



# Impact of thermal convection on CO<sub>2</sub> flux across the earth–atmosphere boundary in high–permeability soils



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## ABSTRACT

Quantifying earth–atmosphere gas exchange is a challenging, yet important problem that is made more complicated by the large number of mechanisms that contribute to this process. This work investigates one mechanism controlling non-diffusive gas transport from high-permeability media that is driven by natural diurnal thermal gradients in the upper vadose zone. We quantified CO<sub>2</sub> migration through 1-m long columns packed with two different permeability values: sand and large soil aggregates – both dry to eliminate chemical reactions. The bottom ends of the columns were exposed to 2000 ppm CO<sub>2</sub>-enriched air and the CO<sub>2</sub> concentration profiles along the columns was continually monitored. The columns were exposed to two different thermal regimes: isothermal conditions and a range of typical nighttime thermal gradients that are known to lead to unstable gas density profiles. Under isothermal conditions, and regardless of the matrix air-permeability, diffusion was the major mechanism for surface–atmosphere gas exchange. Under nighttime conditions, the prevailing mechanism depended upon matrix air-permeability: diffusion controlled CO<sub>2</sub> transport in the low permeability matrix, whereas thermal convection dominated transport in the high permeability matrix. Venting by thermal convection caused a CO<sub>2</sub> flux of up to two orders of magnitude higher than the diffusive flux. Such a mechanism may be implicated in a number of environmental settings. In soil, thermally driven convection can contribute to soil aeration influencing root respiration and microbial activity, and is likely one of the mechanisms associated with rapid CO<sub>2</sub> exchange that is commonly noted to follow tillage. With respect to the global CO<sub>2</sub> output, thermal convective venting is shown to be a permeability-limited mechanism with high gas exchange potential and a continuous diurnal presence. Its characteristic spatial scale could include, geologic sources via fractured rock surfaces, soil cracks, mine tailings, and rock-fill embankments.

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## 1. Introduction

Global climate stability and change depends strongly upon the radiative potential of atmospheric gases. Water vapor, CO<sub>2</sub>, O<sub>3</sub>, CH<sub>4</sub> and N<sub>2</sub>O are the major greenhouse gases that increase radiative potential, with CO<sub>2</sub> considered by mass to be the dominant component (~80%) of greenhouse gases (Johnson et al., 2007). Moreover, soil holds the largest stores of carbon in the planet, even exceeding the amount stored in both the atmosphere and biomass. For this reason, there is a strong research focus on quantifying net exchange of CO<sub>2</sub> between soil and atmosphere, and on identifying the many processes and mechanisms that control this exchange. Processes that control transfer of CO<sub>2</sub> and other relevant gases have time

constants of hours, days, seasons, years and decades, and exhibit a range of spatial scales ranging from leaf to landscape (Baldochi and Meyers, 1998). This span of temporal and spatial scales can create sampling and modeling challenges, nonetheless, understanding each is important since collectively these mechanisms comprise our planet's gas regulation process. This paper brings forth the mechanistic details of a specific non-diffusive, density-driven, gas exchange process that acts at the pedon scale, which is here shown to significantly increase the reach and rate of atmospheric exchange in settings where the media permeability is high. The goal of the paper is to quantify the parameters that control when and where this mechanism participates in gas exchange so that modelers and those making measurements in the field can properly take its impact into account.

Measurements of gas exchange between soil and atmosphere are strongly driven by two main concerns: the need to understand soil microbial function and soil organic matter turnover (e.g., Buyanovsky and Wagner, 1983; Singh and Gupta, 1977; Witkamp, 1966), and the desire to quantify its contribution to the global

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carbon cycle (e.g., Grunzweig et al., 2003; Hanson et al., 2000; Hirano et al., 2003; Norman et al., 1997; Schlesinger and Andrews, 2000; Tang et al., 2003; Turcu et al., 2005). There is a natural link between soil function and climate function since after photosynthesis, CO<sub>2</sub> flux from the soil is the second largest carbon flux in most ecosystems, producing more than 11 times the current rate of industrial CO<sub>2</sub> emissions generated by fossil-fuel combustion (Kuzyakov and Larionovaz, 2005). Thus, since soil can be a potential source as well as a potential sink of atmospheric CO<sub>2</sub>, numerous methods have evolved to quantify soil respiration. Most of the methods associated with soil respiration studies focus on diffusive exchange. Here, we explore the physics of non-diffusive CO<sub>2</sub> flux resulting from the daily atmospheric temperature cycle. We verify the existence of this flux and its magnitude and potential importance under conditions of daily thermal cycles. The mechanism discussed in this paper is driven by convective motion, a far more effective transport mechanism for soil gas and water vapor.

### 1.1. Background

Two mechanisms control gas exchange between soil and atmosphere: (1) diffusion, where transfer of any component of a gas mixture is controlled by its concentration gradient and the matrix diffusion coefficient (Rolston and Møldrup, 2012), and (2) convection, where transfer of any component is carried by mass flow of the whole gas mixture driven by the total gas pressure gradient and media permeability (Scanlon et al., 2001).

Traditionally, diffusion is considered the prevailing mechanism for gas migration in the subsurface (Hillel, 1998; Rolston and Møldrup, 2012), and many studies incorporating gas emission from soils typically make this assumption (e.g., Fang and Moncrieff, 1999; Pumpanen et al., 2003; Šimůnek and Suarez, 1993). However, bias in measurements of soil gas flux can be caused when non-diffusive transport is involved (Liang et al., 2004; Livingston and Hutchinson, 1995).

Convection that can significantly enhance gas migration in soils can be triggered by the following mechanisms: (1) temporal fluctuations in barometric pressure (e.g., Clements and Wilkening, 1974; Elberling et al., 1998; Massmann and Farrier, 1992; Weeks, 2001); (2) winds (Hanson et al., 1993; Kimball and Lemon, 1971; Nachshon et al., 2012; Poulsen and Møldrup, 2006); (3) water infiltration that induces an air pressure wave ahead of the wetting front (Navarro et al., 2008; Renault et al., 1998; Vachaud et al., 1974); and (4) temperature gradients that result in natural convection (Kamai et al., 2009; Nachshon et al., 2008; Rose and Guo, 1995; Schubert and Schulz, 2002; Sturm, 1991; Weisbrod et al., 2009; Weisbrod and Dragila, 2006; Witkamp, 1969).

Advective CO<sub>2</sub> gas transport in soils has been reported in several studies. Witkamp (1969) measured temperature and [CO<sub>2</sub>] in the floor of an oak stand and on a heathland, and found inverse correlation between temperature and [CO<sub>2</sub>] during midnight to dawn, which was explained by thermal convection. Lewicki et al. (2003) measured CO<sub>2</sub> fluxes and wind velocities over a fractured soil, combining [CO<sub>2</sub>] and stable carbon isotope sampling at depths of 10–80 cm. The authors suggested that wind-driven atmospheric air flow affects [CO<sub>2</sub>] in soil and enhances the magnitude and variability of surface CO<sub>2</sub> flux. Takle et al. (2004) measured CO<sub>2</sub> fluxes and wind velocities, together with [CO<sub>2</sub>] and differential pressure measurements in soil at depths of 10, 25 and 50 cm over a bare field. They found that pressure fluctuations penetrate 50 cm into the soil with little attenuation, providing a mechanism for advective mass transfer of gases throughout the porous media. Their mean CO<sub>2</sub> flux results yielded values that were about 5–10 times larger than the soil flux estimated by Fickian diffusion; however, their linear interpolation led to three- to sevenfold larger values for zero pressure pumping ( $\gamma$ -intercept). Takle et al. (2004) suggested various

mechanisms that could supply this additional enhancement, such as thermal expansion of soil air or flushing by evaporating water. Massman and Frank (2006) showed that [CO<sub>2</sub>] variations in 1-m deep snowpack are driven by pressure-induced advective flows created by wind interactions with local terrain irregularities such as mountain peaks and snowdrifts. Reicosky et al. (2008) measured a rapid decline of [CO<sub>2</sub>] at a depth of 30 cm in a plowed organic soil plot, which was explained by the increase in soil-air porosity due to tillage, combined with wind-induced gas exchange. These studies emphasize the importance of advective transport mechanisms, which are expected to be especially prevalent in highly permeable media.

Here, we focus our investigation on one density-driven advective transport mechanism that is generated by diurnal thermal gradients in the upper vadose zone, hereto called thermal convective venting (TCV). CO<sub>2</sub> gas flux rates were quantified for two porosity values spanning the range common to natural and agricultural systems by using large columns packed with either large soil aggregates taken from a tilled agricultural field or sand. Experiments were carried out in a customized climate-controlled laboratory (CCL), which enabled flux measurements under controlled isothermal (ISO) and temperature-gradient (TG) conditions with no wind.

It should be emphasized that the focus of this work was to identify and quantify the mechanisms controlling the CO<sub>2</sub> flux under controlled thermal cycles rather than exploring various chemical reactions and CO<sub>2</sub> production mechanisms. Therefore, the matrices used were air dried, the experimental duration was short and CO<sub>2</sub> source was constant.

## 2. Theory

The energy source for the mechanism of interest is the natural thermal profile of the upper vadose zone, which exhibits diurnal and annual variability driven by solar insolation and atmospheric temperatures. The relatively low thermal conductivity of soils causes retardation of the diurnal thermal signal with depth (Carson and Moses, 1963; Evett, 2001). At night, this typically creates unstable thermal conditions for soil air within the soil profile (i.e., warm, less dense gas underlying cold, denser gas), while stable thermal conditions persist during the day (Fig. 1).

During the night, if the buoyancy forces created by unstable conditions overcome the impeding viscous forces, convective gas motion within the soil profile will proceed. The onset of TCV in homogeneous porous media for conditions where the bottom temperature is higher than the top temperature is described by the Rayleigh–Darcy number ( $Ra$ ), which compares buoyant and viscous forces (Nield and Bejan, 1999):

$$Ra = \frac{g\beta\Delta T k H}{\nu\alpha_s} \quad (1)$$

where  $g$  is the gravitational acceleration,  $\beta$  is the volumetric thermal expansion coefficient,  $\Delta T$  is the temperature difference between the bottom and top of the soil layer,  $k$  is the permeability,  $H$  is the soil layer depth,  $\nu$  is the kinematic viscosity of the fluid and  $\alpha_s$  is the thermal diffusivity of the soil.  $\alpha_s = \kappa_s \kappa_f^{-1} \alpha_f$  where  $\kappa_s$  and  $\kappa_f$  are the thermal conductivities of the soil and the fluid, respectively, and  $\alpha_f$  is the thermal diffusivity of the fluid. Here it is assumed that the soil is homogeneous and isotropic, that Darcy's law and the Boussinesq approximation are valid (Nield and Bejan, 1999), and that air is the only mobile fluid (i.e., dry soil). In this situation, the minimal  $Ra$  value that needs to be exceeded for the onset of TCV is the critical value  $Ra_c \approx 40$  (Nield, 1982). Taking  $\beta$ ,  $\nu$  and  $\alpha_s$  as constants (that are evaluated at some suitable reference temperature, such as at the bottom of the soil layer), leaves  $\Delta T$ ,  $k$ , and  $H$  as the physical parameters that control the value of  $Ra$

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