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

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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 (O₃) 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⁻² 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 (CO₂,
42 N₂O, CH₄, and halocarbons) (IPCC, 2013). Yet, due to its oxidative capacity, O₃ 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, O₃ 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 O₃ 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 O₃ from
57 the atmosphere and therefore an important process governing the tropospheric O₃ 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 O₃ dry
60 deposition, (ii) establish parameterizations for O₃ deposition, and (iii) quantify the terrestrial
61 O₃ 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), O₃ 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 O₃ deposition was the main deposition pathway and
76 represented 55% of the total O₃ 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 O₃ resistance (R_{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 O₃ 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_{soil} was
82 linked to soil surface relative humidity (RH_{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_{soil} and RH_{surf} and its dependence
85 on soil texture, with data obtained only over bare soil are used. A new parameterization of
86 R_{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 (RH_a) as well as turbulent fluxes (sensible (H) and latent (LE) heat
91 fluxes, and momentum (τ) flux from which is deduced friction velocity (u_*)) and O₃
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 O₃
99 chemiluminescent analyzer (OS-G-2, Güsten, 1992). Its calibration was continuously checked
100 against a slow-response O₃ monitor (O₃ 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 O₃ chemiluminescent analyzer
106 (ATDD, NOAA, USA). Owing to the very small and constant offset of the O₃ 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, O₃ deposition velocity was measured by
111 eddy-covariance following the ratio method (Müller et al., 2010) by using a fast-response O₃
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, CO₂ and O₃ 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 CO₂/H₂O Infrared Gas Analyzer (LI-7500, LICOR, USA), and
128 a fast-response O₃ chemiluminescent analyzer (ATDD, NOAA, USA). The coumarin dye of
129 the fast-response O₃ 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 O₃, 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 O₃ 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 O₃ analyser (1308, SIR, E) was used to calibrate the fast-response O₃
 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 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} (s m^{-1}) is the soil resistance, and $R_a(z)$ and R_{bO_3} (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 $R_{soil\ min}$ ($s\ m^{-1}$) is the soil resistance without water adsorbed at the surface (i.e. at
 166 $RH_{surf} = 0\%$) and k is an empirical coefficient of the exponential function.

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

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

$$170 \quad \chi_{H_2O_{surf}} = E(R_a(z) + R_{bH_2O}) + \chi_{H_2Oa} \quad (5)$$

$$171 \quad P_{vapsurf} = \frac{\chi_{H_2O_{surf}} R (T_{surf} + 273.15)}{M_{H_2O}} \quad (6)$$

$$172 \quad P_{sat}(T_{surf}) = p \exp \left[\frac{M_{H_2O} 10^{-3} L}{R} \left(\frac{1}{T_0 + 273.15} - \frac{1}{T_{surf} + 273.15} \right) \right] \quad (7)$$

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

174 with T_{surf} the surface temperature ($^{\circ}C$), ρ the air density ($kg\ m^{-3}$), C_p the air specific heat
 175 ($J\ kg^{-1}\ K^{-1}$), $\chi_{H_2O_{surf}}$ and χ_{H_2Oa} the air concentration of water ($g\ m^{-3}$) at z_0 and reference height,
 176 respectively, E the water vapor flux ($kg\ m^{-2}\ s^{-1}$), $P_{vapsurf}$ the water vapor pressure at z_0 (Pa), R
 177 the universal gas constant ($J\ mol^{-1}\ K^{-1}$), M_{H_2O} the molecular weight of water ($g\ mol^{-1}$),
 178 $P_{sat}(T_{surf})$ the saturation vapor pressure at T_{surf} (Pa), p the atmospheric pressure (Pa), L the
 179 latent heat of vaporization of water ($J\ kg^{-1}$), and T_0 the boiling temperature of water ($^{\circ}C$).

180 3 – RESULTS AND DISCUSSION

181 3.1 – Weather, pedoclimate and ozone deposition

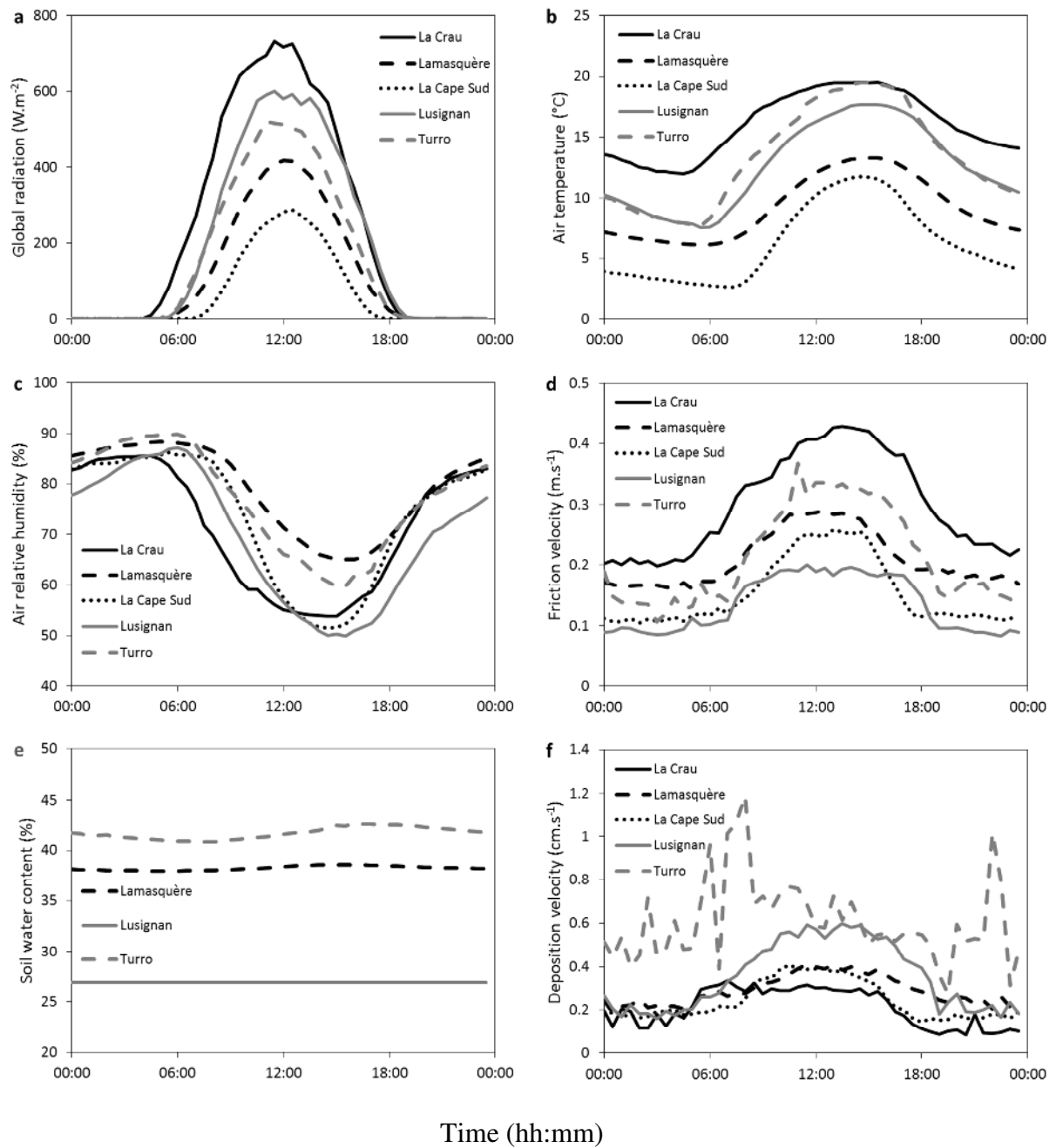
182 Meteorological variables followed typical diurnal trends at each sites. Global radiation and u_*
 183 increased in early morning to reach their maximum at around noon and then decreased to their
 184 minimum in late afternoon (Figures 1a and 1d). Air temperature and RH_a exhibited opposite
 185 trends: 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 $R_g = 232\ W\ m^{-2}$ over the whole experimental period (Table 1), and

193 maximum half hourly R_g around 730 W m^{-2} (Figure 1a). Air temperature and relative
194 humidity were on average at 15.8°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 m s^{-1} on average (Table 1) and varied between 0.2 m s^{-1} during nighttime and
198 0.43 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 W m^{-2} for the whole dataset (Table 1) and its mean
201 half hourly value peaked at 415 W m^{-2} (Figure 1a). Half hourly means of T_a and RH_a ranged
202 between 6°C and 90% during nighttime and 13°C and 65% during daytime (Figure 1b and
203 1c), and averaged 9.2°C and 79% over the whole dataset (Table 1), respectively. Mean u_* was
204 at 0.21 m s^{-1} (Table 1) over the whole dataset and ranged from 0.16 m s^{-1} during nighttime
205 and 0.29 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 W m^{-2}
210 and 6.2°C (Table 1), and half hourly means ranging from 0 to 273 W m^{-2} and from 2.5 to
211 12°C (Figure 1a and 1b), respectively. Dry atmospheric conditions were measured: although
212 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 m s^{-1} (Figure 1d), with
214 mean u_* of 0.15 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 W m^{-2} (Figure 1a) and averaged 195 W m^{-2} over the whole dataset (Table 1).
218 Means of T_a and RH_a were at 12.6°C and 69% (Table 1) and their half hourly means ranged
219 from 7.5°C to 17.7°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 m s^{-1} and 27%,
221 respectively (Table 1). Half hourly means of u_* only varied from 0.09 m s^{-1} during nighttime
222 to 0.20 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 W m^{-2} (Table 1) and its half hourly mean peaked

227 to 516 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°C to 19°C (Figure 1b).
229 Over the whole dataset, T_a was on average at 13.3°C (Table 1). For 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 m s^{-1} during nighttime to 0.37 m s^{-1} during daytime (Figure 1d) while it was on average at
233 0.21 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 cm s^{-1} at La Crau, 0.40 cm s^{-1} at Lamasquère
240 and La Cape Sud, and 0.60 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 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 cm s^{-1} for La Crau, 0.29 cm s^{-1} for Lamasquère, 0.26 cm s^{-1} for La
246 Cape Sud, and 0.36 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 ⁻²	°C	%	m s ⁻¹	mm		%			cm s ⁻¹	
		Mean \pm SD	Mean \pm SD	Mean \pm SD	Mean \pm SD	Mean \pm SD			Mean \pm SD		Mean \pm SD	
La Crau	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	
Lamasquère	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	
La Cape Sud	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	
Lusignan	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	
Turro	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}

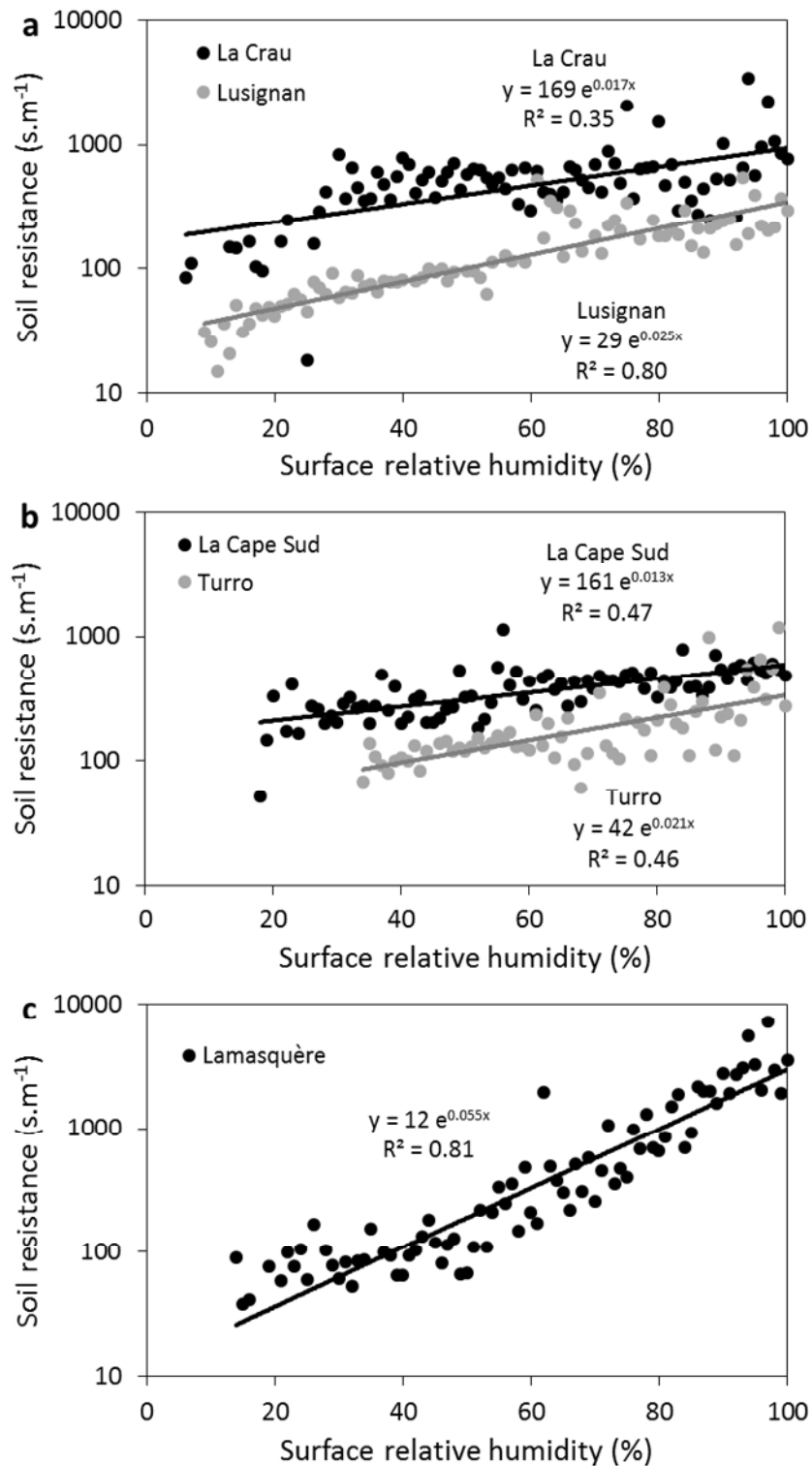
261 Several authors related R_{soil} to the amount of water in soil, and showed that the drier the soil
 262 is, the weaker R_{soil} is (e.g., Bassin et al., 2004; Massman, 2004; Meszaros et al., 2009). More
 263 recently, Stella et al. (2011b) showed that (i) the estimate of R_{soil} as a function of 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_{soil} and RH_{surf} (Figure 2), and are
269 summarized in Table 2.

270 Whatever the site considered, R_{soil} increases exponentially with RH_{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_{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 s m^{-1} . In such conditions, it can be hypothesized that RH_{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_{soil} with RH_{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 ± 0.003 and 0.013 ± 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 ± 0.001 and 0.021 ± 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 $u_* > 0.1\ m\ s^{-1}$ 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_{soil} and RH_{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 ⁻¹	-	%
	Value ± SE	Value ± SE	
Lamasquère	12 ± 1.20	0.055 ± 0.003	54
Stella et al. (2011b)¹	21 ± 1.01	0.024 ± 0.001	31
Lusignan	29 ± 1.01	0.025 ± 0.001	17
Turro	42 ± 1.22	0.021 ± 0.003	30
La Cape Sud	161 ± 1.11	0.013 ± 0.002	6
La Crau	169 ± 1.18	0.017 ± 0.003	22
Güsten et al. (1996)²	800	[-]	0.8

302 ¹ Soil texture: 31% Clay, 62.5% Silt, 6.5% Sand

303 ² Soil texture: 0.8% Clay, 1.3% Silt, 97.9% Sand

304 3.3 – Dependence of R_{soil} parameters to soil texture

305 As shown previously, the dependence of R_{soil} to RH_{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_{soil} with RH_{surf} is due to the presence of water adsorbed at the soil surface,
 308 decreasing the surface active for O_3 deposition since O_3 is hardly soluble in water i.e., R_{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 m² g⁻¹): 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

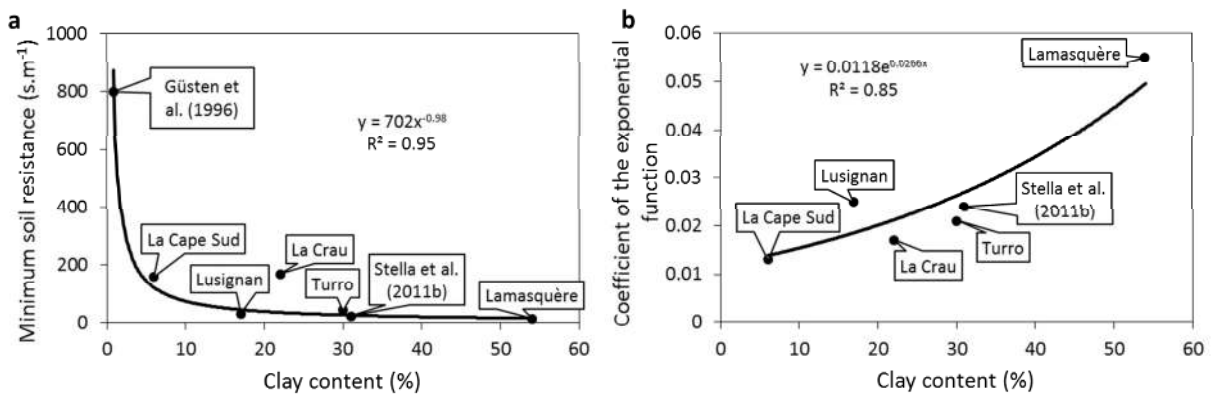
317 larger the surface available at a microscopic scale for O₃ deposition is. To examine this issue,
318 the two parameters $R_{\text{soil min}}$ and k were plotted as a function of soil clay content.

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, O₃ deposition to soil is favored when the soil specific surface increases, since more
332 surface is available for O₃ 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 O₃ deposition onto different soil samples, they reported that for
335 soil moisture content lower than 10% (i.e., close to RH_{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 O₃ is hardly soluble in water, this statement could explain the faster increase of R_{soil}
344 with RH_{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 O₃ 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_{soil} increases with RH_{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_{soil}
 367 $_{\text{min}}$, and the increase of R_{soil} with RH_{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_{soil} as a function of RH_{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 O₃ 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_{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 O₃ deposition at a microscopic scale.
388 Additional chemical processes that remove O₃ at the interface soil-atmosphere remain
389 possible. For instance, the study performed by Vuolo et al. (2017) indicated that soil O₃
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 O₃ 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 O₃ 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

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