min}$  and  $k$  (Equation 3), were

268 estimated from the exponential regression between  $R_{\text{soil}}$  and  $\text{RH}_{\text{surf}}$  (Figure 2), and are  
269 summarized in Table 2.

270 Whatever the site considered,  $R_{\text{soil}}$  increases exponentially with  $\text{RH}_{\text{surf}}$ . These results confirm  
271 those previously obtained by Stella et al. (2011b) at an agricultural field during bare soil  
272 periods. Nevertheless, this relationship substantially differs quantitatively according to the site  
273 considered, as shown in Figure 2 and by the values obtained for  $R_{\text{soil min}}$  and  $k$  (Table 2).

274 The minimum resistance i.e.,  $R_{\text{soil}}$  at  $\text{RH}_{\text{surf}} = 0\%$  corresponding to  $R_{\text{soil min}}$ , strongly varied  
275 according to the site considered (Figure 2 and Table 2). The lowest resistance was observed at  
276 Lamasquère ( $12 \pm 1.20 \text{ s m}^{-1}$ ) and the largest at La Crau ( $169 \pm 1.18 \text{ s m}^{-1}$ ). For Lusignan,  
277 Turro, and La Cape Sud,  $R_{\text{soil min}}$  was  $29 \pm 1.01 \text{ s m}^{-1}$ ,  $42 \pm 1.22 \text{ s m}^{-1}$ , and  $161 \pm 1.11 \text{ s m}^{-1}$ ,  
278 respectively. As far we know, only two studies reported values of  $R_{\text{soil min}}$ . The first concerns  
279 the study of Stella et al. (2011b) for an agricultural field during bare soil periods who found  
280  $R_{\text{soil min}} = 21 \pm 1.01 \text{ s m}^{-1}$ . The second originated from Güsten et al. (1996) who reported from  
281 an experiment in Sahara desert an average daytime resistance of ozone to desert sand of  $800$   
282  $\text{s m}^{-1}$ . In such conditions, it can be hypothesized that  $\text{RH}_{\text{surf}}$  is very close or equal to  $0\%$ , and  
283 hence the value reported by Güsten et al. (1996) corresponds to  $R_{\text{soil min}}$ . Therefore, in spite of  
284 their large range of variation, the values found for our five sites remain in the range reported  
285 by previous studies.

286 Similarly to  $R_{\text{soil min}}$ , the increase of  $R_{\text{soil}}$  with  $\text{RH}_{\text{surf}}$ , corresponding to  $k$ , exhibited large site-  
287 by-site variations (Figure 2 and Table 2). The slowest increase was found at La Crau and La  
288 Cape Sud, with  $k$  equal to  $0.017 \pm 0.003$  and  $0.013 \pm 0.002$ , respectively, while Lamasquère  
289 exhibited the fastest increase ( $k = 0.055 \pm 0.003$ ). The values found at Lusignan and Turro  
290 were intermediate,  $0.025 \pm 0.001$  and  $0.021 \pm 0.003$ , respectively, but are similar to the one  
291 reported by Stella et al. (2011b) who reported  $k = 0.024 \pm 0.001$ .



292

293 **Figure 2:** Soil resistance as a function of surface relative humidity for (a) La Crau (black  
 294 symbols) and Lusignan (grey symbols), (b) La Cape Sud (black symbols) and Turro (grey  
 295 symbols), and (c) Lamasquère. Data are block averaged with a range of 1% surface relative  
 296 humidity. Lines represent the regression (general form:  $R_{soil} = R_{soil\ min} \times \exp(k \times RH_{surf})$ ).  
 297 Only data for  $u_* > 0.1\ \text{m s}^{-1}$  were used.

298 **Table 2:** Minimum soil resistances ( $R_{\text{soil min}}$ ), empirical coefficients of the exponential  
 299 functions ( $k$ ) and clay contents obtained for the sites of this study. The values and their  
 300 standard errors were obtained by the regressions between  $R_{\text{soil}}$  and  $\text{RH}_{\text{surf}}$ . Are also indicated  
 301 the values reported by Stella et al. (2011b) and Gusten et al. (1996) (see text for details).

	$R_{\text{soil min}}$	$k$	Clay content
	s.m <sup>-1</sup>	-	%
	Value ± SE	Value ± SE	
<b>Lamasquère</b>	12 ± 1.20	0.055 ± 0.003	54
<b>Stella et al. (2011b)<sup>1</sup></b>	21 ± 1.01	0.024 ± 0.001	31
<b>Lusignan</b>	29 ± 1.01	0.025 ± 0.001	17
<b>Turro</b>	42 ± 1.22	0.021 ± 0.003	30
<b>La Cape Sud</b>	161 ± 1.11	0.013 ± 0.002	6
<b>La Crau</b>	169 ± 1.18	0.017 ± 0.003	22
<b>Güsten et al. (1996)<sup>2</sup></b>	800	[-]	0.8

302 <sup>1</sup> Soil texture: 31% Clay, 62.5% Silt, 6.5% Sand

303 <sup>2</sup> Soil texture: 0.8% Clay, 1.3% Silt, 97.9% Sand

### 304 3.3 – Dependence of $R_{\text{soil}}$ parameters to soil texture

305 As shown previously, the dependence of  $R_{\text{soil}}$  to  $\text{RH}_{\text{surf}}$  exhibits large site-by-site variations, as  
 306 suggested by Stella et al. (2011a, 2011b). Yet, Stella et al. (2011b) hypothesized that the  
 307 increase of  $R_{\text{soil}}$  with  $\text{RH}_{\text{surf}}$  is due to the presence of water adsorbed at the soil surface,  
 308 decreasing the surface active for  $\text{O}_3$  deposition since  $\text{O}_3$  is hardly soluble in water i.e.,  $R_{\text{soil}}$   
 309 depends on the available dry soil surface.

310 In our study, the main difference that could explain this variability between each site concerns  
 311 the soil texture, which would be consistent with the hypothesis proposed by Stella et al.  
 312 (2011). Indeed, the soil texture and more specifically the clay content determines the specific  
 313 surface area i.e., the mass normalized surface area (in m<sup>2</sup> g<sup>-1</sup>): at a microscopic scale the  
 314 surface for the same amount of soil increases with clay content due to the size and structure of  
 315 these elements. This issue has been proved both theoretically and experimentally in e.g.,  
 316 Petersen et al. (1996) and Pennel (2002). In other words, the greater the amount of clay is, the

317 larger the surface available at a microscopic scale for O<sub>3</sub> deposition is. To examine this issue,  
318 the two parameters  $R_{\text{soil min}}$  and  $k$  were plotted as a function of soil clay content.

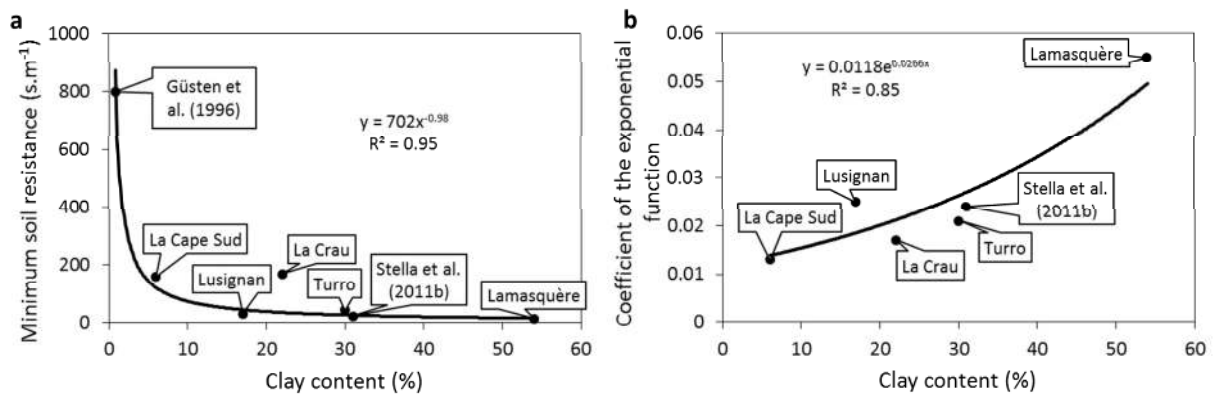
319 The results are presented in Figure 3 and include those obtained from the sites of this study as  
320 well as those from Güsten et al. (1996) and Stella et al. (2011b). The results from La Crau  
321 were not included in fitting the relationships between  $R_{\text{soil min}}$  and  $k$  and the soil clay content,  
322 as discussed at the end of this section. On the one hand,  $R_{\text{soil min}}$  decreases when soil clay  
323 content increases. This decrease is particularly marked for soil clay content lower than 10-  
324 15%. Above this percentage,  $R_{\text{soil min}}$  decreases less rapidly. The best correlation coefficient  
325 was obtained for a power regression ( $R_{\text{soil min}} = 702 \times (\text{clay content})^{-0.98}$ ;  $R^2 = 0.95$ )  
326 (Figure 3a). On the other hand,  $k$  increases with soil clay content and the best correlation  
327 coefficient was found for an exponential relationship  
328 ( $k = 0.0118 \times \exp^{-0.0266 \times (\text{clay content})}$ ;  $R^2 = 0.85$ ) (Figure 3b).

329 As expected from our working hypothesis, when there is no water adsorbed at the soil surface  
330 the soil resistance (i.e.,  $R_{\text{soil min}}$ ) decreases with the increase in soil clay content. In other  
331 words, O<sub>3</sub> deposition to soil is favored when the soil specific surface increases, since more  
332 surface is available for O<sub>3</sub> removal at a microscopic scale. As far we know, only the study of  
333 Sorimachi and Sakamoto (2007) has already examined in this issue with similar results. From  
334 laboratory measurements of O<sub>3</sub> deposition onto different soil samples, they reported that for  
335 soil moisture content lower than 10% (i.e., close to  $\text{RH}_{\text{surf}}$ ) the surface resistance decreases  
336 exponentially with increasing soil surface area. Nevertheless, it must be noted that from our  
337 in-situ measurements it was found a power decrease instead of an exponential one.  
338 Concerning the increase of  $k$  with soil clay content, a possible explanation would be link to  
339 the capacity of a soil to adsorb/desorb water, and therefore to contain water, according to its  
340 amount of clay. According to Schneider and Goss (2012), for the same relative humidity, the  
341 water content is larger for soil with a large amount of clay than for soil with low amount of  
342 clay. In addition, its increase with relative humidity is faster when the amount of clay is large.  
343 Since O<sub>3</sub> is hardly soluble in water, this statement could explain the faster increase of  $R_{\text{soil}}$   
344 with  $\text{RH}_{\text{surf}}$  (i.e. larger  $k$ ) for soils with larger clay content.

345 Results obtained at La Crau site exhibited discrepancies compared to the results obtained on  
346 the other sites, especially concerning  $R_{\text{soil min}}$  (Figure 3a). For this latter, there are only two  
347 possibilities: (i)  $R_{\text{soil min}}$  is underestimated or (ii) the soil clay content is overestimated. The  
348 former seems not to be plausible since the methodology to deduce  $R_{\text{soil min}}$  is identical for all  
349 the sites. However, it is possible that the percentage of clay did not reflect reality. Since O<sub>3</sub> is



350 an highly reactive compound, it can be rapidly depleted. Therefore, properties retained must  
 351 be representative of the soil surface, i.e., the first few millimeters. At this site, soil surface was  
 352 mainly composed of pebbles for which specific surface is small. Thus the “real clay content”  
 353 of the soil surface at this site, in regards with soil specific surface area, would be probably  
 354 closer to a sandy soil as for La Cape Sud, for which both  $R_{\text{soil min}}$  and  $k$  are similar with the La  
 355 Crau ones (Table 2).



356  
 357 **Figure 3:** Relationships between (a) minimum soil resistance ( $R_{\text{soil min}}$ ) and (b) coefficient of  
 358 the exponential function ( $k$ ) and soil clay content. Black lines correspond to the regressions.  
 359 Values from La Crau site were not included in the regressions.

#### 360 4 – CONCLUSIONS AND PERSPECTIVES

361 This study explored the variability of soil  $O_3$  resistance according to soil texture. To this end,  
 362  $O_3$  deposition data over bare soil obtained from micrometeorological measurements under  
 363 contrasted meteorological conditions for five sites were used. The results obtained are  
 364 twofold: (i)  $R_{\text{soil}}$  increases with  $RH_{\text{surf}}$  as found previously by Stella et al. (2011b), but (ii) the  
 365 relationships exhibited large site-to-site variability. From the data analysis, the minimum soil  
 366 resistance without water adsorbed at the surface (i.e., at  $RH_{\text{surf}} = 0\%$ ) corresponding to  $R_{\text{soil}}$   
 367  $_{\text{min}}$ , and the increase of  $R_{\text{soil}}$  with  $RH_{\text{surf}}$  corresponding to  $k$  are linked to soil clay content.  
 368 These patterns can be explained respectively by (i) the surface available for  $O_3$  deposition at a  
 369 microscopic scale which is linked to the soil specific surface area, and (ii) the capacity of a  
 370 soil to adsorb water according to its clay content and therefore to reduce the surface active for  
 371  $O_3$  deposition. From our results (Figure 3) a new parameterization can be established to  
 372 estimate  $R_{\text{soil}}$  as a function of  $RH_{\text{surf}}$  (%) and soil clay content (%):

$$373 R_{\text{soil}} = 702 \times (\text{clay content})^{-0.98} \times \exp^{(0.0118 \times \exp^{-0.0266 \times (\text{clay content})} \times RH_{\text{surf}})} \quad (9)$$

374 This empirical parameterization could be included into surface-atmosphere exchanges models  
375 to assess the O<sub>3</sub> dry deposition budget of continental surface since (i) the soil component can  
376 represent an important fraction of total deposition especially for agroecosystems (Stella et al.  
377 2013) and (ii) current parameterizations accounting for soil water content overestimate  $R_{\text{soil}}$ ,  
378 especially under dry conditions (Stella et al., 2011b). For instance, assuming a soil water  
379 content equal to 0%, the parameterization used in Bassin et al. (2004) and Meszaros et al.  
380 (2009) gives  $R_{\text{soil}} = 200 \text{ s.m}^{-1}$ , values that are particularly high compared to the smallest ones  
381 deduced at Lusignan, Lamasquère and Turro (Figure 2).

382 It must however be noted that our study is limited to only few sites. Ozone deposition is rarely  
383 measured over bare soil, and further efforts should be done to complete our study and assess  
384 the relationships proposed. Yet, we limited our study to the soil texture which is the main  
385 factor controlling the soil specific surface area, but other parameters also modify it, such as  
386 soil compaction. In addition, we only focused our work on the hypothesis of a physical  
387 underlying process i.e., the surface available for O<sub>3</sub> deposition at a microscopic scale.  
388 Additional chemical processes that remove O<sub>3</sub> at the interface soil-atmosphere remain  
389 possible. For instance, the study performed by Vuolo et al. (2017) indicated that soil O<sub>3</sub>  
390 deposition increased following slurry application, suggesting a chemical process linked with  
391 surface reactivity changes due to the added organic matter or volatile organic compound  
392 (VOC) emissions from the slurry. These chemical processes are primarily controlled by  
393 temperature, in a purely reactive way (e.g., Cape et al., 2009). Finally, even if O<sub>3</sub> is weakly  
394 soluble in water, possible dissolution, diffusion and chemical reaction inside the water films at  
395 the soil surface cannot be discarded as suggested by Potier et al. (2015) for O<sub>3</sub> cuticular  
396 deposition on wet leaves, implying an impact of e.g., compound concentrations in water films  
397 (Potier et al., 2015) or water pH (e.g., Flechard et al., 1999). Understanding these effects  
398 could be of particular importance, especially for agroecosystems for which agricultural  
399 practices such as ploughing, crushing or organic fertilization can change both soil  
400 compaction, organic matter content, and soil surface reactivity.

401

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414

**415 References**

416 Ahmad Z., Allam I.M., Abdul Aleem B.J., 2000. Effect of environmental factors on the  
417 atmospheric corrosion of mild steel in aggressive coastal environment. *Anti-Corrosion*  
418 *Methods and Materials* 47, 215–225.

419 Ainsworth E.A., Yendrek C.R., Sitch S., Collins W.J., Emberson L.D., 2012. Ozone on net  
420 primary productivity and implications for climate change. *Annual Review of Plant*  
421 *Biology* 63, 637-661.

422 Altimir N., Kolari P., Tuovinen J.P., Vesala T., Bäck J., Suni T., Kulmala M., Hari P., 2006.  
423 Foliage surface ozone deposition: a role for surface moisture? *Biogeosciences* 3, 209-  
424 228.

425 Altimir N., Tuovinen J.P., Vesala T., Kulmala M., Hari P., 2004. Measurements of ozone  
426 removal by Scots pine shoots: calibration of a stomatal uptake model including the  
427 non-stomatal component. *Atmospheric Environment* 38, 2387-2398.

428 Aubinet M., Grelle A., Ibrom A., Rannik U., Moncrieff J., Foken T., Kowalski A.S., Martin  
429 P.H., Berbigier P., Bernhofer C., Clement R., Elbers J., Granier A., Grunwald T.,  
430 Morgenstern K., Pilegaard K., Rebmann C., Snijders W., Valentini R., Vesala T.,  
431 2000. Estimates of the annual net carbon and water exchange of forests: the  
432 EUROFLUX methodology. *Advances in Ecological Research* 30, 113–175.

433 Bassin S., Calanca P., Weidinger T., Gerosa G., Fuhrer J., 2004. Modeling seasonal ozone  
434 fluxes to grassland and wheat: model improvement, testing, and application.  
435 *Atmospheric Environment* 38, 2349-2359.

436 Béziat P., Ceschia E., Dedieu G., 2009. Carbon balance of a three crop succession over two  
437 cropland sites in South West France. *Agricultural and Forest Meteorology* 149, 1628-  
438 1645.

- 439 Booker F., Muntifering R., McGrath M., Burkey K., Decoteau D., Fiscus E., Manning W.,  
440 Krupa S., Chappelka A., Grantz D., 2009. The ozone component of global change:  
441 Potential effects on agricultural and horticultural plant yield, product quality and  
442 interactions with invasive species. *Journal of Integrative Plant Biology* 51, 337-351.
- 443 Cape J.N., Hamilton R., Heal M.R., 2009. Reactive uptake of ozone at simulated leaf  
444 surfaces: Implications for 'non-stomatal' ozone flux. *Atmospheric Environment* 43,  
445 1116-1123.
- 446 Clifton O.E., Fiore A.M., Munger J.W., Malyshev S., Horowitz L.W., Shevliakova E., Paulot  
447 F., Murray L.T., Griffin K.L., 2017. Interannual variability in ozone removal by a  
448 temperate deciduous forest. *Geophysical Research Letters* 44, 542-552.
- 449 Coyle M., Nemitz E., Storeton-West R., Fowler D., Cape J.N., 2009. Measurements of ozone  
450 deposition to a potato canopy. *Agricultural and Forest Meteorology* 149, 655-666.
- 451 Dizengremel P., Le Thiec D., Bagard M., Jolivet Y., 2008. Ozone risk assessment for plants:  
452 Central role of metabolism-dependent changes in reducing power. *Environmental*  
453 *Pollution* 156, 11-15.
- 454 Doherty R.M., Heal M.R., O'Connor F.M., 2017. Climate change impacts on human health  
455 over Europe through its effect on air quality. *Environmental Health* 16, 33-44.
- 456 Emberson L.D., Ashmore M.R., Cambridge H.M., Simpson D., Tuovinen J.P., 2000.  
457 Modelling stomatal ozone flux across Europe. *Environmental Pollution* 109, 403-413.
- 458 Fares S., Weber R., Park J.H., Genter D., Karlik J., Goldstein A.H., 2012. Ozone deposition to  
459 an orange orchard: Partitioning between stomatal and non-stomatal sinks.  
460 *Environmental Pollution* 169, 258-266.
- 461 Felzer B.S., Cronin T., Reilly J.M., Melillo J.M., Wang X., 2007. Impacts of ozone on trees  
462 and crops. *Comptes Rendus Geoscience* 339, 784-798.
- 463 Flechard C.R., Fowler D., Sutton M.A., Cape J.N., 1999. A dynamic chemical model of bi-  
464 directional ammonia exchange between semi-natural vegetation and the atmosphere.  
465 *Quarterly Journal of the Royal Meteorological Society* 125, 2611-2641.
- 466 Fowler D., Pilegaard K., Sutton M.A., Ambus P., Raivonen M., Duyzer J., Simpson D.,  
467 Fagerli H., Fuzzi S., Schjoerring J.K., Granier C., Neftel A., Isaksen I.S.A., Laj P.,  
468 Maione M., Monks P.S., Burkhardt J., Daemmgen U., Neiryneck J., Personne E.,  
469 Wichink-Kruit R., Butterbach-Bahl K., Flechard C., Tuovinen J.P., Coyle M., Gerosa  
470 G., Loubet B., Altimir N., Gruenhage L., Ammann C., Cieslik S., Paoletti E.,  
471 Mikkelsen T.N., Ro-Poulsen H., Cellier P., Cape J.N., Horvath L., Loreto F.,  
472 Niinemets U., Palmer P.I., Rinne J., Misztal P., Nemitz E., Nilsson D., Pryor S.,  
473 Gallagher M.W., Vesala T., Skiba U., Brüggemann N., Zechmeister-Boltenstern S.,  
474 Williams J., O'Dowd C., Facchini M.C., de Leeuw G., Flossman A., Chaumerliac N.,  
475 Erisman J.W., 2009. Atmospheric composition change: Ecosystem-Atmosphere  
476 interactions. *Atmospheric Environment* 43, 5193-5267.
- 477 Franz M., Simpson D., Arneth A., Zaehle S., 2017. Development and evaluation of an ozone  
478 deposition scheme for coupling to a terrestrial biosphere model. *Biogeosciences* 14,  
479 45-71.
- 480 Freire L.S., Gerken T., Ruiz-Plancarte J., Wei D., Fuentes J.D., Katul G.G., Dias N.L.,  
481 Acevedo O.C., Chamecki M., 2017. Turbulent mixing and removal of ozone within an  
482 Amazon rainforest canopy. *Journal of Geophysical Research: Atmospheres* 122, 2791-  
483 2811.

- 484 Garland J.A., 1977. The dry deposition of sulphur dioxide to land and water surface.  
485 Proceedings of the Royal Society of London A 354, 245–268.
- 486 Güsten H., 1992. A novel ozone sensor for various environmental applications. In: Hudson,  
487 R.D. (Ed.), Ozone in the troposphere and stratosphere, Part 1, NASA Conference  
488 Publication, vol. 3266. Goddard Space Flight Center, Greenbelt, MD, USA, pp. 127–  
489 129.
- 490 Güsten H., Heinrich G., Mönnich E., Sprung D., Weppner J., Bakr Ramadan A., Ezz El-Din  
491 M.R.M., Ahmed D.M., Hassan G.K.Y., 1996. On-line measurements of ozone surface  
492 fluxes: Part II. Surface-level ozone fluxes onto the Sahara desert. Atmospheric  
493 Environment 30, 911-918.
- 494 Hazucha M.J., Lefohn A.S., 2007. Nonlinearity in human health response to ozone:  
495 Experimental laboratory considerations. Atmospheric Environment 41, 4559-4570.
- 496 IPCC, 2013. Climate Change 2013: The Physical Science Basis. Contribution of Working  
497 Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate  
498 Change [Stocker, T.F., D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J. Boschung, A.  
499 Nauels, Y. Xia, V. Bex and P.M. Midgley (eds.)]. Cambridge University Press,  
500 Cambridge, United Kingdom and New York, NY, USA, 1535 pp.
- 501 Ito K., De Leon S.F., Lippmann M., 2005. Associations between ozone and daily mortality:  
502 Analysis and meta-analysis. Epidemiology 16, 446-457.
- 503 Karnosky D.F., Zak D.R., Pregitzer K.S., Awmack C.S., Bockheim J.G., Dickson R.E.,  
504 Hendrey G.R., Host G.E., King J.S., Kopper B.J., Kruger E.L., Kubiske M.E.,  
505 Lindroth R.L., Mattson W.J., McDonald E.P., Noormets A., Oksanen E., Parsons  
506 W.F.J., Percy K.E., Podila G.K., Riemenschneider D.E., Sharma P., Thakur R., Sôber  
507 A., Sôber J., Jones W.S., Anttonen S., Vapaavuori E., Manlovska B., Heilman W.,  
508 Isebrands J.G., 2003. Tropospheric O<sub>3</sub> moderates responses of temperate hardwood  
509 forests to elevated CO<sub>2</sub>: a synthesis of molecular to ecosystem results from the  
510 ASPEN face project. Functional Ecology 17, 289-304.
- 511 Lamaud E., Loubet B., Irvine M., Stella P., Personne E., Cellier P., 2009. Partitioning of  
512 ozone deposition over a developed maize crop between stomatal and non-stomatal  
513 uptakes, using eddy-covariance flux measurements and modelling. Agricultural and  
514 Forest Meteorology 149, 1385-1396.
- 515 Launiainen S., Katul G.G., Grönholm T., Vesala T., 2013. Partitioning ozone fluxes between  
516 canopy and forest floor by measurements and a multi-layer model. Agricultural and  
517 Forest Meteorology 173, 85-99.
- 518 Lee D.S., Holland M.R., Falla N., 1996. The potential impact of ozone on materials in the  
519 U.K. Atmospheric Environment 30, 1053-1065.
- 520 Lombardozzi D., Levis S., Bonan G., Hess P.G., Sparks J.P., 2015. The influence of chronic  
521 ozone exposure on global carbon and water cycles. Journal of Climate 28, 292-305.
- 522 Lombardozzi D., Sparks J.P., Bonan G., 2013. Integrating O<sub>3</sub> influences on terrestrial  
523 processes: photosynthetic and stomatal response data available for regional and global  
524 modeling. Biogeosciences 10, 6815-6831.
- 525 Massman W.J., 2004. Toward on ozone standard to protect vegetation based on effective  
526 dose: a review of deposition resistances and a possible metric. Atmospheric  
527 Environment 38, 2323-2337.

- 528 Meszaros R., Horvath L., Weidinger T., Neftel A., Nemitz E., Dämmgen U., Cellier P.,  
529 Loubet B., 2009. Measurement and modelling ozone fluxes over a cut and fertilized  
530 grassland. *Biogeosciences* 6, 1987-1999.
- 531 Michou M., Laville P., Serça D., Fotiadi A., Bouchou P., Peuch V.H., 2005. Measured and  
532 modeled dry deposition velocities over the ESCOMPTE area. *Atmospheric Research*  
533 74, 89-116.
- 534 Monks P.S., 2005. Gas-phase radical chemistry in the troposphere. *Chemical Society Reviews*  
535 34, 376-395.
- 536 Muller J.B.A., Percival C.J., Gallagher M.W., Fowler D., Coyle M., Nemitz E., 2010. Sources  
537 of uncertainty in eddy covariance ozone flux measurements made by dry  
538 chemiluminescence fast response analysers. *Atmospheric Measurement Techniques* 3,  
539 163–176.
- 540 Paoletti E., 2005. Ozone slows stomatal response to light and leaf wounding in a  
541 Mediterranean evergreen broadleaf, *Arbutus unedo*. *Environmental Pollution* 134,  
542 439-445.
- 543 Pennel K.D., 2002. Specific surface area. p. 308-313. In J.H. Dane and G.C. Topp (ed.)  
544 *Methods of soil analysis. Part 4. SSSA Book Ser. 5. SSSA, Madison, WI.*
- 545 Petersen L.W., Moldrup P., Jacobsen O.H., Rolston D.E., 1996. Relations between specific  
546 surface area and soil physical and chemical properties. *Soil Science* 161, 9-21.
- 547 Potier E., Ogée J., Jouanguy J., Lamaud E., Stella P., Personne E., Durand B., Mascher N.,  
548 Loubet B., 2015. Multilayer modelling of ozone fluxes on winter wheat reveals large  
549 deposition on wet senescing leaves. *Agricultural and Forest Meteorology* 211, 58-71.
- 550 Scalvenzi M., 2015. Resistenza del suolo nudo alla deposizione di ozono. Thesis of the  
551 Master Degree in Physics, Università Cattolica del Sacro Cuore, Brescia, Italia,  
552 Matricola N. 4109094, 44pp.
- 553 Schneider M., Goss K.U., 2012. Prediction of the water sorption isotherm in air dry soils.  
554 *Geoderma*, 170, 64-69.
- 555 Sitch S., Cox P.M., Collins W.J., Huntingford C., 2007. Indirect radiative forcing of climate  
556 change through ozone effects on the land-carbon sink. *Nature* 448, 791-795.
- 557 Stella P., Lamaud E., Brunet Y., Bonnefond J.M., Loustau, D., Irvine M., 2009. Simultaneous  
558 measurements of CO<sub>2</sub> and water exchanges over three agroecosystems in South-West  
559 France. *Biogeosciences* 6, 2957–2971.
- 560 Stella P., Loubet B., Lamaud E., Laville P., Cellier P., 2011b. Ozone deposition onto bare  
561 soil: A new parameterisation. *Agricultural and Forest Meteorology* 151, 669-681.
- 562 Stella P., Personne E., Lamaud E., Loubet B., Trebs I., Cellier P., 2013. Assessment of the  
563 total, stomatal, cuticular, and soil 2 year ozone budgets of an agricultural field with  
564 winter wheat and maize crops. *Journal of Geophysical Research: Biogeosciences* 118,  
565 1–13, doi:10.1002/jgrg.20094.
- 566 Stella P., Personne E., Loubet B., Lamaud E., Ceschia E., Béziat P., Bonnefond J.M., Irvine  
567 M., Keravec P., Mascher N., Cellier P., 2011a. Predicting and partitioning ozone  
568 fluxes to maize crops from sowing to harvest: the Surf<sub>atm</sub>-O<sub>3</sub> model. *Biogeosciences*  
569 8, 2869-2886.
- 570 Stevenson D.S., Dentener F.J. Schultz M.G. Ellingsen K., van Noije T.P.C., Wild O., Zeng  
571 G., Amann M., Atherton C.S., Bell N., Bergmann D.J., Bey I., Butler T., Cofala J.,

- 572 Collins W.J., Derwent R.G., Doherty R.M., Drevet J., Eskes H.J., Fiore A.M., Gauss  
573 M., Hauglustaine D.A., Horowitz L.W., Isaksen I.S.A., Krol M.C., Lamarque J.F.,  
574 Lawrence M.G., Montanaro V., Müller J.F., Pitari G., Prather M.J., Pyle J.A., Rast S.,  
575 Rodriguez J.M., Sanderson M.G., Savage N.H., Shindell D.T., Strahan S.E., Sudo K.,  
576 Szopa S., 2006. Multimodel ensemble simulations of present-day and near future  
577 tropospheric ozone. *Journal of Geophysical Research* 111, D08301,  
578 doi:10.1029/2005JD006338.
- 579 Sorimachi A., Sakamoto K., 2007. Laboratory measurement of dry deposition of ozone onto  
580 Northern Chinese soil samples. *Water, Air, and Soil Pollution: Focus* 7, 181-186.
- 581 Tuovinen J.P., Ashmore M.R., Emberson L.D., Simpson D., 2004. Testing and improving the  
582 EMEP ozone deposition module. *Atmospheric Environment* 38, 2373-2385.
- 583 Van Pul W.A.J., Jacobs A.F.G., 1994. The conductance of a maize crop and the underlying  
584 soil to ozone under various environmental conditions. *Boundary-Layer Meteorology*  
585 69, 83-99.
- 586 Vickers D., Mahrt L., 1997. Quality Control and Flux Sampling Problems for Tower and  
587 Aircraft Data. *Journal of Atmospheric and Oceanic Technology* 14, 512-526.
- 588 Vuolo R.M., Loubet B., Mascher N., Gueudet J.C., Durand B., Laville P., Zurfluh O., Ciuraru  
589 R., Stella P., Trebs I., 2017. Nitrogen oxides and ozone fluxes from an oilseed-rape  
590 management cycle: the influence of cattle slurry application. *Biogeosciences* 14, 2225-  
591 2244.
- 592 Wesely M.L., Hicks B.B., 2000. A review of the current status of knowledge on dry  
593 deposition. *Atmospheric Environment* 34, 2261-2282.
- 594 Wilczak J.M., Oncley S.P., Sage S.A., 2001. Sonic anemometer tilt correction algorithms.  
595 *Boundary-Layer Meteorology* 99, 127-150.
- 596 Wild O., 2007. Modelling the global tropospheric ozone budget: exploring the variability in  
597 current models. *Atmospheric Chemistry and Physics* 7, 2643-2660.
- 598 Wittig V.E., Ainsworth E.A., Naidu S.L., Karnosky D.F., Long S.P., 2009. Quantifying the  
599 impact of current and future tropospheric ozone on tree biomass, growth, physiology  
600 and biochemistry: a quantitative meta-analysis. *Global Change Biology* 15, 396-424.
- 601 Zhang L., Brook J.R., Vet R., 2002. On ozone dry deposition – with emphasis on non-  
602 stomatal uptake and wet canopies. *Atmospheric Environment* 36, 4787-4799.
- 603 Zhang L., Vet R., Brook J.R., Legge A.H., 2006. Factors affecting stomatal uptake of ozone  
604 by different canopies and a comparison between dose and exposure. *Science of the*  
605 *Total Environment* 370, 117-132.  
606