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# Vertical partitioning and controlling factors of gradient-based soil carbon dioxide fluxes in two contrasted soil profiles along a loamy hillslope.

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## 21 Abstract

We assessed soil  $CO_2$  fluxes throughout two contrasted soil profiles along a hillslope in the central loess belt of Belgium. First, we measured time-series of soil temperature, soil moisture and  $CO_2$ concentration at different depths in the soil profiles for two periods of 6 months. Subsequently we calculated the  $CO_2$  flux at different depths, using Fick's diffusion law and horizon specific diffusivity coefficients. The soil diffusivity coefficients were calibrated using profile specific surface  $CO_2$  flux chamber measurements. The calculated fluxes allowed assessing the contribution of different soil layers to surface  $CO_2$  fluxes and elucidating deep soil controlling factors on  $CO_2$  emission.

The results show that approximately 90 to 95 % of the surface CO2 fluxes originate from the first 10 centimeters of the soil profile at the footslope. This indicates that soil OC at such a footslope can be stored along the main part of the soil profile (below 10 cm) and submitted to a long-term stabilization. We also observe that time-series of soil  $CO_2$  emissions at the summit are in accordance with the temporal dynamics of temperature. In contrast, at the footslope, we highlight that long periods of  $CO_2$ accumulation alternate with peaks of important surface release due to the high water filled pore space that limits the transfer of  $CO_2$  along the soil profile at this slope position.

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# 38 **1. Introduction**

Soils play a major role in the global C budget, as they contain 2 to 3 times more C than the atmosphere 39 (Eswaran et al., 1993; Lal et al., 2003). There is now significant concern about the contribution of soil 40 41 OC to future climate change where a climate change driven acceleration of soil OC decomposition 42 could represent a positive feedback on climate. In addition to the role of soil mineralogy and microbial 43 communities, recent studies highlight the importance of soil bio-physical conditions that may vary 44 substantially with time and across landscapes (e.g. Dai et al., 2012). In addition to the combined 45 effects of soil moisture, temperature and OC quality on soil microbial activity (e.g. Wiaux et al., 46 2014b), recent studies show the importance of physical controls on  $CO_2$  fluxes such as gas diffusion barriers along soil profiles .(e.g. Ball, 2013; Maier et al., 2011). Furthermore, most process studies so 47 48 far have focused on the soil surface layer while there is now increasing awareness that subsoil OC represents an important C store that interacts with the atmosphere (Rumpel and Kögel-Knabner, 49 50 2011). Recent studies (Rumpel and Kögel-Knabner, 2011) highlighted that deep soil OC is highly processed, and showed the need to consider C fluxes originating from deeper soil horizons. This is 51 52 particularly relevant in landscapes with complex topography where buried OC in depositional areas contributes substantially to soil C emissions (e.g. Van Oost et al., 2012; Wang et al., 2014 and Wiaux 53 et al., 2014a). Goffin et al. (2014) showed that the upper first 30 centimeters of a forest soil profile 54 contribute substantially to the total surface CO<sub>2</sub> flux. However, to our knowledge, a vertical 55 partitioning has not been evaluated in agro-ecosystems or in systems with contrasting soil physical 56 and/or chemical properties. 57

In this study, we aim to elucidate the role of physical controls on soil-atmosphere  $CO_2$  fluxes and its variation with soil depth. To that aim, we present a comparative analysis between two contrasting soil profiles along an eroded and cultivated hillslope. The objectives of this study are: (i) to quantify the relative contribution of soil surface and subsoil OC to  $CO_2$  fluxes through a vertical partitioning of these fluxes; and (ii) to identify the role of soil physical properties using time-series of soil moisture measurements and gas diffusivity at different depths. The selected study site is characterized by two contrasting soils in terms of soil hydrological regimes and structure.

#### 65 2. Material and methods

#### 66 2.1. Study site description

The study was carried out in the Belgian loam belt along a cultivated hillslope of 150 meters length 67 (50.6669°N, 4.6331° W). The site has a maritime temperate climate, with an average annual 68 69 temperature of 9.7°C and an average annual precipitation of 805 mm. The slope percentage in the backslope area ranges between 8.5 and 16%, with a mean slope of 12%. The slope percentage in the 70 convex shoulder area ranges between 4 to 8.5%, with an average of 6%. The field was plowed (0-30 71 72 cm soil surface layer) every year. Each year, manure and nitrate fertilization was carried out. The 73 previous crop rotation was winter wheat, maize and spring wheat. The study site has been described in 74 detail in Wiaux et al. (2014a,b). For this study, we selected two measurement stations along the 75 hillslope: one at the summit and one at the footslope position. The soil is a Dystric Luvisol type at the 76 summit and a Colluvic Regosol in the depositional area at the footslope (Wiaux et al., 2014a,b). The 77 soil properties of these two soil profiles have been characterized by Wiaux et al. (2014a,b): soil total 78 OC, labile OC and porosity profiles are illustrated in Fig. 1 and 2, respectively.

We measured the total porosity (alpha) in the laboratory by weighing 100 cm<sup>3</sup> undisturbed soil cores both at saturation and after oven drying at 105°C for 48h. We deduced alpha from the mass of water needed to fill sample pores. We calculated the air-filled porosity ( $\varepsilon$ ) as the difference between alpha and volumetric water content (VWC). We calculated average and standard deviation values on triplicate samples for each depth.

We characterized soil water retention (SWR) curves using undisturbed soil cores at 10, 25, 35, 50, 70 and 95 cm depth, with 3 replicates at each depth. We obtained the  $\varepsilon_{100}$  and b parameters of the Campbell (1974) SWR model by fitting the model to the SWR observations (Moldrup et al. 2000).

# 87 2.2. Monitoring of soil CO<sub>2</sub>, water and temperature

We measured soil CO<sub>2</sub> concentrations using purpose-built soil CO<sub>2</sub> probes. The CO<sub>2</sub> sensor in the
probe is based on the CARBOCAP® Single-Beam Dual Wavelength non-dispersive infra-red (NDIR)

90 technology (GMM221, Vaisala corp., Vantaa, Finland). The analytical precision is 1.5% of the measurement range added to 2% of the observed value. The sampling head of the CO<sub>2</sub> probe is a 91 cylinder of 18.5 mm diameter and 40 mm long, covered with a PTFE (polytetrafluoroethylene) 92 membrane, enabling gas exchange and protection against water infiltration. Since the GMM221 93 sensors were not designed for wet soil conditions, the sensors were encapsulated into an additional 94 perforated PVC tube, providing an additional protection against water (Fig. 1). This tubing method is 95 96 an adaptation of the technique presented by Young et al. (2009). We inserted these tubes vertically into the soil, after creating bore holes with a diameter that equals the diameter of the PVC tubes. This 97 approach avoids the need to backfill the bore hole, which will disturb the soil structure and diffusion 98 99 process. Two rubber stoppers, one at 155 mm from the tube head, and another at the top of the tube, prevented atmospheric air from penetrating into the gas sampling volume. Petroleum jelly on these 100 two rubber stoppers ensured a perfect air- and water-tightness and we verified this under laboratory 101 102 conditions before using the probes. We used a nylon membrane to avoid soil particles entering the 103 perforated tube and to limit further water infiltration.

104 We monitored soil temperature using a thermistor probe (Therm107, Campbell Scientific Lt., UK). 105 Analytical precision is 0.4°C. We monitored soil volumetric water content (VWC) using Time 106 Domain Reflectrometry (TDR) probes. We used Topp's equation (Topp et al., 1980) to determine 107 VWC from the measured apparent dielectric constant measured. We used the parameters of the Topp's equation as identified by Beff et al. (2013). In this study latter study, the Topp's equation was 108 calibrated for an experimental field in the close vicinity of our study site, using the method of 109 Heimovaara (1993) and following the protocol described by Garré et al. (2008). We recorded water, 110 temperature and CO<sub>2</sub> concentration profiles measurements with an automatic data logger (CR1000, 111 Campbell Scientific Lt., UK), connected to a multiplexer (AM16/32, Campbell Scientific, Campbell 112 Scientific Lt., UK). 113

114 In order to obtain an equilibrated soil environment around the soil VWC, temperature and  $CO_2$  probes, 115 we started measurements 1 month after the installation of the probes. We covered the measurement plots with a synthetic permeable geotextile during the complete measurement period. This avoidedvegetation growth and any autotrophic contribution to the soil respiration.

The sampling design is depicted in Fig. 4. At each of the 2 slope positions, we measured soil VWC 118 119 and  $CO_2$  concentrations profiles with 3 replicates on each measurement depth (Fig. 4). We collected 120 18 VWC profiles (6 soil depths, 3 replicates), at each of the 2 slope positions. We measured VWC at a depth of 10, 25, 35, 50, 70 and 95 cm depths (Fig. 4). We measured the temperature at 4 soil depths 121 122 (10, 25, 45, 85 cm) without replicates (Fig. 4). We measured CO<sub>2</sub> concentrations at a depth of 10, 25, 123 45 and 85 cm. We also performed surface  $CO_2$  fluxes measurements with an infra-red gas analyzer 124 (IRGA) linked to a survey chamber at 16 dates (profile and surface sampling time was within a 30 125 minutes time interval). Note that the averaged values of CO<sub>2</sub> concentration for each observation depth 126 cover the same area as the IRGA chamber network located at the soil surface (Fig. 4). These reference 127 surface CO<sub>2</sub> fluxes allowed calibrating parameters of the soil gas diffusion model, ensuring the accuracy of profile  $CO_2$  fluxes (section 2.3). 128

We adjusted the concentration ranges of the  $CO_2$  probe for each soil depth and for each slope position. This allowed an optimal fit of the probes to the local concentrations. Each probe has to characterize the entire range of values encountered during the seasons while at the same time, it should have a sufficiently narrow measurement range to ensure measurement precision At the summit position, measurements ranged between 0-2 % at 12, 25, 45 cm depth and between 0-5 % at 85 cm depth. At the footslope position, measurements ranged between 0-5 % at 12 cm depth, between 0-10 % at 25 and 45 cm depth and between 0-20 % at 85 cm depth.

We recorded hourly time-series of VWC, temperature and  $CO_2$  concentrations from 12 May to 13 December 2012 and from 14 May to 22 November 2013 at the footslope position and from the 2 June to 13 December 2012 and from the 14 June to 22 November 2013 at the summit position. In 2012, important parts of  $CO_2$  measurements were not recorded as a result of sensors failures and/or the use of an unsuitable initial measurement range of some sensors. To increase the quality of the soil concentration data time-series, we removed observations where the battery voltage was lower than 11.5 V. We also corrected soil profile  $CO_2$  concentrations measurements for temperature variations using the empirical formulas described by Tang et al. (2003). This allowed removing the impact of temperature on the  $CO_2$  reading of the  $CO_2$  probe, since the CARBOCAP® technology is temperature dependent. The probe manufacturer (Vaisala corp., Vantaa, Finland) provided probe specific parameters values for the correction formulas.

We averaged triplicate VWC and  $CO_2$  concentrations data, providing an average value for each soil depth and slope position. Note that averaging strategy allows to account for the spatial variability of VWC and  $CO_2$  concentrations (Maier and Schack-Kirchner, 2014), by extending the measurement footprint to an area of c. 5 m<sup>2</sup>.

We calculated soil temperature and VWC profiles using a linear interpolation between the depth specific values within the profile. We kept the values constant between the sampling point at the top of the profile and the soil surface. We calculated the  $CO_2$  concentrations profiles by fitting Eq. 2 to the observations. We evaluated the performance of this fitting by means of the regression coefficient ( $R^2$ ). When the  $R^2$  values were lower than a threshold value of 95%, we considered the  $CO_2$ concentration profile as unreliable and we did not retain the resulting  $CO_2$  fluxes in final analysis.

# 157 **2.3. Calculation of the CO<sub>2</sub> fluxes**

We calculated the CO<sub>2</sub> flux using Fick's first law of diffusion according to the gradient method (Eq. 1,
e.g. Maier and Schack-Kirchner, 2014):

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$$F_{CO_2} = -D_s \frac{\partial CO_2}{\partial z}$$
(Eq. 1)

where  $F_{CO_2}$  is the soil CO<sub>2</sub> flux [µmol m<sup>-2</sup> s<sup>-1</sup>], D<sub>s</sub> the diffusivity of CO<sub>2</sub> in soil [m<sup>2</sup> s<sup>-1</sup>], CO<sub>2</sub> the soil CO<sub>2</sub> concentration [µmol m<sup>-3</sup>], and  $\frac{\partial CO_2}{\partial z}$  the vertical soil CO<sub>2</sub> gradient.

In order to calculate the vertical soil CO<sub>2</sub> gradient, we used a double sigmoidal equation (Eq. 2), which
allows accounting for some curve concavity variations (Maier and Schack-Kirchner, 2014):

165 
$$\operatorname{CO}_{2}(z) = 0.04 + A\left(\left(\frac{1}{1+e^{-\gamma_{1}z}}\right) + \left(\frac{1}{1+e^{-\gamma_{2}(z-d)}}\right) - \left(\frac{1}{2} + \frac{1}{e^{\gamma_{2}d} + 1}\right)\right)$$
(Eq. 2)

where z is the soil depth [cm], d is the soil depth [cm] at which the sharpness of the curve changes due to a diffusion barrier,  $\gamma_1$  and  $\gamma_2$  [cm<sup>-1</sup>] are fitted parameters which characterize the sharpness of the curve, respectively above and below the soil depth d, and A [%] is a reference value used to define the fitted asymptotic value of the CO<sub>2</sub> concentration at infinite depth. We fitted the A, d,  $\gamma_1$  and  $\gamma_2$ parameters for each CO<sub>2</sub>-profile using the trust-region-reflective optimization algorithm in Matlab ©. The derivative of Eq. 2 provided the CO2 gradient ( $\frac{\partial CO_2}{\partial z}$ ) used in Eq. 1.

172 The diffusivity of CO<sub>2</sub> in soil, D<sub>s</sub> (Eq. 1) is a function of the diffusivity of CO<sub>2</sub> in free air (varying 173 with temperature T and pressure, e.g. Davidson *et al.*, 2006) and of the gas tortuosity factor ( $\xi$ ) (Eq. 3):

174 
$$D_s = \xi \, 1.47 \, 10^{-5} \left(\frac{T+273}{273}\right)^{1.75}$$
(Eq. 3)

where  $\xi$  depends on soil physical and hydrological properties. We used the Moldrup *et al.* (2000) model (Eq. 4) which was shown to provide the most accurate and precise results (Davidson et al., 2006; Goffin et al., 2014);

178 
$$\xi = (2\varepsilon_{100}^{3} + 0.04\varepsilon_{100}) \left(\frac{\varepsilon}{\varepsilon_{100}}\right)^{2+3/b}$$
(Eq. 4)

179 where  $\xi$  is the gas tortuosity factor,  $\varepsilon$  [m<sup>3</sup> m<sup>-3</sup>] is the soil air-filled porosity, b [-] is the slope of the 180 Campbell (1974) soil water retention curve model between -100 and -500 cm H<sub>2</sub>O water suction, and 181  $\varepsilon_{100}$  [m<sup>3</sup> m<sup>-3</sup>] is the soil air-filled porosity at a soil water potential of -100 cm H<sub>2</sub>O.

182  $CO_2$  fluxes, as assessed by the gradient based method, were calculated on an hourly time-scale, and 183 then integrated on a daily basis. Temperature, VWC, diffusivity and  $CO_2$  concentration values were 184 also averaged on a daily basis.

185 In contrast to other studies (e.g. Pingintha et al., 2010; Turcu et al., 2005), we did not aggregate the 186 soil diffusivity coefficient for the entire soil profile or for an entire soil layer. We considered the vertical distribution explicitly, and integrated Eq. 4 in the finite difference numerical solution of Eq. 1. In this numerical integration, we used a depth increment of 0.1cm and constrained the surface  $CO_2$ concentrations with atmospheric  $CO_2$  levels (i.e. 400 ppm).

190 We calibrated the diffusion model by adjusting the parameters related to the gas diffusion coefficient (i.e. b and  $\varepsilon_{100}$ ) such that calculated fluxes fit punctual CO<sub>2</sub> fluxes observations at 16 dates spread 191 192 along the measurement period. We obtained these observations by means of a portable infra-red gas analyzer with an automated closed dynamic chamber (LI-8100A system, LI-COR, United-States), 193 following Davidson et al. (2002). The sampling design of these surface chamber CO<sub>2</sub> fluxes 194 measurements on the same study site has been described in Wiaux et al. (2014 b). Comparing the 195 196 gradient-based CO<sub>2</sub> fluxes with directly measured IRGA CO<sub>2</sub> fluxes, we obtained a good prediction with a R2 of 92% for all soil types together. This ensures the consistency (and consequently the 197 198 precision) of the calculated fluxes. The slope of the fit (i.e. 1.05 and 1.22, respectively in 2012 and 199 2013) was used to correct the calculated fluxes and to ensure the accuracy.

#### 200 **2.4. Vertical partitioning of CO<sub>2</sub> fluxes**

We partitioned the continuous  $CO_2$  flux profiles obtained using Eq.2 into 10 slides of 10 centimeters along the soil profile. For each soil slide, we calculated the difference between the top and bottom fluxes. We divided this difference by the total  $CO_2$  flux (e.g. the value at the soil surface). This provides the relative contribution in terms of both  $CO_2$  production and transfer (in %) of each soil slide to the surface  $CO_2$  flux (e.g. Goffin et al., 2014; Maier and Schack-Kirchner, 2014).

In order to allow an easy representation of the temporal dynamic of this vertical partitioning, we
averaged values on a semi-seasonal time-scale. Standard deviation values reflect the variability over
time during each semi-season.

## 209 **3. Results**

# 210 **3.1. Spatio-temporal analysis of measured soil variables**

Fig. 5 shows the spatio-temporal variation of soil temperature and moisture, while Fig. 6 shows the spatio-temporal variation of  $CO_2$  fluxes, concentrations and diffusion. All these values correspond to in-situ measurements during a 6 month period in 2013. Similar measurements have been carried out in 2012 and display similar spatio-temporal trends (data not shown).

During the observation period, the soil temperature (Fig. 5A) did not significantly differ between the
summit and the footslope, although higher temperatures were observed at the summit profile for some
shorter periods (e.g. day of year 180 to 220 where temperatures are approximately 2 to 3 °C higher).
The mean daily surface temperatures range between 4°C to 28°C at the summit, and between 4°C to
25°C at the footslope.

220 The space-time dynamics of the soil volumetric water content (VWC, Fig. 5B) differ substantially 221 between the summit and the footslope profiles. At the footslope, the observed soil VWC at different soil depths varied in a narrow range (0.36 to 0.39 cm<sup>3</sup> cm<sup>-3</sup>). In contrast, soil VWC at the summit 222 varied between 0.23 to 0.34 cm<sup>3</sup> cm<sup>-3</sup> for the plow layer (0-30cm depth) and higher values 223 (approximately 0.5 cm<sup>3</sup> cm<sup>-3</sup>) were observed for the rest of the soil profile. The soil at the summit 224 position was the wettest during the early spring and the late autumn and driest in the summer. At the 225 226 footslope, soil VWC reached the saturation level in the early summer after an important rainfall event and then slowly decreased until the early autumn and reached saturation again in the late autumn. 227

In contrast to the VWC, the soil gas diffusivity (Fig. 6C) reached its maximum value in the summer at the summit while it was low at the footslope. Soil gas diffusivity was approximately 10 times lower at the footslope than at the summit.

The soil  $CO_2$  concentrations at both the summit and the footslope increased gradually from spring to late summer. Thereafter, concentrations dropped again and lowest values were observed in the late autumn. The ranges of  $CO_2$  fluxes obtained for the footslope and summit profiles were very similar. However, their temporal distribution was different: the periods characterized by high  $CO_2$  fluxes did not occur at the same time and had a different duration. For all soil profiles,  $CO_2$  fluxes decreased with depth and reached null values at approximately30 cm depth at the summit and at approximately 15 cm depth at the footslope.

#### **3.2. Shape and variability of CO<sub>2</sub> concentrations and fluxes profiles**

The observed soil  $CO_2$  concentrations (Fig. 6Bb) increased with soil depth, from the atmospheric value of 0.04 % at the surface to concentrations which were two orders of magnitude higher at 100 cm depth ( $CO_2$ ,(z) in Eq.2) (Fig. 6Bb). For the measurement period of 6 months considered here,  $CO_2$ concentration values at 100 cm depth were three to four times higher at the footslope position than at the summit position. In 2013, these values ranged between 0.86 to 3.46 % at the summit position and between 3.68 to 9.12 % at the footslope position.

245 The observed  $CO_2$  concentration profiles followed a double exponential trend (Eq. 2). This particular 246 model fits our observations relatively well, with regression coefficients ranging between 97 to 100%. 247 The second exponential curve starts approximately at the middle of the profile, and is particularly 248 pronounced at the footslope, reflecting a shift of nearly 4% CO<sub>2</sub> between 44 and 100 cm depth. 249 Standard deviations around averaged values of observed hourly CO<sub>2</sub> concentrations at each depth are given in Table 1. The small-scale spatial variability is low relative to the mean values of CO<sub>2</sub> 250 concentrations, the only exception being the footslope at 25 cm depth where the maximum standard 251 deviation exceeded the maximum mean value. 252

The CO<sub>2</sub> fluxes (Fig. 6A) were calculated based on both CO<sub>2</sub> concentrations and diffusivity. For all soil profiles, CO<sub>2</sub> fluxes decreased with depth and reached null values at c.30 cm depth at the summit and at c. 15 cm depth at the footslope.

# 256 **3.3. Vertical partitioning of CO<sub>2</sub> fluxes**

The distribution of the soil  $CO_2$  fluxes in the profile is illustrated in Fig. 7. At the footslope, 90 to 95 % of the surface  $CO_2$  fluxes were generated in the first ten centimeters of the soil profile. The soil 259 layer between 10 and 20 cm contributed for only 5 to 10 %, and the deeper layers did not significantly contribute to the surface fluxes. At the summit, the relative contribution of the different soil layers was 260 261 more dynamic in time, with a contribution of the first ten centimeters of the soil profile ranging from 80 % at the late spring, decreasing to 60 % in the early summer, and reaching 40 % from late summer 262 to the late autumn. At the summit, the first 30 centimeters of the soil profile significantly contributed 263 to surface fluxes. This contribution decreased with depth in the late spring and the early summer, but 264 265 is homogeneously distributed with depth for the rest of the time. At the summit, soil layers deeper than 30 cm depth sometimes contributed for up to 20% of the total flux, especially in the autumn. Between 266 40 to 50 cm depth, and 80 to 90 cm depth, some negative contribution (i.e. CO<sub>2</sub> uptake) up to -20% is 267 268 also observed.

269 **4. Discussion** 

# 270 4.1. Soil physical control on CO<sub>2</sub> emissions

The observed differences in the temporal dynamics of surface soil CO<sub>2</sub> fluxes between the footslope 271 and summit soil profiles (Fig. 6A) indicates that the controlling factors are not the same. At the 272 273 summit, on one hand, the dynamic of surface soil CO<sub>2</sub> fluxes (Fig. 6A) clearly follows the temperature variations (Fig. 5A, maximum during the summer). At the footslope, on the other hand, the soil 274 surface CO<sub>2</sub> flux was small even when temperature increased and remained relatively small 275 throughout the summer period (Fig. 6A). This is most likely related to the high VWC values observed 276 277 at the footslope (Fig. 5B), as it is well known that VWC negatively impacts soil CO<sub>2</sub> emissions (e.g. 278 Webster et al., 2008b; Perrin et al., 2012; Wiaux et al., 2014b). More precisely, we suggest that the factor controlling CO<sub>2</sub> emissions at the footslope is not only VWC but also the difference between the 279 280 VWC and the water saturation level of the soil pore spaces. While the VWC at the footslope remained high throughout the year, we observed that the soil surface CO<sub>2</sub> flux dramatically increased when the 281 gas diffusivity exceeded a threshold value of c.  $0.1 \text{ cm}^2 \text{ d}^{-1}$  (i.e. from day 255 to 305 of year 2013, Fig. 282 6A). Hence, we argue that the  $CO_2$  emissions at the footslope profile are related to the fact that a high 283 VWC both: (i) strongly limits the transfer of biotic  $CO_2$  along the soil profile, and (ii) reduces the 284

production of  $CO_2$  in itself due to the lack of oxygen for the microbial community. In both cases, the lower  $CO_2$  emissions at the footslope profile relative to the summit, are due to gas diffusion limitations (even indirectly in the case of oxygen lack). This is in sharp contrast to the summit profile where gas can easily diffuse throughout the year and along the entire soil profile (Fig. 6C).

289 In the period preceding the important CO<sub>2</sub> emissions (i.e. from day 255 to 305 of year 2013, Fig. 6A), 290 the soil  $CO_2$  cannot move along the soil profile and accumulates within soil pores. This results in a CO<sub>2</sub> concentration increase both in the early and the late summer, especially below 50 cm depth (Fig. 291 292 6). This phenomenon is particularly evident below the compacted soil layer between 40 cm and 50 cm 293 depth. Based on the porosity profile illustrated in Fig. 1, this suggests that for our footlslope soil 294 profile, which is a Colluvic Regosols, gas diffusion barriers strongly impact the CO<sub>2</sub> concentration 295 profile, and hence the temporal dynamics of soil surface  $CO_2$  fluxes. This is in agreement with recent 296 studies (e.g. Ball, 2013) that show that soil pore continuity and size are key to understand the 297 mechanisms regulating the soil gases emissions.

298 As a consequence, the significantly higher  $CO_2$  concentrations observed at the footslope, especially for 299 deeper soil layers, are probably not related to the large amount of labile OC that was found at this 300 position (shown in Wiaux et al., 2014a,b), but more likely result from the accumulation of CO2 during 301 periods with a very low diffusivity. Maier et al. (2011) showed that the  $CO_2$  efflux can deviate from 302 the instantaneous soil respiration due to  $CO_2$  storage into soil pore spaces. Hence, we suggest that at 303 the footslope, soil physical properties are the dominant control on surface  $CO_2$  fluxes. In summary, we 304 highlight that the mechanisms that govern soil surface  $CO_2$  emissions are highly variable in both space 305 and time. On a well-drained soil at the summit of a hillslope, the observed soil  $CO_2$  emissions were 306 directly related to soil microbial respiration and CO<sub>2</sub> production (demonstrated in Wiaux et al., 2014b). 307 However, at the footslope of the hillslope, which is characterized by a different hydrological regime, we observed that the temporal dynamic of soil  $CO_2$  emissions were more closely related to physical 308 309 transfer mechanisms: long periods of CO<sub>2</sub> production and accumulation alternate with periods of 310 important release at the soil surface.

#### **4.2. Soil organic carbon storage in downslope deposits**

312 The soil respiration rate can be interpreted as an indicator of soil OC persistence (e.g. Gregorich et al., 313 1994). However, a further analysis of what occurs along the soil profile is needed to thoroughly answer the question of the persistence of OC. The vertical partitioning of the soil CO<sub>2</sub> fluxes, as 314 illustrated in Fig. 7, shows that during the observation period, 90 to 95 % of the surface CO<sub>2</sub> flux 315 originated from the first ten centimeters of the soil profile at the footslope. Given the important 316 317 amount of OC until up to 100 cm depth in our study site (Fig. 1, Wiaux et al., 2014 a), this observation 318 is not in agreement with the study of Goffin et al. (2014), who suggested that the relative contribution of a soil layer to the surface CO<sub>2</sub> fluxes is related to OC distribution along the soil profile. However, 319 320 while similarities exist in the physical controls and the method used to calculate the vertical 321 partitioning, the study of Goffin et al. (2014) reports on CO<sub>2</sub> production in forest soils, preventing any 322 direct quantitative comparison.

In addition, the substantial contribution of the upper soil layers found here was not related to higher 323 324 temperatures (Fig. 5A), contrary to what was suggested by Takahashi et al. (2004). According to the CO<sub>2</sub> concentration and diffusivity profiles (Fig. 6C), the relative contribution of the soil layers to the 325 326 surface CO<sub>2</sub> flux is more likely governed by soil physical controls (Ball, 2013) rather than by biological production depending on thermal energy and OC substrate. Here, soil gas diffusivity 327 strongly decreases from 10 to 40 cm depth (where diffusivity is null) at the two slope positions, and 328 the profile of CO<sub>2</sub> concentration displays no gradient between 10 and 40 cm depth, particularly at the 329 330 footslope (Fig. 6A).

Here, we show that despite the fact that the footslope profiles generates  $CO_2$  fluxes which exceed those observed at the summit position (demonstrated in Wiaux et al., 2014b), the contribution of soil layers below 10 cm depth is very small (Fig. 7). The OC in the top layer of the soil profile (i.e.0-10 cm) contributed for c. 90% of the total  $CO_2$  flux at the footslope position (Fig. 7). This can be explained by environmental conditions specific to this 0-10 cm layer playing in favor of both microbial respiration and gas diffusion. There are no limitations related to both diffusion barriers and access to the oxygen disappear close to the soil surface. Hence, the only impact of soil VWC on soil
respiration is its positive effect as it provides a more easy access for soil micro-organisms to their OC
substrate, and to the enhancement of their metabolic activities by water (Akinremi et al., 1999;
Castellano et al., 2011; Herbst et al., 2008; Howard and Howard, 1993; Šimůnek and Suarez, 1993).
The combination of this high amount and high quality of soil OC (Fig. 1, as described by Wiaux et al.,
2014a) with this net positive effect of soil VWC results in a strong increase of microbial respiration
rates.

Finally, our results suggest that buried soil OC in colluvial deposits is effectively protected from mineralization below 10 cm depth, which corroborates the assumption of a long-term stabilization of buried OC in colluvial soils as suggested in the literature (e.g. Doetterl et al., 2012; Berhe et al., 2008, 2012a).

# 348 **5.** Conclusion

In this study, we evaluated the factors controlling soil carbon dioxide fluxes for two soil profiles along 349 350 a hillslope characterized by contrasting physical and chemical characteristics. At the summit position of the studied hillslope, the time course of surface soil CO<sub>2</sub> fluxes was strongly related to soil 351 temperature and maximum CO2 fluxes were observed during the summer. Here, the observed soil  $CO_2$ 352 353 emissions are directly related to soil micro-organisms respiration and associated biotic CO<sub>2</sub> 354 production. In contrast, the higher levels of water filled pore space observed at the footslope profiles, 355 strongly limited the transfer of biotic CO<sub>2</sub> throughout the soil profile. Here, the soil surface CO<sub>2</sub> flux substantially increased for limited amounts of time when the gas diffusivity exceeded a threshold 356 value. As a result, the time course of observed soil CO<sub>2</sub> emissions was to a large extent explained by 357 358 physical transfer mechanisms: long periods of accumulation alternate with shorter periods of important CO<sub>2</sub> release. The vertical partitioning of the soil CO<sub>2</sub> fluxes for the footslope profiles showed that, 359 during the observation period, 90 to 95 % of the surface CO<sub>2</sub> fluxes originated from the first 10 360 centimeters of the soil profile. This study highlights the need to include soil physical properties and 361 their dynamics directly into soil OC models. 362

# **363** Author contribution

F.W. designed the experiments, and carried out the research. M.V., K.V.O. and F.W. analyzed the results. F.W. wrote the main part of the paper and prepared the manuscript with contributions from all co-authors.

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# 528 Tables

529 Table 1. Range of standard deviation and averaged values of triplicated measured hourly CO<sub>2</sub> concentrations at each

530 depth, both at the summit and at the footslope position. This range is indicated by minimum (Min) and maximum

531 (Max) values encountered along time (hourly time series) during the 6 months measurement period.

	Summit position				Footslope position			
Soil depth [cm]	Min mean [%]	Max mean [%]	Min S.D. [%]	Max S.D. [%]	Min mean [%]	Max mean [%]	Min S.D. [%]	Max S.D. [%]
10	0.07	1.39	0.00	0.71	0.26	4.75	0.00	3.13
25	0.06	1.83	0.00	0.68	0.30	3.93	0.00	5.32
45	NI	NI	NI	NI	0.12	3.96	0.00	1.96
95	0.15	2.83	0.00	1.42	0.48	7.52	0.00	2.48

532

533

# 535 Figures



536

Fig. 1.Soil profiles (0-100 cm) of both soil total OC and labile OC pool concentrations [C%], at the summit and
footslope positions. Error bars indicate 1 standard deviation (n≥3).



540 Fig. 2. Soil porosity profiles at the footslope (plain line) and at the summit (dashed line) positions. Error bars indicate



543 Fig. 3. Description of the probes used for CO2 concentration measurements inside the soil.



Fig. 4. Schematic description of the experimental plot (sampling design) at each slope position showing how temperature, VWC, CO<sub>2</sub> concentrations and CO<sub>2</sub> fluxes probes collocate with each others. Probes have been inserted at different locations both vertically and horizontally. Consequently, all of them are not in the same plane (i.e. depth lines with axes labels on the right hand-side illustrate the foreground profile and depth lines with axes labels on the left hand-side illustrate the background profile).





Fig. 5. Space-time dynamic of soil temperature (A) and moisture (B) at the summit (red) and the footslope (black)
position in 2013: (a) time series at different depths; (b) Profile at different dates.







Fig. 6. Space-time dynamic of soil CO<sub>2</sub> fluxes (A) concentrations (B) and diffusivity (C), at the summit (red) and the
footslope (black) position in 2013: (a) time series at different depths; (b) Profile at different dates.





Fig. 7. Depth distribution of the relative contribution to soilsurface  $CO_2$  fluxes in year 2013 averaged by semi-seasons (error bars represent the standard deviation of the time aggregation for each soil layer): (a) at the summit, and (b) at the footslope position.