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Water transfer in soil at low water content. Is the local equilibrium assumption still appropriate?

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Abstract

The dynamics of water content in the superficial layers of soils is critical in the modelling of land-surface processes. In arid regions, vapour flux contributes significantly to the global water mass balance. To account for it in theoretical descriptions, most of the models proposed in the literature rely on the local equilibrium assumption that constrains the vapour pressure to remain at its equilibrium value. It implicitly amounts to consider an instantaneous phase change. Recent works underlined a retardation time and a decrease in phase change rate as the water content gets lower. Therefore, the objective is to revisit water transport modelling by rejecting the local equilibrium assumption. This requires developing a non-equilibrium model by taking into account the phase change kinetics. To assess the interest of this approach, a natural soil of Burkina-Faso has been experimentally characterized from independent tests and soil column experiments have been carried out. The comparison of experimental drying kinetics and water content pro-

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files with computational predictions confirms the reliability of this description. Liquid/gas non-equilibrium is significant in a limited subsurface zone which defines explicitly the transition from liquid transport in lower layers to vapour transport in upper layers, i.e., the evaporation front. The overall moisture dynamics is governed by the coupling between water transport mechanisms (liquid filtration, vapour diffusion, phase change) that mainly occurs in this transition zone.

Keywords: water transport, evaporation, non-equilibrium, experimental, modelling

1. Introduction

Land-atmosphere mass exchanges are concentrated in the superficial layers of soils and control most of biological processes required for plant growth. An accurate description of these layers is of first importance when developing realistic boundary conditions to be implemented in large-scale environmental models. Intensive research has been done to develop efficient numerical models of water transport in the vadose zone. In particular, the quantification of total water evaporation from soil is a crucial issue since it governs the water content dynamics near the surface (Gowing et al., 2006).

Following the pioneered work of Philip and de Vries (1957), most of models have associated the Richard's equation in the liquid phase with a classical diffusive equation in the gas phase while both transport phenomena are coupled through the heat equation (Bittelli et al., 2008; Garcia-Gonzalez et al., 2012; Grifoll et al., 2005; Novak, 2010; Parlange et al., 1998; Saito et al., 2006; Sakai et al., 1999; Thomas and Missoum, 1999; Xiang et al., 2012;

16 Yanful and Mousavi, 2003). Although thermal gradients affect liquid water
17 redistribution in soils, the most important coupling process is the transport
18 of latent heat by vapour flux. These models have been successfully used to
19 describe the land-atmosphere water and energy balance of various natural
20 field soils over a large period (Bittelli et al., 2008; Garcia-Gonzalez et al.,
21 2012; Grifoll et al., 2005; Novak, 2010; Saito et al., 2006).

22 Most of these models have been developed for temperate regions where
23 water content evolves in the capillary domain from saturation to the milting
24 point. Since water vapour flow in semi-arid and arid regions can represent a
25 major part of the overall water flow, it is important to take it into account
26 together with liquid water flow when evaluating hydrologic fluxes (Garcia-
27 Gonzalez et al., 2012; Sakai et al., 1999). In this framework, classical models
28 have been extrapolated towards low water contents without carefully check-
29 ing the validity of such formulations. For instance, usual descriptions of the
30 water retention curve can significantly diverge from experimental points at
31 very low water content (Thakur et al., 2006). Correction functions have been
32 proposed without being fully satisfactory. However, as water content goes
33 to very low values, liquid water takes the form of adsorbed layers onto solid
34 surface and does not behave as “free” water. It has been shown that film
35 flows replace conventional capillary flows at low water content, induces a
36 change in the relative permeability description (Tuller and Or, 2002). It is
37 therefore noteworthy to consider that modelling water transport phenomena
38 at low water content calls for a specific description.

39 In particular, the *local equilibrium assumption* is extensively used in most
40 of the theoretical models proposed in the literature (Bittelli et al., 2008;

41 Garcia-Gonzalez et al., 2012; Grifoll et al., 2005; Novak, 2010; Parlange et al.,
 42 1998; Saito et al., 2006; Sakai et al., 1999; Thomas and Missoum, 1999; Xi-
 43 ang et al., 2012; Yamanaka and Yonetani, 1999; Yanful and Mousavi, 2003).
 44 This hypothesis governs the liquid/gas mass exchange by assuming that the
 45 partial pressure of vapour remains equal to its equilibrium value. It is gen-
 46 erally written as an explicit relation between the relative humidity and the
 47 hydraulic head. It allows combining liquid and vapour mass balance equa-
 48 tions in a single one describing the global soil moisture content. Implicitly, it
 49 amounts to consider that the evaporation process is instantaneous in compar-
 50 ison with the other transport phenomena. This should be fairly satisfactory
 51 when capillary forces are predominant. Nevertheless, under particular condi-
 52 tions, a volatilization retardation time has been observed (Armstrong et al.,
 53 1994; Bénet and Jouanna, 1982; Chammari et al., 2008). In the hygroscopic
 54 domain where adsorption phenomena predominate, phase change kinetics is
 55 strongly influenced by the thermodynamic state of water and evaporation
 56 rate is drastically reduced (Bénet et al., 2009; Lozano et al., 2008). These
 57 experimental observations conduced us to reconsider the *local equilibrium*
 58 *assumption* in order to assess its reliability.

59 The aim of this contribution is to revisit water transport modelling in the
 60 lower range of water content. The prominence of hygroscopic effects leads
 61 us to reject the liquid/gas equilibrium assumption. This requires developing
 62 a two-equation model by taking into account the phase change kinetics. It
 63 means that, in some specific configurations, the characteristic times of the
 64 three transport mechanisms considered (liquid flow, vapour diffusion, liquid-
 65 gas phase change) are of the same order of magnitude. To fulfil this objective,

66 a natural soil of Burkina-Faso has been experimentally characterized from
67 independent tests and soil column experiments have been carried out. The
68 comparison of experimental drying kinetics and water content profiles with
69 computational predictions supports the validation of this description. There-
70 fore, the *local equilibrium assumption* can be discussed based on numerical
71 simulations. In particular, the location of the evaporation front is identified
72 as it propagates from the upper surface towards deeper layers.

73 **2. Theoretical and numerical modelling of water transfer**

74 *2.1. Liquid and vapour transfer model*

75 A natural soil can be idealized by a triphasic porous medium by consid-
76 ering a solid, a liquid and a gaseous phase, while the gaseous phase consists
77 of two components: dry air and water vapour. Theoretical modelling relies
78 on the following assumptions:

- 79 • Temperature is uniform and constant. This hypothesis will be validated
80 *a posteriori* in the last section based on numerical simulations.
- 81 • The solid skeleton is undeformable. Assuming a rigid structure is a
82 strong hypothesis since evaporation and dehydration processes in non-
83 consolidated soils generally lead to global shrinkage. However, we focus
84 our attention on the hygroscopic domain where these effects are negli-
85 gible.
- 86 • The total gas pressure is constant and uniform, since the convective
87 transport in the gas phase is negligible. Actually, it means that the gas
88 permeability is large enough to assume that a pressure gradient will

89 be instantaneously equilibrated when compared to the other transport
 90 phenomena.

Therefore, three elementary phenomena are considered: liquid filtration governed by capillary and gravity effects, vapour diffusion in the gas phase and liquid-gas phase change of water. Fundamental mass balance equations of water in liquid and gas phase are written:

$$\frac{\partial \rho_l}{\partial t} + \nabla \cdot (\rho_l \mathbf{v}_l) = -\hat{\rho}_v \quad (1)$$

$$\frac{\partial \rho_v}{\partial t} + \nabla \cdot \mathbf{J}_v = \hat{\rho}_v \quad (2)$$

91 where ρ_l and ρ_v (kg m^{-3}) are, respectively, the apparent density of liquid
 92 water and its vapour, $\mathbf{v}_l$ (m s^{-1}) is the intrinsic velocity of liquid water
 93 and $\mathbf{J}_v$ ($\text{kg m}^{-2} \text{s}^{-1}$) is the vapour diffusion flux. The phase-change rate $\hat{\rho}_v$
 94 ($\text{kg m}^{-3} \text{s}^{-1}$) is thus a volumetric scalar flux, representing the mass of water
 95 transforming from a liquid state to a vapour state by unit volume and unit
 96 time.

97 From an experimental point of view, appropriate state variables are the
 98 mass water content, $w(\%)$, defined as the ratio between the apparent mass
 99 densities of liquid and solid:

$$w = \frac{\rho_l}{\rho_s} \quad (3)$$

100 and the vapour partial pressure in the gas phase, $p_v(\text{Pa})$, linked to the ap-
 101 parent density of vapour, ρ_v , through the ideal gas law:

$$\phi_g p_v = \frac{RT}{M_w} \rho_v \quad (4)$$

102 where R (J kg^{-1}) is the ideal gas constant and M_w (kg) is the molar mass
 103 of water. The volume fraction of the gas phase, ϕ_g , is related to the water

104 content by:

$$\phi_g = \frac{V_g}{V} = 1 - \frac{\rho_s}{\rho_s^*} - w \frac{\rho_s}{\rho_l^*} \quad (5)$$

105 where ρ_s^* and ρ_l^* (kg m^{-3}) are, respectively, the real density of solid and liquid
106 phase.

107 2.2. Vapour diffusion

108 The vapour diffusion flux, $\mathbf{J}_v$, is classically described by a first order Fick's
109 law:

$$\mathbf{J}_v = -D_{vs} \nabla \rho_v^* \quad (6)$$

110 where D_{vs} is the effective vapour diffusion coefficient in the soil. It gen-
111 erally depends on the tortuosity as presented in next section dealing with
112 experimental characterization.

113 2.3. Liquid filtration

114 The liquid filtration flux appearing in Eq. 1 can be expressed by the
115 Darcy's law extended to the non-saturated case. The validity of such de-
116 scription toward very low water content is questionable since the concept of
117 liquid pressure is meaningless (Baker and Frydman, 2009; Low, 1961; Nitao
118 and Bear, 1996). An alternative proposed by Bénet et al. (2012) is to rely
119 on the chemical potential. Therefore, filtration transport is written:

$$\rho_l \mathbf{v}_l = -K (\nabla \mu_l - \mathbf{g}) \quad (7)$$

120 where μ_l (J kg^{-1}) is the mass chemical potential of liquid water, $\mathbf{g}$ (m s^{-2}) is
121 the gravity acceleration vector. The filtration coefficient, K , refers to the soil
122 effective conductivity by means of:

$$K = K_r K_{sat} \frac{\rho_l^*}{g} \quad (8)$$

123 where K_r is the relative permeability function and K_{sat} (m s^{-1}) the hydraulic
124 conductivity at saturation.

125 The chemical potential is a function of the water content described by
126 the soil/water retention curve. It is usually built by merging measurements
127 from classical tensiometry and sorption isotherm. Depending on the research
128 area (civil, petrol or food engineering, agronomy, pedology), various names
129 are used to designate this potential (suction, soil matrix potential, capillary
130 pressure, liquid activity). The chemical potential can be seen as a unifying
131 concept well defined in a thermodynamic framework (Job and Hermann,
132 2006). From its energetic definition, the chemical potential can consistently
133 describe the thermodynamic state of water over the whole range of water
134 content (Bénet et al., 2012).

135 2.4. *Liquid-gas phase change*

136 From thermodynamic considerations, it can be shown that the volumetric
137 rate of phase change $\hat{\rho}$ ($\text{kg m}^{-3} \text{ s}^{-1}$) is proportional to the water chemical
138 potential difference between the liquid and vapour states (Bedeaux and Kjel-
139 strup, 2004; Bénet and Jouanna, 1982; Kuiken, 1994). A detailed develop-
140 ment of this phase change theoretical relation has been given by Bénet et al.
141 (2009) and the main results are recalled here. In the isothermal case, the
142 non-equilibrium phase change rate is expressed as a function of the vapour
143 partial pressure by:

$$\hat{\rho}_v = -L \frac{R}{M_w} \ln \left(\frac{p_v}{p_{veq}} \right) \quad (9)$$

144 where L (kg K s m^{-5}) is a phenomenological coefficient to be determined ex-
145 perimentally as detailed in next section. The vapour pressure at equilibrium,

146 p_{veq} , is defined as the product of the saturating vapour pressure at the given
 147 temperature multiplied by the water activity:

$$p_{veq} = a_w(w) p_{vs}(T) \quad (10)$$

148 The liquid activity is defined by the sorption isotherm curve. Since it is
 149 an equilibrium properties, it can only describe an equilibrium situation as
 150 required by the local equilibrium assumption. Rejecting this fundamental
 151 hypothesis means that the vapour pressure, p_v , can diverge from its equilib-
 152 rium value, p_{veq} .

153 2.5. Numerical discretization

154 Finally, mass balance equations are written:

$$\frac{\partial w}{\partial t} - K_{sat} \frac{\rho_e^*}{\rho_s g} \nabla \cdot \left(K_r \frac{\partial \mu_l}{\partial w} \nabla w - K_r \mathbf{g} \right) - L \frac{R}{\rho_s M_w} \ln \left(\frac{p_v}{p_{veq}} \right) = 0 \quad (11)$$

$$\frac{\partial}{\partial t} (\phi_g p_v) - \nabla \cdot (D_{vs} \nabla p_v) + L \frac{R^2 T}{M_w^2} \ln \left(\frac{p_v}{p_{veq}} \right) = 0 \quad (12)$$

156 These non-linear equations are strongly coupled, on one hand, through the
 157 phase change term, $\hat{\rho}_v$, and on the other hand, through physical characteris-
 158 tics that depends on the water content: $\phi_g(w)$, $D_{vs}(w)$, $L(w)$.

159 Balance equations are discretized based on finite-volume formulation us-
 160 ing a one-dimensional regular mesh where the unknowns, w and p_v are located
 161 at the centre of grid blocks. A first-order upstream scheme is used to describe
 162 the convective term appearing in the liquid mass balance. Temporal integra-
 163 tion is performed using a fully implicit scheme to get numerical stability. To
 164 handle with non-linearities, a Newton-Raphson method ensures an accurate
 165 convergence for a moderate time step.

Figure 1: Soil granulometric curve.

166 3. Materials: a typical soil from Burkina-Faso

167 The material under investigation is a natural soil from Nasso, Burkina-
 168 Faso. From its particle size distribution (Fig. 1), this soil can be classified as
 169 a fine sand, the silty or clayey fraction is almost negligible. This is in agree-
 170 ment with the sorption isotherm curve which highlights a limited hygroscopic
 171 domain that ranges from 0% to 2% (Fig. 2).

172 3.1. Vapour diffusion

173 Through Fick's equation (Eq. 6), the vapour diffusion flux depends on
 174 the effective diffusion coefficient in soil, D_{vs} . It is generally defined as:

$$D_{vs} = \tau \phi_g D_{va} \quad (13)$$

Figure 2: Experimental sorption isotherm curve and theoretical modelling (Fredlund et al., 2002).

175 where τ represents the tortuosity and D_{va} the free diffusion coefficient of
 176 vapour in air. Standard correlation gives a value at 30°C, $D_{va} = 26.1 \times$
 177 10^{-6} m s^{-2} (Campbell, 1985). Several relations have been proposed to de-
 178 scribe the tortuosity coefficient as a function soil characteristics (Abu-El-Shar
 179 and Abriola, 1997; Moldrup et al., 2000). The classical relation proposed by
 180 Millington & Quirck (Moldrup et al., 2001) has been chosen:

$$\tau = \phi_g^{\frac{7}{3}} / \phi^2 \quad (14)$$

181 Anyway, numerical simulations have brought out a weak dependence of total
 182 water transport on the diffusion model. Regarding the case under investiga-
 183 tion, the various relations proposed in the literature are roughly equivalent.

184 3.2. *Liquid filtration*

185 The soil water retention curve has been determined by merging measures
 186 from tensiometry and sorption isotherm. This is based on classical devices
 187 such as the pressure plate and standard saline solutions. Relying on both ex-
 188 perimental techniques leads to characterize the soil/water equilibrium state
 189 over the whole range of water content. As discussed by Bénet et al. (2012),
 190 a variety of micro-scale mechanisms governs the soil/water interaction. De-
 191 scribing their macro-scale manifestation by means of pressure head or suction
 192 is meaningless in the low water content range. The classical theory based on
 193 capillary effects leads to misinterpretations and cannot be extended without
 194 careful investigation. Therefore, the chemical potential is more appropri-
 195 ate. Following this point of view, the soil/water retention curve (Fig. 3) is
 196 presented with the water mass chemical potential in abscissa.

Figure 3: Experimental and fitted theoretical soil water retention curve (Fredlund and Xing, 1994).

Several models have been presented in the literature to represent the soil/water retention curve. By analysing the behaviour of several soils, Thakur et al. (2006) highlighted that the fitting function proposed by Fredlund and Xing (1994) is the most appropriate description valid for very low water content. This model is given by:

$$w = \left[1 - \frac{\ln \left(1 + \frac{\mu}{\mu_r} \right)}{\ln \left(1 + \frac{10^6}{\mu_r} \right)} \right] \frac{w_{sat}}{\left[\ln \left(e + \left(\frac{\mu}{\mu_i} \right)^n \right) \right]^m} \quad (15)$$

where μ_r , α , n and m are soil parameters. Numerical identification with experimental points lead to values: $\mu_i = 3.78$, $\mu_r = 9.02$, $n = 2.87$ and $m = 1.10$, as represented in Fig. 3.

The hydraulic conductivity at saturation, K_{sat} , was measured experimentally with saturated samples compacted to reach a solid apparent mass density, $\rho_s = 1500 \text{ kg m}^3$, which corresponds to a porosity, $\phi = 43\%$. The average value obtained from several trials is: $K_{sat} = 3.995 \times 10^{-6} \text{ m s}^{-1}$.

The relative permeability function, K_r , is assumed to be described by the Mualem's predictive model explicited in the case of the soil/water retention curve proposed by Van Genuchten (1980). The mathematical development leads to the following relation (Van Genuchten and Nielsen, 1985):

$$K_r = \sqrt{S_e} \left[1 - \left(1 - S_e^{\frac{1}{m}} \right)^m \right]^2 \quad (16)$$

where the effective saturation, S_e , is defined by

$$S_e = \frac{w - w_{res}}{w_{sat} - w_{res}} \quad (17)$$

The water content at saturation, w_{sat} , the residual water content, w_{res} , and the exponent, m , are soils characteristic identified on the soil water retention curve (Fig. 3) using the relation proposed by Van Genuchten (1980).

217 Parameter values obtained from the numerical identification procedure are:
 218 $w_{sat} = 0.289$, $w_{res} = 0.00942$, $m = 0.526$.

219 3.3. Liquid-gas phase change

220 The phase change coefficient, L , introduced in this relation, should de-
 221 pends on the state variables such as the water content, the temperature and
 222 on the nature of the soil. This coefficient must be determined experimentally
 223 and has been the focus of several works (Lozano et al., 2008, 2009; Ruiz and
 224 Bénét, 2001). An original experimental device has been developed to analyse
 225 the return back to equilibrium of a soil sample subjected to non-equilibrium
 226 conditions. This non-equilibrium situation is caused by, first, extracting the
 227 gas phase of the soil sample, and then, replacing it by dry air, what results
 228 in a macroscopic thermodynamic non-equilibrium between the liquid phase
 229 and its vapour. Thus, the dependence of the phase change coefficient L on
 230 several physical variables (temperature, water content, total gas pressure)
 231 has been experimentally investigated (Lozano et al., 2009; Ruiz and Bénét,
 232 2001). The influences of the nature of the liquid and of the soil texture have
 233 also been underlined.

234 From a large set of experimental data carried out in isothermal conditions
 235 with pure water in clayey silty sand, Lozano et al. (2008) have provided a
 236 complete model of the phase change coefficient. Its variations as functions
 237 of the water content and the vapour partial pressure are characterised by 3
 238 coefficients (L_{eq} , k , r) through the following expressions:

$$\text{close to equilibrium, i.e.,} \quad r \leq \frac{p_v}{p_{veq}} \leq 1 \quad : \quad L = L_{eq} \quad (18)$$

239

$$\text{far from equilibrium, i.e.,} \quad 0 \leq \frac{p_v}{p_{veq}} \leq r \quad : \quad L = L_{eq} + k \left(r - \frac{p_v}{p_{veq}} \right) \quad (19)$$

240 The neighbourhood of an equilibrium situation, i.e., when the vapour partial
 241 pressure is close to its equilibrium value, corresponds to the validity domain
 242 of the linear thermodynamics of irreversible processes and a constant phe-
 243 nomenological coefficient is observed. Outside of this domain, i.e., far from
 244 equilibrium, an affine dependence on the vapour partial pressure is obtained
 245 and the phase change rate is highly increased. Concerning the soil under
 246 investigation, the influence of the water content on model parameters (L_{eq} ,
 247 r) is presented in Figs. 4 and 5 while the third parameter r has be found
 248 to be fairly constant, $r = 0.93$. Some bell-shaped curves are generally ob-
 249 served, where the maximum around 3% is roughly the upper limit of the
 250 hygroscopic domain. Above this maximum, the phase change rate decreases
 251 since the liquid-gas interface area reduces. For water content greater than
 252 7%, the gas phase is occluded and phase change cannot be activated. Below
 253 the maximum, when hygroscopic effects become predominant, the intensity
 254 of solid-liquid interactions increases in the adsorbed layers. The supplemen-
 255 tary energy required for water desorption decreases the phase change rate
 256 leading to lower values of the coefficient.

257 4. Validation: laboratory column experiments

258 4.1. Laboratory column experiments

259 Homogenized wet soil was compacted in a PVC tube to reach a solid
 260 apparent mass density, $\rho_s = 1500 \text{ kg m}^3$, which corresponds to a porosity, $\phi =$

Figure 4: Phenomenological coefficient close to equilibrium, Leq , versus the water content, w . Experimental points and fitting relation.

Figure 5: Phenomenological coefficient far from equilibrium, k , versus the water content, w . Experimental points and fitting relation.

261 43%. Sample dimensions were 30 cm height and 7.5 cm diameter. The initial
 262 water content of the soil was fixed at $w = 6\%$ to focus on low water content
 263 cases. Then, soil samples were placed in a regulated drying atmosphere at
 264 controlled temperature $T = 30^\circ\text{C}$ and relative humidity $RH_1 = 30\%$ for
 265 the first case, and $RH_2 = 50\%$ for the second case. The lower surface was
 266 hermetically closed while the upper surface was in contact with air.

267 Columns were weighed at regular time steps to determine the average
 268 water content leading to the drying kinetics plotted in Figs. 6 and 7. In
 269 each case ($RH_1 = 30\%$ and $RH_2 = 50\%$), the six experimental kinetics were
 270 achieved with a good reproducibility. The determination of water content
 271 profiles was based on a destructive method. Indeed, soil columns were cut
 272 into 2 cm slices and water content was measured by differential weighing
 273 after 48 hours drying at 105°C .

274 The evolutions of water content profiles presented in Figs. 8 and 9 clearly
 275 highlight water transport towards the upper side. Surface water content
 276 quickly decreases to the residual water content given by the soil/water reten-
 277 tion curve (Fig. 3): $w_{res} \approx 0.01$. At beginning, one can observe that the water
 278 content at bottom slightly exceeds the initial water content. This underlines
 279 that, in this range of water content, gravitational and capillary forces are of
 280 the same order of magnitude.

281 4.2. Comparison with theoretical model

282 Using identical boundary and initial conditions, numerical simulation of
 283 Eqs. 11 and 12 are plotted in solid line on Figs. 6 through 9. A very good
 284 agreement between the experimental drying kinetics and theoretical predic-
 285 tions is obtained in both cases (Figs. 6 and 7). Furthermore, numerical sim-

286 ulations are able to fairly represent the overall evolutions of water content
287 profiles (Figs. 8 and 9).

288 Even if the overall agreement is good, some local discrepancies are ob-
289 served. For instance, at the end of experiments, the gradient of water con-
290 tent is roughly constant what numerical profiles cannot represent. This is-
291 sue mainly depends on the relative permeability description (Eq. 16) which
292 has not been developed specifically for the soil under investigation. Further
293 experimental characterization is required to completely define the specific
294 relative permeability relation associated with this soil. Anyway, by relying
295 on the literature, the relation chosen in this case seems to be appropriate.

296 On can notice an evaporation front that penetrates inside the soil column
297 from the top (Rose et al., 2005; Yamanaka and Yonetani, 1999). It corre-
298 sponds to the propagation of the upper boundary condition through the soil
299 resulting from diffusion and phase change processes. Therefore, a major issue
300 is that the response of soil columns to the imposed relative humidity is ade-
301 quately represented by our numerical model. Without any fitting parameter,
302 the competition of three phenomena (filtration, diffusion and phase change)
303 is an accurate description of water transport in soils.

304 **5. Is liquid/gas equilibrium is a valid assumption?**

305 First of all, the validity of the isothermal assumption can be asserted us-
306 ing numerical simulation. From the numerical results presented in previous
307 section, the heat power consumed by phase change has been computed along
308 the soil column for each time step. At short times ($t < 2$ days), absolute value
309 of the heat power reaches a maximum of 4500 W m^{-3} in the first layer at

Figure 6: Drying kinetics. Comparison between theoretical prediction and experimental results in the first case: $RH_1 = 30\%$.

Figure 7: Drying kinetics. Comparison between theoretical prediction and experimental results in the second case: $RH_2 = 50\%$.

Figure 8: Water content profiles along soil columns. Comparison between theoretical prediction and experimental results in the first case: $RH_1 = 30\%$.

Figure 9: Water content profiles along soil columns. Comparison between theoretical prediction and experimental results in the second case: $RH_2 = 50\%$.

310 the upper surface. This value quickly decreases below 1000 W m^{-3} reaching
 311 80 W m^{-3} at the end of experiment. Since the heat power is maximal in
 312 the first layer of soil in contact with the thermo-regulated atmosphere, small
 313 temperature deviations could be observed in the upper part of soil columns
 314 in the first 2 days. Temperature gradient would enhance superficial vapour
 315 flux and eventually speed up the drying process. This issue could account for
 316 the discrepancies observed in drying kinetics at the beginning of experiments
 317 (Figs. 6 and 7). Anyway, the heat power computed beyond the first 2 days
 318 is not sufficient to generate significant temperature deviations. Indeed, the
 319 isothermal assumption is valid as long as the overall drying kinetics is suffi-
 320 ciently slow what is fairly verified in the cases under investigation. Further
 321 work is required to couple our model with heat flows and provide a detailed
 322 analysis of the isothermal assumption. However, this issue is beyond the
 323 scope of this paper.

324 Most of water (vapour and liquid) transport models developed to describe
 325 drying or water management processes relies on the *local equilibrium assump-*
 326 *tion*. This hypothesis specifies that, on each point of the domain, liquid and
 327 gas phases are in equilibrium. In our case, it can be written as:

$$\forall t, p_v = p_{veq} \quad (20)$$

328 postulating that the vapour pressure remains at its equilibrium value. Phys-
 329 ically, it amounts to saying that, when compared to diffusion, phase change
 330 processes are sufficiently fast to assume it instantaneous. This usual as-
 331 sumption has been taken into account without carefully checking its domain
 332 of validity. A large set of experimental investigations on phase change kinet-
 333 ics have suggested that evaporation in soils may not be as fast as supposed

334 (Bénet and Jouanna, 1982; Lozano et al., 2008; Ruiz and Bénet, 2001). For
 335 instance, phase change kinetics is drastically decreased when the binding en-
 336 ergy of water layers adsorbed on fine-scale grains increased (Lozano et al.,
 337 2009). This is mainly the case in the hygroscopic range of water content and
 338 is emphasized in the presence of a clayey fraction.

339 To evaluate the liquid/gas non-equilibrium, the ratio of the vapour pres-
 340 sure divided by its equilibrium value is proposed.

$$\theta = \frac{p_v}{p_{veq}} \quad (21)$$

341 This criterion is plotted along the soil column in Fig. 10. One can note that
 342 the vapour pressure can strongly deviate from its equilibrium value. Along
 343 the profile, average and largest value of non-equilibrium are given as functions
 344 of time in Fig. 11. The short time behaviour ($t < 4$ days) corresponds to the
 345 establishment of the vapour profile governed by diffusion. It is a transient
 346 behaviour which is not of particular interest with regards to the practical
 347 applications concerned. For larger time ($t > 10$ days), as transport processes
 348 are fully established, average non-equilibrium is close to 1 while local values
 349 can go up to 0.6.

350 Thus, deviation from equilibrium is significant only in a limited region
 351 ($0.25 < z < 0.20$). To analyse this point, computed water fluxes in liquid
 352 and gas phases are shown in Fig. 12. Liquid and gas water fluxes are the 1D
 353 representations of, respectively, $\rho_l \mathbf{v}_l$ in Eq. 1 and $\mathbf{J}_v$ in Eq. 2. To compare
 354 these fluxes with phase change rate, it has been rewritten in terms of surface
 355 flux:

$$\hat{J}_v = \hat{\rho}_v dz \quad (22)$$

356 where dz is the spatial discretization used in numerical simulation. The
 357 intensities of water fluxes and phase change rate cannot be directly compared
 358 since they do not take part identically in mass balance equations (Eqs. 11
 359 and 12). Indeed, water fluxes are cumulative through the gradient operator
 360 while phase change is a volumetric rate. Nevertheless, the comparison of the
 361 three plots supports a qualitative analyse.

362 Water flux occurs predominantly in liquid phase in the lower part of
 363 columns and almost exclusively in gas phase in the upper part (Gowing
 364 et al., 2006; Yamanaka and Yonetani, 1999; Yanful and Mousavi, 2003). The
 365 switch from one behaviour to the other occurs precisely in the region where
 366 non-equilibrium is maximum ($0.25 < z < 0.20$ in Fig. 10). Since the non-
 367 equilibrium criterion corresponds to the generalized thermodynamic force
 368 governing the phase change flux (Eq. 9), phase change rate is maximum in
 369 this region which creates the main source term in vapour transfer processes.
 370 Thus, the 3 water transport phenomena coexist only in this transition zone.

371 As the evaporation front evocated in previous section penetrates in the
 372 soil column, this transition zone propagates from the upper boundary condi-
 373 tion towards soil bottom. This defines the dry surface layer (DSL) a few cen-
 374 timetres below the surface identified by Yamanaka et al. (1998); Yamanaka
 375 and Yonetani (1999). After all, the only manifestation of coupling effects
 376 between liquid and vapour transport mechanisms occurs in a limited region
 377 as well as the deviation from equilibrium. However, this particular zone
 378 governs the overall water transport kinetics. Actually, the cases under inves-
 379 tigation underline that the vapour diffusion is not impeded and that there is
 380 no specific limiting phenomena. Indeed, the kinetics results from a complex

Figure 10: Theoretical liquid/gas non-equilibrium, $\theta = \frac{p_v}{p_{veq}}$, along the soil column in the first case: $RH_1 = 30\%$ at $t = 38.2$ days.

competition of filtration, diffusion and phase change.

Taking into account the liquid/gas equilibrium assumption would lead to artificially increase the phase change rate and enhance the vapour flux. In such a case, the diffusion process would govern the overall kinetics. Since the equilibrium vapour pressure is directly linked to the water content through the sorption isotherm, the vapour diffusive flux would only depend on the water content vertical gradient. This means that a vapour diffusive flux could only occur in the hygroscopic domain where equilibrium vapour pressure deviates from the saturated vapour pressure. From our mind, this constraint is too restrictive while it is usually hidden by the non-isothermal vapour flux.

The material used in this work is a fine sand that does not present im-

Figure 11: Evolution of maximum and average liquid/gas non-equilibrium, $\theta = \frac{p_v}{p_{veq}}$, as function of time in the first case: $RH_1 = 30\%$.

Figure 12: Theoretical water fluxes in gas phase, $\mathbf{J}_v$, and liquid phase, $\rho_l \mathbf{v}_l$, along the soil column in the first case: $RH_1 = 30\%$ at $t = 38.2$ days. The surface representation of phase change rate, $\hat{J}_v$ (Eq. 22), is added for comparison.

392 portant hygroscopic effects, i.e., the hygroscopic domain represents a limited
 393 range of water content $0\% < w < 2\%$ (Fig. 2). It has been shown that
 394 phase change rate is drastically lowered as the water activity deviates from
 395 1 (Bénet et al., 2009). For instance, a small clay fraction ($\approx 10\%$) sig-
 396 nificantly increases hygroscopic effects and decreases phase change kinetics
 397 (Lozano et al., 2008). Thus, similar investigations carried out with silty sand
 398 or clayey sand would enhance the liquid/gas non-equilibrium calling for a
 399 two-equation model such as the one developed in this paper.

400 **6. Conclusion**

401 By rejecting the local liquid/gas equilibrium assumption, a non-equilibrium
 402 model of isothermal water transport in soil has been proposed. Liquid water
 403 and vapour transport are ruled by 2 mass balance equations while the cou-
 404 pling between them is described by a non-equilibrium phase change relation.
 405 It amounts to consider that the characteristic times associated with each wa-
 406 ter transport mechanism (liquid filtration, vapour diffusion, phase change)
 407 are of the same order of magnitude. By comparing numerical simulations
 408 to drying kinetics and water content profiles obtained from laboratory soil
 409 column experiments, a validation of this description is proposed. It must be
 410 recalled that the comparison test does not rely on any adjusted parameters
 411 since each soil characteristics has been identified with an independent and
 412 specific experimental procedure.

413 In the cases under investigation, local liquid/gas equilibrium can clearly
 414 not be a reasonable assumption. Even if average non-equilibrium is not
 415 particularly pronounced, it is locally significant as the vapour pressure can

416 drop down to half of its equilibrium value. The limited region where a large
417 non-equilibrium is observed defines the evaporation front. It corresponds to
418 the transition from liquid transport in deeper layers to vapour transport in
419 superficial layers which controls the moisture dynamics in bare soils. From
420 this model, the overall evaporation rate can be estimated directly without
421 resorting to empirical correlations (Bittelli et al., 2008; Yanful and Mousavi,
422 2003).

423 The more questionable aspect of this model concerns the description of
424 the relative permeability function. Indeed, it is now well established that the
425 relation proposed by Van Genuchten (1980) is not appropriate at low water
426 content. Further investigations are required to develop accurate modelling of
427 liquid flow phenomena at very low water content as proposed by Tuller and
428 Or (2002) and Peters and Durner (2002).

429 Since the coupling with heat flux is not taken into account, this model
430 cannot be applied directly on practical situations to describe natural field
431 observations. It is rather an heuristic model dedicated to discuss the local
432 equilibrium assumption and assess its eventual reliability. Atmospheric ther-
433 mal evolutions highly influence water content dynamics in the first layers of
434 bare soils, particularly in arid regions where diurnal temperature variations
435 are large (Gowing et al., 2006). Thus, the approach proposed in this work
436 must be included in a wider modelling such as existing land-surface models
437 (Garcia-Gonzalez et al., 2012; Simunek et al., 2008).

438 As evidenced previously, phase change kinetics is highly influenced by
439 hygroscopic effects (Lozano et al., 2008). For instance, a clayey fraction will
440 decrease the phase change rate and extend the hygroscopic domain. Such a

441 situation would lead to intensify non-equilibrium, or at least to widen the
442 transition zone and the evaporation front. In this framework, the introduc-
443 tion of organic matter like compost should modify the moisture dynamics.
444 Based on this approach, our objective is to revisit the traditional techniques
445 developed in subsaharian regions to prevent from soil evaporation and opti-
446 mize water resources management.

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