1 **Free and forced gas convection in highly permeable, dry porous media**

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Abstract

The spatial and temporal distribution of gas species within the vadose zone is determined by biochemical sources and by transport mechanisms. Here, two mechanisms that can transfer gas at high rates across the earth-atmosphere interface are studied. The first is thermal convection venting (TCV), a free convection process that develops under conditions of sufficient temperature and density gradients. The second mechanism is wind-induced convection (WIC), an outcome of atmospheric surface winds that drive air movement within the porous media by a forced-convection process. Both of these advective mechanisms can dominate gas transport in high permeability porous media, and the objective of this study was to determine the permeability values that are relevant for these mechanisms to become significant for gas transport. Experiments were performed using large columns filled with four different single-sized spherical particles of 1 to 4 cm in diameter. The experiments were conducted in a climate-controlled laboratory, where surface winds and temperature gradients were imposed and monitored. A tracer gas of CO₂-enriched air was used to quantify the impact of TCV and WIC on gas exchange between the porous media and the atmosphere. A permeability range of 10^{-7} to 10^{-6} m² was found to be sufficient for the onset of TCV when imposed temperature gradients were similar to standard nighttime atmospheric conditions, leading to full or partial venting of the column. Surface wind with a velocity of 1.5 m s⁻¹ drove WIC to a depth of 0.3 m in most experimental conditions. The impact of WIC on net gas transport was not observed at the bottom-most sensor (0.9 m), except under conditions of very high permeability (2.4×10^{-6} m²) and a large temperature difference (6.5 °C m⁻¹), when both TCV and WIC worked simultaneously. Results confirm that TCV and WIC can significantly contribute to gas transport through porous media with sufficiently high permeability.

33	Keywords:
34	Gas transport
35	Thermal convection
36	Wind-induced convection
37	High-permeability porous media
38	Soil aeration
39	

1. Introduction

Gas movement within the earth's subsurface and its exchange with the atmosphere is one of the principal processes contributing to soil, ecosystem and atmospheric changes. Gas transport is controlled by diffusive and advective mechanisms (e.g., Kuang et al., 2013 and references therein), and its fate is impacted by biological, chemical, and physical processes. Vadose zone source gases moving across the earth-atmosphere interface are an important source of greenhouse gas emission. Significant emissions are feasible from terrestrial soils (Blagodatsky and Smith, 2012; Maier and Schack-Kirchner, 2014), Landfills (Allaire et al., 2008) and other environmental settings (Kang et al., 2014). Consequently, to improve our understanding of the abovementioned processes and their impacts on the environment, we need to quantify the mechanisms that control gas transport at the earth-atmosphere interface. This study quantifies how gas exchange is controlled by conditions imposed by the physical properties of the upper vadose zone and atmospheric temperature (Blagodatsky and Smith, 2012; Weisbrod et al., 2009).

Diffusion occurs across all permeable interfaces (Allaire et al., 2008) and historically has been considered the dominant or even the only significant process for gas transport in low-permeability media (Moldrup et al., 2004). With increasing permeability, transport by advection may increase gas exchange rates (You and Zhan, 2013) and even dominate the process of net gas flux across the earth-atmosphere interface (Massman et al., 1997; Weisbrod and Dragila, 2006). Several mechanisms can drive advective gas transport at this interface: (1) thermal convection venting (TCV), a type of free convection, which may develop in response to density gradients. Density gradients may result from either temperature gradients in the subsurface or a temperature contrast at the interface between atmospheric and porous media air (e.g., Ganot et al., 2014; Kamai et al., 2009; Lahmira et al., 2014a; Nachshon et al., 2008; Nield and Kuznetsov, 2013);

(2) forced wind-induced convection (WIC), which develops from surface winds that create pressure differences that drive convective air circulation (e.g., Amos et al., 2009; Nachshon et al., 2012; Reicosky et al., 2008; Turner and Pinshow, 2015); and (3) barometric pumping, which is the inward and outward motion of subsurface air due to cycles in atmospheric pressure (Neeper, 2002; Rossabi and Falta, 2002; Tillman and Smith, 2005; You and Zhan, 2013).

The effectiveness of the various gas transport mechanisms are determined by media properties, gas properties, and atmospheric conditions. The most significant porous media properties that affect gas transport are porosity, permeability, and water content (Abu-El-Sha'r and Abriola, 1997; Choi and Smith, 2005). Gas transport is also affected by the gas properties, namely density, viscosity and solubility (Hillel, 1998). In the upper part of the vadose zone, atmospheric conditions of wind, temperature and barometric pressure control the boundary condition and the energy gradient driving gas transport (Poulsen and Moldrup, 2006). In this work, we focused on the effect of the media's permeability and the temperature gradient on the development and magnitude of TCV. The potential contribution of WIC to gas convection within the media was also studied.

1.1. Thermal convection venting: free convection

Unstable gas density gradients of sufficient magnitude may drive free (natural) convection within the porous media or across the porous media-atmosphere interface. A special case of convective instability is driven by temperature gradients (TCV) (Lebeau and Konrad, 2009). The criteria for the onset of TCV is typically defined by the Rayleigh-Darcy number (Ra), a dimensionless number that relates convection to the ratio between gravitational (buoyancy) and dissipative potentials (viscosity and thermal diffusion) for a given fluid and a given media

permeability. The Ra number for a nonlinear temperature gradient is given as (Nield and Bejan, 2006; Tan et al., 2003):

$$Ra = \frac{g\beta k H^2}{\nu \alpha_s} \frac{dT}{dZ} \quad (1)$$

where g [$m\ s^{-2}$] is the gravitational acceleration, β [K^{-1}] is the volumetric thermal expansion coefficient of the fluid, k [m^2] is the permeability of the porous media, H [m] is the characteristic length of the porous media defined here as the distance from the impermeable boundary, ν [$m^2\ s^{-1}$] is the kinematic viscosity of the fluid, α_s [$m^2\ s^{-1}$] is the thermal diffusivity of the bulk porous media, T [K] is the temperature, and Z [m] is the vertical spatial coordinate.

Numerical studies have explored the value of the critical Ra ($Ra_{critical}$), which is defined as the threshold value for the onset of TCV. A value of $Ra_{critical} = 27.1$ is typically used as a threshold for the onset of convection in porous media, under a nonlinear temperature gradient, as is the case in natural soils, and assuming homogeneous and isotropic permeability (Tan and Sam, 1999; Tan et al., 2003).

A large number of analytical methods have been suggested for estimating the permeability of the media. For the case of coarse media composed of uniform spherical particles, the Kozeny-Carman equation is commonly used (Bear, 1972):

$$k = \frac{d_p^2}{180} \frac{f^3}{(1-f)^2} \quad (2)$$

where d_p [m] is the particle diameter, and f is the media's air-filled porosity. Substituting **Eq. 2** for **Eq. 1** yields:

$$Ra = \frac{g\beta H^2}{\nu \alpha_s} \frac{d_p^2 f^3}{180(1-f)^2} \frac{dT}{dZ} \quad (3)$$

which implies that $Ra \propto d_p^2$ and $Ra \propto dT/dZ$.

Only a few studies have explored TCV in porous media under natural atmospheric conditions. These studies include several applications, including: waste rock dump (Lahmira et al., 2014b) and natural aggregates from an agricultural field (Ganot et al., 2014). To the best of our knowledge, a general relationship between TCV and either permeability or particle size (PS) has not been studied. We also note that in some studies, Ra was calculated using an average linearized dT/dZ value to represent the entire system (e.g., Ganot et al., 2014). In this study, we use a continuous function to represent the temperature gradient (dT/dZ) for the nonlinear temperature profile of the media, which is more representative of natural soil settings (Hillel, 1998; Holmes et al., 2008).

1.2. Wind-induced convection: forced convection

Surface wind (SW) can drive gas transport within the porous media and across its interface with the atmosphere via two mechanisms: (1) Wind blowing over an irregular interface will drive turbulent eddies that penetrate the subsurface zone (Brickner-Braun et al., 2014; Hillel, 1998). This mechanism is effective for high-permeability media corresponding to particle diameters greater than 1-2 cm (Ishihara et al., 1992). (2) High-frequency pressure changes at the interface cause a pressure gradient between the interface and the porous media (Nachshon et al., 2012; Poulsen and Moldrup, 2006). Even a small pressure deficit of 1 Pa m^{-1} at the earth-atmosphere interface can drive a significant advective gas flux (Amos et al., 2009; Kimball and Lemon, 1972).

In general, relatively few studies have explored WIC gas transport within porous media and across the earth-atmosphere interface. Ishihara et al. (1992) investigated water transfer beneath bare soil driven by WIC on a laboratory scale. Riley et al. (1999) studied subsurface radon transport due to time-varying winds. Takle et al. (2004) examined the possible role of WIC on the efflux of CO₂ from the earth's subsurface. Poulsen and Moldrup (2006) and Poulsen and Sharma (2011) investigated WIC effects on earth-atmosphere gas exchange for porous media, focusing on landfills. Because gas fluxes in the shallow subsurface resulting from WIC can be of up to one order of magnitude higher than the fluxes originating from diffusion (Takle et al., 2004), further research to understand this mechanism is warranted, particularly regarding the manner in which permeability mediates the effectiveness of WIC.

1.3. Conditions leading to the onset of free and forced convection in natural systems

To illustrate the potential likelihood that natural atmospheric conditions may lead to TCV and WIC, temperature and wind data are presented from the Negev Desert, Israel, for a typical summer (**Fig. 1a**) and winter day (**Fig. 1b**). Significant temperature differences (TDs) between the atmosphere and the earth's subsurface at a depth of 1 m occur during the second part of the night. Maximum TDs > 10 °C and 7 °C are evident in the summer and winter data, respectively. Depending on the media permeability, these differences can potentially activate TCV. Higher maximum TD values are typically expected in winter since the vadose zone is warmer than the mean ambient temperature, and arid climates, like the Negev Desert, usually experience a higher diurnal temperature range in the winter than in the summer (Dai et al., 1999; Ganot et al., 2014). We emphasize that the TD values in **Fig. 1** only indicate potential conditions for TCV. The values in **Fig. 1** were calculated from temperature data at only two depths as described above. In order to define the location and magnitude of TCV, data of the temperature profile is needed

because natural profiles are nonlinear. While TCV is likely to occur at night, SW conditions suitable for WIC are evident on both summer and winter days during the afternoon. The wind velocity measured at an elevation of 3.5 m above ground exceeded 6 m s^{-1} in summer and 3 m s^{-1} in winter (the blue zones in **Fig. 1a** and **Fig. 1b**). Using **Eq. 2** from Nachshon et al. (2012), it can be calculated that the maximum observed wind of 6 m s^{-1} at an elevation of 3.5 m is equal to 1.5 m s^{-1} at 0.1 m above the surface.

Subsurface gas transport by convection is still an elusive research topic in soil physics and subsurface transport literature. Most studies to date explored the onset of convection numerically or by conducting experiments in ideal enclosures surrounded by impermeable walls (e.g., Laguerre et al., 2008; Tan et al., 2003). In addition, most studies related to TCV were performed with water as the convective fluid in conditions of low to moderate permeability values, lacking an investigation of gas transport in high permeability media (e.g., Nield and Kuznetsov, 2013 and references therein). Moreover, we could not find any published data regarding the combined impact of TCV and WIC on the overall gas transport across the earth-atmosphere interface.

Here, we report on the relationships between the porous media permeability, Ra values, and the development and magnitude of TCV and WIC. The reported data were obtained from experiments that were conducted with large-scale columns, packed with idealized media containing spherical particles. The experiments were conducted under controlled conditions, by artificially forcing temperature and wind conditions.

2. Materials and Methods

Experiments were performed in vertical polyvinyl chloride columns, filled with spherical particles. The columns were located in a climate-controlled laboratory, in which the ambient

atmospheric (top) boundary condition for temperature and surface wind was controlled and monitored continuously. The bottom boundary condition was controlled for temperature and carbon dioxide concentration [CO₂].

2.1. Climate-controlled laboratory and configuration of columns

All experiments were performed under a controlled ambient air temperature of 11 ± 0.5 °C (see Nachshon et al., 2011 and Neriah et al., 2014 for additional technical details). The columns were 1.40 m in height and 0.3 m in diameter, and were subdivided into two sections: the top part was 1.05 m tall, filled with porous media, and the second part that was at the bottom was a 0.35 m tall gas chamber (**Fig. 2a**), separated by a metal grid. Inside the gas chamber, an electric heater (100W, Satec, Israel) maintained the desired temperature at the bottom of the column. Temperature was monitored and controlled by thermocouples (type T, Omega Engineering, UK). Data from all sensors were logged (CR1000, Campbell Scientific, UT, USA) at 1-min intervals. Air enriched with 2000 ± 40 ppm CO₂ was chosen as the tracer gas. To maintain a constant tracer concentration within the gas chamber, an enriched air tank was connected to the gas chamber (**Fig. 2a**) via a flow control valve and inlet pipe to the gas chamber (without additional outlets). A continuous flow of 0.5 ± 0.05 L min⁻¹ was maintained and monitored using a flowmeter (MR3000, Key Instruments, PA, USA). Two differential pressure sensors (1MBAR-D-4V, All Sensors, CA, USA) were installed at the inlet pipe and in the gas chamber just below the upper section of the column (**Fig. 2a**). These sensors were used to confirm that the pressure that built up between the gas chamber and the above porous media did not exceed 1 Pa (accuracy limit). Additional sensors included: (1) 20 thermocouples inside the column in groups of five at four depths of 0.1, 0.4, 0.7, and 1 m below the column's top surface; (2) a relative humidity sensor (O2H-9903SD, Lutron, PA, USA) outside the column obtained the ambient relative

humidity; (3) a barometric pressure sensor (MPX2100AP, Freescale Semiconductors, TX, USA); and (4) two CO₂ sensors (IRGAs GMD-20, Vaisala, Finland) within the column at depths of 0.3 and 0.9 m below the column's top surface, one inside the gas chamber, and one for measuring the ambient concentration inside the laboratory. Further discussion on the IRGAs CO₂ sensor can be found at Turcu et al. (2005). The experimental setup is depicted in **Fig. 2**, which shows the setting of the columns and sensors (**Fig. 2a**) and the overall laboratory (**Fig. 2b**).

2.2. *Porous media*

Polystyrene spherical particles with diameters of 1, 2, 3 and 4 cm were used as the porous media; each column contained particles of a single size. These non-compressible spheres were chosen because of their ideal spherical shape that allows accurate permeability calculations, and because they have minimal influence on the gas, as they are impermeable and are made from non-reactive materials. Each column was packed using the same packing method of continuous filling while gently shaking the column. The porosity of each column was calculated as the ratio of the total sphere volume to the overall column volume (Hillel, 1998). The permeability values for the columns, calculated using **Eq. 2**, are 1.6×10^{-7} , 6.5×10^{-7} , 1.3×10^{-6} , and 2.4×10^{-6} m² for the PS diameters of 1, 2, 3, and 4 cm, respectively.

2.3. *Tracer gas*

The tracer gas for the experiment was air enriched with 2000 ppm CO₂ (Maxima, Israel). This concentration was chosen as it has no significant impact on the air density (Kowalski and Sánchez-Cañete, 2010). Additionally, because the experiments were conducted in dry media, the high solubility of CO₂ in water had no impact on its mobility in the gas phase.

2.4. *Wind tunnel*

A wind tunnel with a constant wind velocity was used for simulating natural SW conditions corresponding to those measured by a meteorological station in the Negev Desert (**Fig. 1**, blue lines). The tunnel size and cross-sectional area were customized to our columns following Ishihara et al. (1992). The tunnel was constructed from a standard air ventilation fan connected to a $0.5 \times 0.4 \times 0.4$ m wood tunnel. The tunnel outlet included a grid of 81 metal pipes, each 0.038 m in diameter and 0.18 m in length. The wind was measured at different heights above the column, at a distance of 0.2 m from the outlet, using a handheld device (C400, Lufft, Germany). An average wind velocity of 1.5 m s^{-1} was sustained 0.1 m above the column in all experiments where wind was applied.

2.5. *Experimental procedure*

Experiments were designed to test gas transport for four media properties (i.e., sphere diameters) under six different conditions that varied in either TD or SW: (1) isothermal conditions (TD0), with no SW (control condition); (2) TD1 and no SW; (3) TD2 and no SW; (4) TD0 and SW; (5) TD1 and SW; and (6) TD2 and SW. Each experimental condition was performed on all four PSs, with three repetitions, such that overall, 72 experiments were conducted. The TD conditions imposed were: TD0 = $0 \pm 0.5^\circ$, TD1 = $3.5 \pm 0.5^\circ$, and TD2 = $6.5 \pm 0.5^\circ$ °C. The TD values represent the difference between the temperature measured at the bottom of the porous media and that of the ambient air measured above the column. The temperature differences were imposed by heating the bottom of the column using an electric heater inside the gas chamber that was automated to maintain the desired temperature, except for TD0. A warmer temperature at the bottom was imposed in order to mimic nighttime soil conditions with cold atmospheric air above warm soil air (Holmes et al., 2008). Experiments

with imposed SW applied a constant wind of 1.5 m s^{-1} 0.1 m above the top of the column surface.

2.6. *Tracer gas analysis: relative concentration calculation*

The extent of the convective mechanism in the mixing of air between the atmosphere and the media was quantified by comparing the $[\text{CO}_2]$ at a depth of 0.9 m in the media to the ambient concentration. To separate the contribution of convection from background fluxes resulting from diffusion and the small measured advective component resulting from the inlet flow rate (0.5 L min^{-1}), we normalized each value to the value obtained during the isothermal experiment runs. The extent of venting is thus represented by a relative concentration parameter, calculated as:

$$\text{Relative Concentration} = \frac{C_{0.9} - C_{\text{atm}}}{C_{\text{TD0}} - C_{\text{atm}}} \quad (4)$$

where $C_{0.9}$ [ppm] is the $[\text{CO}_2]$ under convective venting conditions, C_{TD0} [ppm] is the average $[\text{CO}_2]$ under isothermal (TD0), steady-state conditions, both at a depth of 0.9 m for the same PS, and C_{atm} [ppm] = 365 ppm is the $[\text{CO}_2]$ of the laboratory room. Steady-state conditions were defined as less than a 1% change in $[\text{CO}_2]$ in all sensors over at least 10 minutes. It is expected that the relative concentration will equal unity when there is no convective venting, and zero under full convective venting.

3. **Results and Discussion**

Porosity values for all columns were between 0.44 and 0.45 (**Table 1**, column A), close to the ideal 0.40 value for cubic tetrahedral packing (Hillel, 1998).

3.1. *Air movement under isothermal conditions*

Under isothermal conditions ($TD = 0 \pm 0.5$ °C; **Table 1**, column C), we verified that convective motion did not occur within any of the columns (within measurement error). $[CO_2]$ values at the 0.9-m depth were close to the influx $[CO_2]$ of 2000 ppm (**Table 1**, column D), indicating minimal mixing. When wind was imposed, WIC venting reached approximately a 0.3-m depth in all columns except for $PS = 1$ cm, dropping the tracer concentration by 15 ± 5 %. The observed decrease in the $[CO_2]$ at a depth of 0.3 m agrees with previous studies. Takle et al. (2004) observed wind-driven high pressure fluctuations at a depth of 0.15 m. WIC at a depth of 0.3 m in tillage soils was reported by Reicosky et al. (2008), and in large aperture fractures, up to a depth of 0.5 m (Nachshon et al., 2012). Since wind tunnel may generate less turbulence than natural wind, WIC may be more pronounced under field conditions (Waddington et al., 1996) than observed here.

3.2. *Air movement under temperature differences*

When TD conditions were imposed between the bottom of each column and the laboratory air, TCV occurred in all columns except for $PS = 1$ cm (**Table 1**, column E). The magnitude of convective gas mixing increased with increasing PS and TD values, indicated by a decrease in $[CO_2]$ at 0.9 m. For example, for TD2, $[CO_2]$ at 0.9 m decreased by 10% and 71% for $PS = 2$ and $PS = 4$ cm, respectively. These results agree with the general behavior predicted by **Eq. 3**, which shows that TCV, which is amplified with the increase of the Ra number (Nield and Bejan, 2006), should be proportional to the temperature gradients and the permeability (square of the PS). TD values used in the experiment were chosen to be in the range of field data (**Fig. 1**, red zone) and reported in previous studies (e.g., Ganot et al., 2012; Ninari and Berliner, 2002; Smith et al., 2012). The imposed TD conditions resulted in nonlinear temperature gradients, with the gradient exponentially decaying away from the heat source (**Fig. 3**). The slight differences between the

temperature profiles for the same TD are the results of small variations in laboratory temperature and thermocouple locations between the different experiments.

A continuous temperature profile was obtained by fitting to the data an exponential temperature distribution function that decays from the heat source (the electric heater in the gas chamber at the bottom of the column),

$$T = X_1 + X_2 \exp^{[X_3(Z+1.05)]} \quad (5)$$

where T [°C] is the calculated temperature, Z [m] is the depth from the surface (negative downward), and X_i (with $i = 1, 2$, or 3) are fitting parameters. For each experimental condition, **Eq. 5** was fitted using a least squares optimization to the data from the three repetitions of each experiment, using temperature data measured at the six different depths (**Fig. 3**). Eight equations corresponding to four PS and two TD values were obtained from this fitting exercise, with $R^2 > 0.96$.

Because the Ra number provides a theoretical indicator for the onset of free convection and the occurrence and magnitude of TCV, continuous Ra values were evaluated from the temperature data. The derivative of **Eq. 5** (**Eq. 6**) was used to obtain a continuous dT/dZ function for calculating Ra values for each column (**Fig. 4**).

$$\frac{dT}{dZ} = X_3 X_2 \exp^{[X_3(Z+1.05)]} \quad (6)$$

Regions within the media are locally active with TCV when $Ra > Ra_{critical}$, and, in contrast, are not active when $Ra < Ra_{critical}$ corresponding to sections of stable density gradients. For our analysis, we used $Ra_{critical} = 27.1$, typical for a nonlinear temperature distribution (Tan and Sam, 1999; Tan et al., 2003). The section within the column that is active in TCV is quantified by the

convection cell length, calculated to be the distance from the bottom of the porous media (where it is warmest, and the Ra greatest) up to where the Ra line crosses $Ra_{critical}$ (**Fig. 4, Table 1**, column G). For each TD condition, the convection cell lengths increased with increasing PS value; and for each PS value, all cell lengths increased with the increasing TD. An average Ra for each convection cell (Ra_{avg} ; **Table 1**, column H) was calculated using nonlinear averaging of **Eq. 1** with the temperature values of the top and bottom boundaries of the convection cell as the integration boundaries. The convection cell length and Ra_{avg} characterize the TCV state, with the first describing the spatial extent of the convective mixing region and the second parameter describing the magnitude of gas mixing. Two mixing regions can have similar spatial distributions, but their magnitudes can be different, thus causing different gas concentration changes inside a similar cell length. The magnitude of gas mixing will always be greatest at the location of the greatest temperature gradient; in this experiment, the location is the end of the cell closest to the heat source, but under natural conditions, the depth of the maximum temperature gradient varies diurnally.

The relationship shown in **Fig. 5a** provides the link between experimental results and the theoretical prediction of convection with Ra . If Ra predicts the convective venting intensity and location, then increases in cell length will correlate to decreasing gas concentration at the 0.9-m-depth sensor. Indeed, our results show a linear relationship ($R^2 > 0.91$) between the convection cell length and the relative $[CO_2]$ (**Fig. 5a**) measured by the 0.9-m sensor. Results in **Fig. 5a** demonstrate that internal cells driven by temperature gradients within the porous media can control gas exchange with the atmosphere, as evidenced by the decrease in tracer gas concentrations.

While the sensor at 0.9 m is an indication of net loss, the mixing caused by convection within the porous media is evidenced by the decrease of the tracer concentrations in the profile. A progressive decrease in tracer concentrations along the profile between the 0.3- and 0.9-m sensors was more pronounced with increase in PS and TD (**Fig. 5b**). For PS = 4 and TD2, the convection cell reached up to the 0.3-m sensor; consequently, the gas was fully mixed and the concentration profile was flat. In all other cases, the cell did not reach the 0.3-m sensor, but its effect was evident in the reduced concentration differences between these two sensors. Ganot et al. (2014) also observed flat concentration gradients when exploring convection in a column packed with large aggregates collected from an agricultural field (estimated permeability of $2.4 \times 10^{-6} \text{ m}^2$, similar to the permeability for the PS = 4 cm) when a TD promoting TCV was applied. Under natural conditions, TCV acting within the root zone can accelerate the equilibration of soil gases with the atmosphere, subsequently increasing soil aeration.

In the scenarios explored in this study, the calculation of Ra values indicates that all the convection cells ended below the upper boundary, thus causing “closed inner cells” in the media. It is evident from the data that even internal cells increase the net gas flux by reducing the diffusion transport distance, thus increasing the concentration gradient driving diffusion (Rose and Guo, 1995). In the case of cells that reach the earth-atmosphere interface, atmospheric CO_2 values are expected throughout the media whenever the time constant of gas production is lower than the convective time constant. The extreme case of such conditions is an open fracture system (Kamai et al., 2009; Nachshon et al., 2008; Weisbrod and Dragila, 2006; Weisbrod et al., 2009).

While the effect of wind did not reach deep into the profile ($\sim 0.3 \text{ m}$), WIC mixing of the upper profile resulted in a reduction in the gas concentration at the 0.3-m sensor for all columns in

which $PS > 1$ cm. In the case of TD2 and $PS = 3$ and 4 cm, WIC mixing and the TCV convection cell linked to flatten the entire gas profile. These findings indicate the potential for a linkage between free and forced convection mechanisms in natural systems. Each mechanism has a region of influence; when these overlap, the two mechanisms could synergize into a more complete and effective venting process. In high-permeability media under field conditions, the diurnal variability of the media's thermal gradient could lead to regular, short-lived, but effective, media gas-flushing events.

3.3. *Impact of natural soil temperature profiles on soil gas convection: theoretical calculations*

The same analysis to determine location and extent of internal TCV cells which was used for the laboratory columns is applied here to soil profile temperatures to investigate the extent of the impact of TCV on subsurface gas motion and gas efflux. The soil temperature profile resulting from downward conduction of a surface sinusoidal temperature wave is given by Hillel (1998):

$$T(Z, t) = T_{\text{avg}} + A_0 \left[\sin \left(\omega t + \frac{Z}{d} \right) \right] \exp^{\frac{Z}{d}} \quad (7)$$

where T_{avg} [K] is the average temperature of the soil surface, A_0 [K] is the daily amplitude of the surface temperature, ω [s^{-1}] is the radial diurnal frequency, t [s] is the time, Z [m] is the depth (negative downward), and d [m] is the damping depth defined as

$$d = \left(\frac{2\alpha_s}{\omega} \right)^{0.5} \quad (8)$$

where α_s [$m^2 s^{-1}$] is the thermal diffusivity of the bulk soil.

A continuous thermal gradient function was obtained by differentiating **Eq. 7**, and Ra values were calculated using **Eq. 1**, where H was equaled to the damping depth to represent the distance over which the temperature difference is significantly expressed. Incorporating a nonlinear temperature gradient elucidated an additional phenomenon not previously reported. As an example, two times of day are here analyzed, 04:00 AM and 08:00 AM. The temperature profile at 04:00 AM (**Fig. 6a1**) shows an unstable upper section of cold air above warmer air, starting at 11.3 °C at the soil-atmosphere interface and increasing with depth, reaching $Ra_{critical}$ (i.e., $Ra = 27.1$) at a depth of ~0.33 m (**Fig. 6a3**). During this time period, a convection cell stretching from the surface to a depth of 0.33 m is predicted. At 08:00 AM (**Fig. 6b1**), the upper 0.09 m has a stable gradient, but between a depth of 0.09 m and a depth of 0.46 m, the thermal gradient is unstable (**Fig. 6b2**). Ra values predict an internal convection cell in the middle section where cold resides above warm air, with the cell stretching from depth of 0.10 to 0.41 m (**Fig. 6b3**). In both cases, the convection cell was ~0.3 m long, but located very differently within the profile. A cell that reaches the soil-atmosphere interface (e.g., 04:00 AM) will enable direct advective gas transport between the soil and the atmosphere. Internal TCV cells will homogenize internal gas gradients and will shorten the distance over which diffusion controls transport, increasing net efflux. At 04:00 AM, the cell is already connected to the surface, and venting is presumably already faster than gas generation processes within the soil profile.

Our study emphasizes the need to quantify Ra as a continuous property for the entire porous media profile, rather than using temperature values from a few discrete points, as sparse discrete values can lead to erroneous TCV interpretations. For example, using **Figs. 6a3** and **6b3**, and calculating the Ra number based on a linear $\Delta T/\Delta Z$ between only two points ($Z = 0$ and 0.5 m) gives $\Delta T/\Delta Z$ equal to 17.2 and 0, and Ra equal to 308.3 and 0, at 04:00 and 08:00 AM,

respectively. These results will lead to the wrong conclusion that a full convection cell of a length of 0.5 m is initiated at 04:00 AM, and no cell at 08:00 AM as $Ra < Ra_{critical}$.

The experiments and theoretical analysis presented here investigated dry porous media conditions and homogeneous permeability. Under natural conditions, heterogeneous permeability, water content and vapor dynamics (evaporation and condensation) are expected to affect the convection process by changing the temperature profile and the air-filled porosity (Eqs. 2, 7 and 8), impacting the criteria leading to TCV onset. Our results show that for low permeability ($< 10^{-7} \text{ m}^2$) TCV is unlikely to occur under natural daily changes of temperature. In other words, in sandy or finer soils, unless there are large aperture cracks ($\sim 1 \text{ cm}$; Nachshon et al., 2008), TCV is unlikely to be developed. Also large pores (above capillary size) between aggregates tend to drain fast by gravity and therefore will be empty most of the time, facilitating convection.

4. Summary and conclusion

The main objective of this research was to establish relationships between permeability and convective venting under imposed (1) thermal gradients within the porous media; (2) surface wind; and (3) these conditions combined. Column experiments were conducted under controlled laboratory conditions, using columns filled with single-sized spherical particles.

For the temperature gradients imposed, results clearly show a permeability cutoff ($\sim 1.6 \times 10^{-7} \text{ m}^2$) below which thermal convection did not develop. Above this permeability cutoff, convective venting increased with permeability and temperature gradient. TCV caused significant gas mixing between the porous media and the ambient atmospheric air above. Relative $[\text{CO}_2]$ at a depth of 0.9 m decreased down to 29% during TCV for the largest particle size (4 cm) with an

imposed temperature difference of 6.5 °C, compared to the isothermal case. The permeability cutoff value identified by this study suggests that for sand size particles and below, TCV is an insignificant contributor to gas transport.

The addition of a 1.5 m s⁻¹ surface wind enhanced the circulation of atmospheric air to a depth of 0.3 m for particles greater than 1 cm. For the largest particle size (4 cm) and under the highest temperature difference (TD2 = 6.5 °C), the upper convective cell driven by WIC was linked with the lower TCV cell venting the entire column, lowering the concentration at the bottom of the column to 9%, the lowest value compared to all the other conditions.

By measuring tracer gas concentration and calculating *Ra* values from temperature data, it was possible to link between experimental data and the theoretical prediction of convection.

Observed changes in tracer gas concentration showed an excellent correlation to the convection cell length derived from the theoretical *Ra* values. This correlation supports the potential of the future prediction of TCV from measured soil temperature profiles and physical properties, mainly permeability and thermal parameters. This approach was tested using field soil temperature profiles. The nonlinear temperature gradient required that dT/dZ and the resulting *Ra* values should be analyzed as a continuous curve and not as discrete values along the profile. This analysis elucidated the existence of internal convection cells that develop and do not necessarily reach the earth-atmosphere interface. Such cells would cause internal mixing of soil gases within the convection cell with an effect on the net gas transport by shortening the distance over which diffusion is acting. Our analysis also demonstrates that only a small temperature data set is necessary to provide an accurate prediction of convective gas transport.

431 Different transport mechanisms affect gas movement across the earth-atmosphere interface.
432 Here, we evaluated the impact of free convection driven by temperature gradients, internal to the
433 porous media, on the enhancement of gas transport across the media-atmosphere interface. In
434 addition, we also tested the influence of wind-induced convection and the potential synergy
435 between the two mechanisms. The results of this study indicate that convection, even if only
436 existing internally within the media, can enhance gas transport significantly above the rates
437 driven by diffusion.

438

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Figure captions

Fig. 1. Soil temperature difference between the soil surface and 1 m below the surface, and surface wind measured 3.5 m above the land surface during a summer (a) and a winter (b) day. Data taken from the Ben-Gurion University Meteorological Station located at Sede Boquer. Red zones labeled TD represent time segments with unstable density gradients potentially leading to TCV, and blue zones represent wind conditions (SW) that could potentially lead to WIC according to similar wind velocities that activated WIC in fractures (Nachshon et al., 2012). Note that the maximum observed wind of 6 m s^{-1} at an elevation of 3.5 m corresponds to 1.5 m s^{-1} at 0.1 m above the surface (Nachshon et al., 2012)

Fig. 2. Experimental setting. (a) Scheme of the column showing sensor locations (modified from Ganot et al., 2014), (b) A photo of the overall system located within the climate-controlled laboratory.

Fig. 3. Continuous temperature profiles obtained by fitting an exponential function (Eq. 5) to each dataset, for different particle sizes (PSs) and temperature differences (TDs). (a) $\text{TD1} = 3.5 \pm 0.5 \text{ }^{\circ}\text{C}$, (b) $\text{TD2} = 6.5 \pm 0.5 \text{ }^{\circ}\text{C}$. $R^2 > 0.96$ for the entire dataset.

Fig. 4. Calculated Ra profiles (Eqs. 3, 5, and 6) for different media particle sizes (PSs) under different imposed temperature differences (TDs). (a) $\text{TD1} = 3.5 \pm 0.5 \text{ }^{\circ}\text{C}$, (b) $\text{TD2} = 6.5 \pm 0.5 \text{ }^{\circ}\text{C}$. Ra_{critical} equals 27.1 (Tan and Sam, 1999; Tan et al., 2003). TCV stage represents conditions that exceed the theoretical Ra_{critical} values for the onset of TCV. Reference depth represents the CO_2 sensor location in the porous media 0.9 m below the surface and 0.15 m above the lower boundary of the porous media with the gas chamber.

Fig. 5. Correlations between convection cell length and (a) relative concentration, (b) differences

between the $[\text{CO}_2]$ at 0.3-m and 0.9-m depths. Regression lines exclude data from experiments in which no TCV transport was observed: PS = 1 cm for TD1 and TD2; and PS = 2 cm for TD1. Convection cell lengths were based on Ra numbers presented in **Fig. 4**, and relative concentrations were calculated using **Eq. 4**. Error bars represent a 5% deviation from the average.

Fig. 6. Vertical distribution of three parameters in a theoretical soil profile, at two different times. (a) 04:00 AM, and (b) 08:00 AM. (a1, b1): Temperature profile calculated with **Eqs. 7** and **8**. For a1: $T_{\text{avg}} = 20^\circ\text{C}$, $A_0 = 10^\circ\text{C}$, $t = 72000 \text{ sec}$, $\alpha_s = 5 \times 10^{-7} \text{ m}^2 \text{ s}^{-1}$ corresponding to dry sand conditions (Al Nakshabandi and Kohnke, 1965). For b1: $t = 86400$. For a2 and b2: dT/dZ as a function of depth using the first derivative of **Eq. 7**. For a3 and b3: Ra number as a function of depth calculated by substituting dT/dZ into **Eq. 1**, $k = 1.6 \times 10^{-7} \text{ m}^2$ (same as the 1-cm PS experiments), $H = d$. Constants used for air at 20°C : $\beta = 3.4 \times 10^{-3} ^\circ\text{C}^{-1}$ and $\nu = 1.5 \times 10^{-5} \text{ m}^2 \text{ s}^{-1}$ (Incropera et al., 2007). TCV stage represents conditions of $Ra > Ra_{\text{critical}}$, indicating onset of TCV. The Ra value for positive (stable) temperature gradients is set to $Ra = 0$. Note that the Ra axis is in log scale.

633 (Figure 1)

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636 (Figure 2)

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638 (Figure 3)

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640 (Figure 4)

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642 (Figure 5)

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644 (Figure 6)

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646 **Table**

647 **Table 1.** Summarized results organized by particle size (PS)

A	B	C	D	E	F	G	H
Column properties ^a	Conditions	Temperature differences (TD) ^b [°C]	CO ₂ reading at -0.9 [ppm]	Relative concentration at -0.9 [ppm] ^c	CO ₂ reading at -0.3 [ppm]	Convection cell length [m] ^d	Ra_{avg}
1 cm PS $k = 1.6 \times 10^{-7} \text{ m}^2$ $f = 0.45$	TD0	-0.35±0.06	1909±4	1±0	1845±47	0	≈0
	TD1	3.26±0.15	1915±10	1±0	1840±3	0	≈0
	TD2	3.53±0.03	1919±5	1.01±0	1841±4	0.01	28.7
	SW	-0.12±0.07	1899±10	0.99±0	1827±6	N/A	N/A
	TD1 with SW	6.66±0.08	1906±2	1±0	1811±22	N/A	N/A
	TD2 with SW	6.63±0.16	1909±7	1±0	1804±33	N/A	N/A
2 cm PS $k = 6.5 \times 10^{-7} \text{ m}^2$ $f = 0.45$	TD0	-0.4±0.04	1955±2	1±0	1576±47	0	≈0
	TD1	3.2±0.10	1914±10	0.97±0.01	1570±51	0.09	32.3
	TD2	6.5±0.02	1800±32	0.90±0.02	1505±47	0.31	45.1
	SW	-0.4±0.04	1942±3	0.99±0	1284±45	N/A	N/A
	TD1 with SW	3.6±0.04	1920±6	0.98±0.01	1352±17	N/A	N/A
	TD2 with SW	6.4±0.06	1759±37	0.88±0.02	1268±70	N/A	N/A
3 cm PS $k = 1.3 \times 10^{-6} \text{ m}^2$ $f = 0.44$	TD0	-0.5±0.12	1872±20	1±0	1225±174	0	≈0
	TD1	3.8±0.03	1506±43	0.76±0.03	1201±39	0.34	60
	TD2	6.5±0.07	1353±21	0.66±0.01	1212±19	0.5	78.1
	SW	-0.2±0.02	1873±21	1±0.02	1006±49	N/A	N/A
	TD1 with SW	3.9±0.05	1643±7	0.85±0.01	1101±15	N/A	N/A
	TD2 with SW	6.7±0.03	1141±45	0.52±0.03	1011±47	N/A	N/A
4 cm PS $k = 2.4 \times 10^{-6} \text{ m}^2$ $f = 0.45$	TD0	-0.5±0.08	1655±13	1±0	978±57	0	≈0
	TD1	3.7±0.013	919±29	0.43±0.02	827±30	0.54	79.3
	TD2	6.2±0.12	735±24	0.29±0.02	751±22	0.73	96.2
	SW	-0.2±0.01	1766±26	1.09±0.03	887±49	N/A	N/A
	TD1 with SW	3.4±0.14	868±37	0.39±0.03	751±26	N/A	N/A
	TD2 with SW	6.3±0.71	484±35	0.09±0.03	594±38	N/A	N/A

648 Average and standard deviation values calculated from three experiment repetitions.^a

649 Permeability was calculated using **Eq. 2**.^b TDs between the bottom of the porous media in the

650 column and the air inside the laboratory.^c Relative concentration was calculated using **Eq. 4**.^d

651 Calculations in columns G and H are explained in section 3.2. N/A represents the conditions in

652 which Ra number calculations are not applicable due to an additional wind mechanism.