



HAL
open science

Water transfer in bread during staling: Physical phenomena and modelling

Jean-Yves Monteau, Emmanuel Purlis, Emna Besbes, Vanessa Jury, Alain Le-Bail

► **To cite this version:**

Jean-Yves Monteau, Emmanuel Purlis, Emna Besbes, Vanessa Jury, Alain Le-Bail. Water transfer in bread during staling: Physical phenomena and modelling. *Journal of Food Engineering*, 2017, 211, pp.95 - 103. 10.1016/j.jfoodeng.2017.04.016 . hal-01669317

HAL Id: hal-01669317

<https://hal.science/hal-01669317>

Submitted on 12 Sep 2023

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Accepted Manuscript

Water transfer in bread during staling: Physical phenomena and modelling

Jean-Yves Monteau, Emmanuel Purlis, Emna Besbes, Vanessa Jury, Alain Le-Bail



PII: S0260-8774(17)30152-8

DOI: [10.1016/j.jfoodeng.2017.04.016](https://doi.org/10.1016/j.jfoodeng.2017.04.016)

Reference: JFOE 8851

To appear in: *Journal of Food Engineering*

Received Date: 12 September 2016

Revised Date: 8 March 2017

Accepted Date: 8 April 2017

Please cite this article as: Monteau, J.-Y., Purlis, E., Besbes, E., Jury, V., Le-Bail, A., Water transfer in bread during staling: Physical phenomena and modelling, *Journal of Food Engineering* (2017), doi: 10.1016/j.jfoodeng.2017.04.016.

This is a PDF file of an unedited manuscript that has been accepted for publication. As a service to our customers we are providing this early version of the manuscript. The manuscript will undergo copyediting, typesetting, and review of the resulting proof before it is published in its final form. Please note that during the production process errors may be discovered which could affect the content, and all legal disclaimers that apply to the journal pertain.

Water transfer in bread during staling: physical phenomena and modelling

Jean-Yves Monteau^{a,b,*}, Emmanuel Purlis^{a,b,c}, Emna Besbes^{a,b},
Vanessa Jury^{a,b}, Alain Le-Bail^{a,b}

^aUBL, Oniris, département GPA, rue de la Géraudière, B.P. 82 225
44322 Nantes CEDEX 3, France

^bGEPEA UMR CNRS 6144, Nantes, France

^cCentro de Investigación y Desarrollo en Criotecnología de Alimentos (CIDCA), UNLP,
CONICET, Facultad de Ciencias Exactas, 47 y 116, La Plata (1900), Argentina

Abstract

Starch retrogradation and water loss have effects of the same intensity on the increase in firmness in the phenomenon of bread staling. Writing the equations of this system, in order to understand the mechanisms of water transfer in the vapour and liquid phases, is apparently simple. Nevertheless, choices are necessary for the simplifying hypotheses. Two models, differing in their geometry and their equations, were developed. Besides investigating the water transfer mechanisms in the vapour and liquid phases, the aim of this study is to compare the results of the two models and to conclude as to their individual interest. Concerning the physical phenomena, the study shows that a part of the water lost by the crumb escapes into the atmosphere while another part is absorbed by the crust. As regards the modelling the study shows that the most complete model is not the best choice and specifies the simplifying assumptions that should be retained or eliminated.

*Corresponding author

Email address: jean-yves.monteau@oniris-nantes.fr (Jean-Yves Monteau)

Keywords: Sandwich bread, staling, water transfer modelling

1 **1. Introduction**

2 The staling of bread is defined as its firming over time and results in a
3 loss of smoothness (Roussel and Chiron, 2002). Not only are its textural
4 properties modified but also its aromatic properties. This evolution is per-
5 ceptible for a product like sandwich bread sold in a tight packaging. The
6 retrogradation of starch is essential for staling (Hug-Iten et al., 1999), but
7 water migration plays an important part through its distribution among the
8 bread components. In fact, bread with crust loses its freshness quickly while
9 bread without crust stays fresh (Bechtel et al., 1953; Besbes et al., 2014).
10 However, the effect of the presence of the crust is exerted on the exchange of
11 water between the crumb, the crust and the atmosphere and not on starch
12 retrogradation. According to Ronda et al. (2011), starch retrogradation and
13 water loss have effects of the same intensity on the increase in bread firming.
14 Water migration results in an equilibration of the water content between the
15 crust and the crumb at the macroscopic scale and in the redistribution of
16 moisture between the components at the microscopic scale. This equilibrium
17 is unstable and may drift towards crust softening and crumb drying.

18
19 Water transfer in a product is driven by different mechanisms: molecular
20 diffusion (gas phase), convection and liquid capillarity. Several authors have
21 used a simple model based on molecular diffusion in dense, homogeneous and
22 isotropic media with Fick's second law to describe the mass transfer (Datta,
23 2005). However, the mass transfer of water in liquid or in vapour form was

24 not differentiated. Other authors, for instance Thorvaldsson and Janestad
25 (1999), preferred to use Fick's law in separating liquid water transfer and
26 water vapour transfer. Nevertheless, Fick's law alone does not explain the
27 mass transfer in porous media because diffusion is not the only mechanism
28 responsible. Therefore, it is necessary to add a convection term to the diffu-
29 sion term based on Darcy's law: the transfer is then also due to a pressure
30 gradient. A synthesis of formulations of heat and mass transfer problems has
31 been provided by Datta (2007a).

32

33 To understand the mechanisms of water transfer in bread during staling,
34 we have developed models of the system. Modelling a process involves a
35 number of assumptions and choices for the equations. Thus the same pro-
36 cess can be described by several models, with the most complex in terms of
37 its equations not necessarily being the best. For bread staling we tested two
38 models with the following main assumptions, respectively: constant pres-
39 sure and incorporation of the crust via the water vapour permeability in the
40 boundary conditions; a composite domain (crumb and crust) with variable
41 pressure. The aim of this work is to compare the results given by these two
42 models and to conclude about their application scope.

43 **2. Modelling**

44 The modelling is greatly inspired by the model of Whitaker (1977) devel-
45 oped for the drying process, with some adaptations to the staling problem.
46 The problem is isothermal. So, there is no heat transfer equation. We are
47 particularly interested in three quantities varying in time and 2D-space: the

48 local content of liquid water, water vapour and dry air, these two gases form-
49 ing the gaseous phase. The geometry is the upper right quarter of a slice
50 of bread, placed horizontally in order to eliminate the gravitational effect
51 (Figure 1). For the first model, the crust is assumed to be a membrane char-
52 acterised by its water vapour permeability.

53

54 In the first model, the pressure of the bread slice is assumed equal to atmo-
55 spheric pressure. The system modelling is based on two governing equations:
56 one for liquid water and one for the local water vapour content. Because the
57 total pressure is constant, the dry air pressure and the local dry air content
58 are derived from the local water vapour content.

59

60 In the second model, the crust is a domain with its own characteristics
61 (density, porosity, etc.) different from the crumb. The total pressure is no
62 longer supposed constant. The local dry air content is assumed to be gov-
63 erned by a similar equation to that which governs the local water vapour
64 content. Partial pressures of each gas and thus the total pressure can vary.

65

66 Figure 1 presents a summary of assumptions of each model, which are
67 further discussed next.

68 *2.1. First Model*

69 The variation in the local water content, X_{liq} , is described by Darcy flow
70 (due to liquid pressure: $P_{\text{liq}} = P - P_c$) with the addition of an evaporation-

71 condensation volumetric rate, I :

$$\frac{\partial X_{\text{liq}}}{\partial t} = \nabla \cdot \left[\rho_{\text{liq}} \frac{k_{\text{liq}}}{\mu_{\text{liq}}} \nabla (P - P_c) \right] - I \quad (1)$$

72 where P_c is the capillary pressure and P the total pressure. ρ_{liq} and μ_{liq} are
73 the density and the dynamic viscosity of liquid water, and k_{liq} is the crumb
74 permeability to liquid water. Since liquid water is strongly bound to starch,
75 the total pressure of the driven flow is not significant in our case, so the
76 following expression is used:

$$\frac{\partial X_{\text{liq}}}{\partial t} = \nabla \cdot (D_{\text{liq}} \nabla X_{\text{liq}}) - I \quad (2)$$

77 where D_{liq} is the capillary diffusivity of liquid water, defined as (Datta,
78 2007b):

$$D_{\text{liq}} = -\rho_{\text{liq}} \frac{k_{\text{liq}}}{\mu_{\text{liq}}} \frac{\partial P_c}{\partial X_{\text{liq}}} \quad (3)$$

79 The variation in water vapour, X_{vap} , is driven by a diffusion equation
80 with the corresponding source term:

$$\frac{\partial X_{\text{vap}}}{\partial t} = \nabla \cdot \left[\rho_g \Phi (1 - S) D_{\text{av}} \nabla \frac{\rho_{\text{vap}}}{\rho_g} \right] + I \quad (4)$$

81 The air-vapour diffusion coefficient D_{av} is weighted by the ratio of gas vol-
82 ume to total volume, i.e. the porosity Φ multiplied by the gas fraction in the
83 pores, $1 - S$; S is the water saturation in the pores and ρ_g is the gas density.

84

85 The evaporation-condensation rate is assumed proportional to the dif-
86 ference between the equilibrium water vapour pressure, $a_w P_{\text{sat}}(T)$, and the
87 vapour pressure in the pores P_{vap} . a_w is the crumb water activity and $P_{\text{sat}}(T)$
88 is the saturation pressure of the water vapour at temperature T :

$$I = C [a_w P_{\text{sat}}(T) - P_{\text{vap}}] \quad (5)$$

89 C is the proportionality coefficient. The crumb water activity was experi-
 90 mentally determined to fill a table function of the local water content (Besbes
 91 et al., 2013). This table is interpolated using piecewise cubic functions.

92 The water vapour pressure is derived from the perfect gas law:

$$P_{\text{vap}} = \frac{X_{\text{vap}} R T}{M_{\text{vap}} \Phi (1 - S)} \quad (6)$$

93 where R is the perfect gas law constant and M_{vap} is the molar mass of water
 94 vapour.

95
 96 This first model is characterised by a total pressure assumed constant
 97 and equal to atmospheric pressure. Thus, the dry air pressure is derived by
 98 subtracting the water vapour pressure from the total pressure:

$$P_{\text{air}} = P - P_{\text{vap}} \quad (7)$$

99 Then, the dry air content X_{air} is derived using the perfect gas law:

$$X_{\text{air}} = \frac{P_{\text{air}} M_{\text{air}} \Phi (1 - S)}{R T} \quad (8)$$

100 where M_{air} is the molar mass of dry air.

101
 102 The water vapour and dry air densities can be calculated from the mass
 103 concentrations in the porous matrix:

$$\rho_{\text{vap}} = \frac{X_{\text{vap}}}{\Phi (1 - S)} \quad (9)$$

$$\rho_{\text{air}} = \frac{X_{\text{air}}}{\Phi (1 - S)} \quad (10)$$

104 The saturation pressure is calculated with the Dupré formula, which can
 105 be used from -50 to 200 °C:

$$P_{\text{sat}}(T) = 133.32 \exp\left(46.784 - \frac{6435}{T} - 3.868 \ln T\right) \quad (11)$$

106 where T is in K and P_{sat} in Pa. The problem is isothermal, thus P_{sat} is
 107 constant.

108

109 The pores saturation is calculated with the following equation:

$$S = \frac{X_{\text{liq}}}{\rho_{\text{liq}} \Phi} \quad (12)$$

110 Lastly, the gas density is the sum of the dry air and water vapour densities:

111

$$\rho_{\text{g}} = \rho_{\text{vap}} + \rho_{\text{air}} \quad (13)$$

112 *2.1.1. Boundary conditions:*

113 The mass transfer with the ambient air is assumed to occur only in the
 114 gaseous phase. Consequently, the liquid water flow at the surface is equal to
 115 zero, as well as at the left and bottom boundaries because of the symmetries
 116 (see Figure 1 a). Thus, in all boundaries, the boundary condition for liquid
 117 water is

$$n \cdot (D_{\text{liq}} \nabla X_{\text{liq}}) = 0 \quad (14)$$

118 where n is the outward normal to the boundary. Regarding X_{vap} , on the left
 119 and bottom boundaries, the vapour flow is equal to zero,

$$n \cdot \left[\rho_{\text{g}} \Phi (1 - S) D_{\text{av}} \nabla \frac{\rho_{\text{vap}}}{\rho_{\text{g}}} \right] = 0 \quad (15)$$

120 and at the surface it is determined by the water vapour permeability of the
 121 crust WVP:

$$n \cdot \left[\rho_g \Phi (1 - S) D_{av} \nabla \frac{\rho_{vap}}{\rho_g} \right] = \text{WVP} \frac{P_{vap,a} - P_{vap}}{e} \quad (16)$$

122 where e is the crust thickness. The vapour pressure in the ambient air is
 123 calculated conventionally by

$$P_{vap,a} = RH P_{sat}(T) \quad (17)$$

124 where RH is the ambient relative humidity.

125 2.2. Second Model

126 For the second model, the presence of the crust is explicitly incorporated.
 127 An external layer (5 mm in thickness) is added to the crumb (Figure 1 b)
 128 to represent the crumb-crust composite system. Furthermore, the crumb
 129 and crust have different properties, mainly due to differences in structure
 130 developed during baking. For instance, the following expressions inspired
 131 from (Datta, 2007b) and fitted to our product were used for the capillary
 132 diffusivity of liquid water:

$$D_{liq,crumb} = 1.5 \times 10^{-9} e^{(-2.8+2 X_{dm})} \Phi \quad (18)$$

$$D_{liq,crust} = 1 \times 10^{-9} e^{(-2.8+2 X_{dm})} \Phi \quad (19)$$

133 where X_{dm} is the local water content on a dry basis. The relationship between
 134 X_{liq} and X_{dm} is

$$X_{dm} = \frac{X_{liq}}{\rho_{app,dm}} \quad (20)$$

135 where $\rho_{\text{app,dm}}$ is the apparent density of the desiccated matrix.

136

137 Regarding the water activity of the crust, a water sorption isotherm ob-
138 tained at 15 °C is used (Besbes et al., 2013).

139

140 The total pressure a priori can vary, thus the governing equation (1) for
141 the water vapour content is modified by the addition of a flow term.

$$\frac{\partial X_{\text{vap}}}{\partial t} = \nabla \cdot \left[\rho_{\text{g}} \Phi (1 - S) D_{\text{av}} \nabla \frac{\rho_{\text{vap}}}{\rho_{\text{g}}} + \rho_{\text{vap}} \frac{K_{\text{g}}}{\mu_{\text{g}}} \nabla P \right] + I \quad (21)$$

142 where K_{g} is the permeability of the media to the gaseous phase and μ_{g} the
143 dynamic viscosity of this phase.

144 K_{g} is given by Equation (22) (Jury, 2007):

$$K_{\text{g}} = 6.55 \times 10^{-11} \Phi^{3.03} \quad (22)$$

145 μ_{g} is taken equal to air dynamic viscosity (Table 1).

146

147 X_{air} is assumed to vary according to a governing equation similar to that
148 governing the evolution of X_{vap} , but without the evaporation-condensation
149 term:

$$\frac{\partial X_{\text{air}}}{\partial t} = \nabla \cdot \left[\rho_{\text{g}} \Phi (1 - S) D_{\text{av}} \nabla \frac{\rho_{\text{air}}}{\rho_{\text{g}}} + \rho_{\text{air}} \frac{K_{\text{g}}}{\mu_{\text{g}}} \nabla P \right] \quad (23)$$

150 Other equations are unchanged, except for the total pressure P which is no
151 longer equal to atmospheric pressure. From a mathematical point of view,
152 Equation (23) replaces the equation $P = P_{\text{atm}}$, where P_{atm} refers to the at-
153 mospheric pressure.

154

155 *2.2.1. Boundary conditions*

156 For X_{liq} , the boundary conditions are insulation conditions on all the ex-
 157 ternal boundaries, and at the crumb-crust interface there is continuity of the
 158 water content. The addition of a partial derivative equation to calculate the
 159 dry air content involves defining the boundary conditions for this variable.
 160 For the left and bottom boundaries (Figure 1 b) symmetry conditions or zero
 161 flow conditions apply. For the surface boundary, a Dirichlet condition, thus
 162 a value of the dry air content, cannot be fixed because this value is unknown.
 163 In fact, there may be a discontinuity in the dry air content at this boundary.
 164 A Neumann condition, i.e. a flow condition, cannot be used because it is
 165 not better known. The natural boundary condition is total pressure equal
 166 to atmospheric pressure, assuming that it varies without discontinuity from
 167 the crumb to the atmosphere. However, a boundary condition for the dry air
 168 content cannot be deduced from this pressure condition because the condi-
 169 tions above lead to a Dirichlet condition involving the water vapour content
 170 on this boundary. Yet, this Dirichlet condition is not known. Because of this
 171 technical difficulty, equations were rewritten to use, as dependent variables,
 172 the liquid water and water vapour contents, and the total pressure. The
 173 following pressure equation, as governing equation, is thus obtained:

$$\begin{aligned}
 \frac{1}{RT} \left(\Phi - \frac{X_{\text{liq}}}{\rho_{\text{liq}}} \right) \frac{\partial P}{\partial t} + \nabla \cdot \left[-\rho_{\text{g}} \Phi (1 - S) D_{\text{av}} \left(\frac{1}{M_{\text{vap}}} \nabla \frac{\rho_{\text{vap}}}{\rho_{\text{g}}} + \frac{1}{M_{\text{air}}} \nabla \frac{\rho_{\text{air}}}{\rho_{\text{g}}} \right) \right. \\
 \left. - \left(\frac{\rho_{\text{vap}}}{M_{\text{vap}}} + \frac{\rho_{\text{air}}}{M_{\text{air}}} \right) \frac{K_{\text{g}}}{\mu_{\text{g}}} \nabla P \right] = \\
 \frac{1}{\rho_{\text{liq}} \Phi - X_{\text{liq}}} \left(\frac{X_{\text{vap}}}{M_{\text{vap}}} + \frac{X_{\text{air}}}{M_{\text{air}}} \right) [\nabla \cdot (D_{\text{liq}} \nabla X_{\text{liq}}) - I] + \frac{I}{M_{\text{vap}}} \quad (24)
 \end{aligned}$$

174 For P , there are symmetry conditions on the left and bottom boundaries,
 175 and the Dirichlet condition $P = P_{\text{atm}}$ at the surface. There is pressure con-
 176 tinuity at the interface.

177

178 For X_{vap} , the same symmetry conditions apply on the left and bottom
 179 boundaries as for model 1, and the flow condition (16) is applied at the
 180 crumb-crust interface. A boundary condition is necessary at the external
 181 surface in contact with the atmosphere. The same difficulties appear as for
 182 the air content boundary condition: the water vapour content and the flow
 183 of water vapour are unknown at the crust surface. To close the system of
 184 equations, the vapour pressure at the surface is assumed equal to that of the
 185 atmosphere, which leads to the following Dirichlet condition:

$$X_{\text{vap}} = \frac{P_{\text{vap,a}} M_{\text{vap}} \Phi (1 - S)}{RT} \quad (25)$$

186 2.3. Parameters

187 The models constants are either known physical constants (D_{av} , R , M_{vap} ,
 188 etc.), values specific to our product (D_{liq} , Φ , WVP, etc.), or environmental
 189 parameters T , P_{atm} and RH . They are given in Table 1. Except for the pa-
 190 rameter C , the characteristic values of the product were measured, as were
 191 the environmental parameters. The material and methods used and the ex-
 192 perimental results are described in (Besbes, 2012).

193

194 2.4. Initial conditions

195 For the first model, the initial conditions necessary to solve the two dif-
 196 ferential equations were:

$$X_{\text{liq}} = 160 \text{ kg m}^{-3} \quad (X_{\text{dm}} = 0.83) \quad (26)$$

$$X_{\text{vap}} = 7.849 \times 10^{-3} \text{ kg m}^{-3} \quad (27)$$

197 The initial liquid water content was set at the experimental value and the
198 water vapour content was determined to be equal to that of the atmosphere.

199

200 For the second model, the initial condition necessary to solve Equa-
201 tion (24) was added:

$$P = P_{\text{atm}} \quad (28)$$

202 For this model, initial conditions for the crust are also necessary. As for the
203 first model X_{liq} was set equal to the experimental value and X_{vap} equal to
204 that of the atmosphere.

$$X_{\text{liq}} = 75.9 \text{ kg m}^{-3} \quad (X_{\text{dm}} = 0.23) \quad (29)$$

$$X_{\text{vap}} = 7.756 \times 10^{-3} \text{ kg m}^{-3} \quad (30)$$

$$P = P_{\text{atm}} \quad (31)$$

205 2.5. Programming, geometry and mesh

206 Programming was achieved using Comsol 4.2 and 4.3. The geometry
207 used the symmetries of the product: a quarter of a bread slice was drawn.
208 The mesh for model 2 was refined until the results at the selected points no
209 longer changed. For model 1 a coarser mesh was sufficient but the same level
210 of refining as for the second model was kept. Meshes are shown in Figure 2.

211 3. Model fitting

212 The second model is assumed to be the most realistic as it considers the
213 real geometry with the crust as a distinct area with its own physical char-
214 acteristics. Besides, it uses the most complete equations without hypotheses
215 about the pressure inside, calculating the dry air content in the same way
216 as the water vapour content and with the flow term. For these reasons,
217 it was selected as the model to best-fit (i.e. fine-tune) the unknown model
218 parameter (namely, the evaporation coefficient) against experimental results.

219
220 For the fitting, experimental and computed local water contents averaged
221 on discs were compared. These discs are shown in Figure 2. This resulted
222 in a modification of the mesh. All the discs are in the crumb. In order
223 to obtain water content profiles, averages of the values obtained for a given
224 x-coordinate, or on the height of the slice, were calculated for 4 values of
225 staling time. These values were 2 h (D0), 3 days (D3), 7 days (D7) and
226 10 days (D10).

227
228 The evaporation coefficient C cannot be experimentally determined. C
229 was used as a fitting parameter to fine-tune the numerically simulated water
230 content profiles against experimental counterparts. The value obtained for
231 C is $5 \times 10^{-4} \text{ s m}^{-2}$. Figure 3 illustrates the quality of the fit. The model
232 underestimates the water loss at the start of staling (D0) because the com-
233 puted water content profile, being constant and nearly equal to the initial
234 value, is above the experimental profile. The profiles computed at D3, D7
235 and D10 are relatively close to the experimental ones except for the averages

236 at $x = 38$ mm for the profiles D3 and D7. For these two cases, the computed
237 values are significantly over estimated. However, it is not possible to obtain
238 a better fit with this model, which has only one fitting parameter.

239 4. Results

240 4.1. Time variation of the main variables

241 The analysis in this section is performed on the results given by the 2nd
242 model because it is a priori the most faithful to reality. The local water
243 content is given on a dry basis, X_{dm} , according to the use in food process
244 engineering.

245
246 Figure 4 shows the variation in the liquid water content at the different
247 points considered. In the crumb, the local water content decreases constantly.
248 For the inner points, this decrease is linear, starting at point (25, 25) as
249 expected due to its proximity to crust. Note that the centre of the product
250 (point (0, 0)) also shows this trend, which may be due to evaporation since
251 gradients are not significant at the beginning of the process. Underneath the
252 crumb-crust interface (point (34.9, 34.9)), the local water content decreases
253 rapidly during the first day and then very slowly, once initial dehydration due
254 to proximity to crust is diminished and thus the driving force for transport is
255 reduced. On the other hand, above the interface (point (35.1, 35.1)), liquid
256 water content increases rapidly during the first day, but then decreases very
257 slowly. At the midpoint of the crust (point (37, 37)) and surface, liquid water
258 content increases constantly.

259 These results are explained by a reequilibration of water from the crumb

260 to the crust, with a decrease at the crumb-crust interface to supply the
261 crust. This rebalancing has already been observed experimentally (Besbes
262 et al., 2014). Above the interface, the increase in the water content at the
263 beginning is also explained by water vapour condensation (see Figure 7).
264 Then, although there is still condensation, the water content decreases. This
265 means that the liquid water diffusion overtakes the condensation. There is
266 a sharp drop in the water content at the crumb-crust interface, although a
267 continuity condition for the water content on a wet basis is imposed. This
268 discontinuity is normal on a dry basis because the relationship between the
269 water content on a wet basis and the water content on a dry basis is given
270 by Equation (20), with $\rho_{app,dm}$ being different for the crumb and the crust.
271 However, this discontinuity is present on a wet basis despite the continuity
272 boundary condition. In fact, the software complies with the condition but
273 the computation shows that the water content decreases drastically on pass-
274 ing the boundary.

275

276 In the crumb the water vapour content increases linearly but the three
277 lines at points (0, 0), (15, 15) and (25, 25) are almost parallel instead of
278 diverging as for the liquid water content (Figure 5). Below the crumb-crust
279 interface, the water vapour content increases rapidly during the first day and
280 afterwards more slowly. Above the interface (point (35.1, 35.1)), it decreases
281 rapidly during day 1, but then rises slowly after 1.6 days. The two curves
282 at the interface end by merging asymptotically. Both the crust and surface
283 points show a similar trend for the water vapour content, which decreases
284 continuously with different rates. Overall, the water vapour content is higher

285 in the crust than in the crumb until days 6-7; afterwards, the trend is the
286 opposite.

287 In the crumb, the linear and parallel increase in the water vapour content
288 at the three points considered can be explained by the combination of evap-
289 oration and diffusion. Evaporation is almost constant over time for these
290 three points, although with different values (see Figure 7): the closer to the
291 interface, the greater the evaporation. If there was only evaporation, the
292 lines should diverge. A result of this simulation is that the total pressure is
293 constant in the product (Figure 10). Thus, the only driving terms for the
294 movement of the water vapour are evaporation and diffusion, not pressure
295 gradient. Below the interface, the water vapour content increases because of
296 evaporation. In the crumb, the vapour diffusion goes from the centre towards
297 the interface because the water vapour content gradient is oriented inwards.
298 However, at the interface it goes from the crust to the crumb. In the end, the
299 two interface curves (at points (34.9, 34.9) and (35.1, 35.1)) merge and the
300 water vapour content in the crumb becomes higher than that in the crust,
301 because there is still condensation in the crust and evaporation in the crumb
302 (Figure 7).

303

304 The dry air content increases linearly in the crumb and faster close to the
305 interface (Figure 6). Below the interface, it first increases rapidly and then
306 more slowly while above it first decreases rapidly before increasing slowly. In
307 the crust and at the surface, it decreases more and more slowly.

308

309 The dry air and liquid water contents behave inversely. In the crumb,

310 the air content increases because the liquid water content decreases, thus de-
311 creasing the water saturation of pores. The dry air takes the place released
312 by liquid water. Overall, the dry air content shows the same behaviour as
313 the gas saturation.

314

315 In the crumb, the evaporation flow is constant over time but not in space
316 (Figure 7). It is lower at the centre and increases toward the interface.
317 Below the interface, it is initially significant and decreasing exponentially
318 while above the interface it is negative. Thus there is condensation. At the
319 beginning, it rapidly approaches 0, and then decreases very slowly (the con-
320 densation increases). In the crust and at the surface, the evaporation flow
321 increases slowly (the condensation decreases).

322

323 The evaporation flow results from an imbalance between the terms $a_w P_{\text{sat}}$
324 and P_{vap} , the water vapour pressure (Equation 5). Logically, until the bal-
325 ance is reached, the evaporation flow does not vanish. At the end of the
326 simulation the steady state is still not reached and the condensation con-
327 tinues to decrease at the surface and in the crust. Over time there would
328 probably be a balance, $a_w P_{\text{sat}} = P_{\text{vap}}$, but different for the crumb and the
329 crust because the isotherm is not the same for these two areas. The evapo-
330 ration flow would be zero everywhere but the shelf-life of the product would
331 be greatly exceeded.

332

333 In the equations, the water saturation differs from the liquid water con-
334 tent only by a coefficient. The variations are thus similar for the two variables

335 (Figure 8).

336

337 Figure 9 shows the variation in the overall water content for a 1 cm-bread
338 slice in the crumb, the crust and the whole slice.

339

340 In the whole slice and the crumb, the water content decreases in two al-
341 most parallel lines. However, the water content in the crust increases very
342 slightly in the first two days of staling and then tends to stabilise.

343

344 Thus the water that migrates to the atmosphere comes from the crumb,
345 passing by and slightly moistening the crust.

346

347 Figure 10 compares the water vapour pressure and dry air pressure in the
348 slice along an axis from the centre to the surface at the end of the simulation
349 (10.2 days). The water vapour pressure in the slice is very low compared
350 to the air pressure. Vapour pressure and air pressure seem constant in the
351 whole product. In reality, for these two gases there is a difference in pressure
352 between the crumb and the crust but due to the y-scale these differences
353 cannot be seen on the figure. The total pressure, although calculated, stays
354 constant and equal to atmospheric pressure. It is a realistic hypothesis that
355 can be used when writing the model equations. An interesting consequence
356 is that there is no Darcy flow because there is no total pressure gradient.
357 Gas transfers only occur by diffusion and evaporation-condensation. The
358 flow terms computed in the equations are found equal to zero.

359

360 To resume, these simulation results show that throughout staling there is
361 evaporation in the crumb and condensation in the crust, with water transfer
362 from the crumb to the crust. This results in water loss for the crumb and a
363 gain for the crust, and that overall the bread loses water.

364 *4.2. Comparison of the two models*

365 In the first model, the water vapour content is calculated with a diffusion
366 equation, the total pressure is fixed equal to atmospheric pressure, and the
367 dry air content is deduced. In the second model, the water vapour and the
368 dry air content are calculated in the same way, using a diffusion equation.
369 The crust is considered to be a 5 mm-thick area. The equations are the same
370 as in the crumb but the physical properties differ.

371

372 Figures 11 to 15 enable a comparison of the results given by the two mod-
373 els for the crumb (as the crust is represented only by a limit and a boundary
374 condition for the first model, it cannot be used for a comparison). The results
375 are taken at the point $(0, 0)$, the centre of the crumb, and at $(34.9, 34.9)$ just
376 below the crumb-crust interface.

377

378 In Figure 11, the comparison of the two models reveals significant differ-
379 ences. The first model overestimates the local water content compared to
380 the second.

381

382 Similarly, the first model overestimates the water vapour content com-
383 pared to the second, either at the centre or below the crumb-crust interface
384 (Figure 12).

385

386 Conversely, model 1 underestimates the dry air content compared to
387 model 2 (Figure 13) which is expected as dry air and liquid water contents
388 behave inversely.

389

390 The evaporation flow is underestimated by the first model (Figure 14).

391

392 The water loss of the crumb given by model 1 is lower than that given by
393 model 2 (Figure 15).

394

395 In summary, model 1 overestimates the liquid water and water vapour
396 contents, and underestimates the evaporation flow and the water loss of the
397 crumb compared to model 2. These results show the importance of the water
398 transfer from the crumb to the crust, which cannot be calculated with the
399 first model.

400

401 It is worth noting that the model 2, more realistic, based on physical
402 mechanisms (in contrast with purely empirical or semi-empirical models, e.g.
403 using an effective diffusion coefficient that involves several mechanisms) gives
404 us the chance to couple transport models with quality models, for example,
405 to predict rheological properties related to texture and sensory attributes
406 evolution during bread storage, and thus to optimise storage conditions and
407 product formulation. This is because parameters of the model are actually
408 product properties, that is, physical properties of the material. In this way,
409 we could assess different product formulations and their evolution during

410 storage by relating some key ingredients (e.g. starch content, water content)
411 with the material properties.

412

413 Another model intermediate between the two presented here has been
414 tested. In this model, as the first one, the crust was assumed to be a mem-
415 brane, but the water vapour and the dry air content are calculated, as in
416 the model 2, using a diffusion equation. The total pressure was no longer
417 equal to atmospheric pressure but equal to the sum of the partial pressure
418 of dry air and water vapour. The results were strictly equal to those given
419 by the first model (the curves were the same). This is due to the pressure
420 staying constant in the product. The teaching of this model is that when the
421 hypothesis of constant pressure is realistic, there is no need to complicate
422 the model by computing the pressure for the sake of precision, because it
423 involves another equation and additional flow terms.

424

425 Regarding the modelling of the crust, considering that model 2 gives the
426 most realistic results, it is clear that the crust cannot be assumed to be a
427 membrane through which the evaporation flows. Such a model leads to sig-
428 nificantly different results from a model in which the crust is represented as
429 a zone distinct from the crumb. Moreover, it does not allow the water equi-
430 libration phenomenon from the crumb to the crust to be represented. It is
431 necessary to take the crust into account as a zone with its own characteristics.

432

433 Finally, the optimal model, that is to say the simplest model in its equa-
434 tions, without sacrificing to the accuracy of the physical phenomena repre-

435 sented, would be a simplification of the second model by imposing the total
436 pressure equal to atmospheric pressure. The governing equation for the dry
437 air content would be removed as well as the flow term in the water vapour
438 equation. The dry air content would be calculated from algebraic equations.
439 In this way, the crust would be considered a zone with its own characteris-
440 tics. This would provide the simplest possible model, limiting the numerical
441 problems and the computation time, and giving results strictly identical to
442 those given by model 2.

443 5. Conclusion

444 This work has provided results on the water transfer occurring during
445 bread staling and on the techniques of modelling water transfer in bread.

446
447 Concerning the transfer phenomena, the numerical results indicate that a
448 1 cm-thick bread slice lost 1.3 g over 10.2 days, with the crumb losing almost
449 1.8 g and the crust gaining 0.4 g. The missing 0.1 g is probably due to numer-
450 ical errors. Thus, nearly 25 % of the water lost by the crumb was gained by
451 the crust while the rest evaporated into the atmosphere. This water transfer
452 from crumb to crust has already been shown experimentally (Besbes et al.,
453 2014).

454
455 Regarding the modelling of these transfers, the comparison of the results
456 given by the two models shows that it is not necessary to compute the trans-
457 fer for each gas (here water vapour and dry air) by a governing equation with
458 the terms of accumulation, diffusion, flow and a source term, when the total

459 pressure is known to be constant and equal to atmospheric pressure. It is
460 simpler, and probably limits the numerical problems, to remove the governing
461 equation for one of the gases, calculating its partial pressure by subtracting
462 the partial pressure of the other gases from atmospheric pressure. Then, the
463 content of this gas in the product is deduced. Moreover, the flow terms for
464 the other gases disappear from the equations.

465

466 Other information provided by this study is that, for realistic results, the
467 crust cannot be considered a membrane of negligible thickness and charac-
468 terised by its water vapour permeability. The crust is a zone interacting with
469 the crumb for the transfers and considering this crust to be only a Neumann
470 boundary condition gives significantly different results.

471 **References**

- 472 Bechtel, W. G., Meisner, D. F., Bradley, W. B., 1953. The effect of the crust
473 on the staling of bread. *Cereal Chemistry* 30, 160–168.
- 474 Besbes, E., 2012. Dynamique de l'eau dans les matrices céréalières : étude de
475 l'effet des conditions de cuisson sur le rassissement du pain de mie. Ph.D.
476 thesis, Université de Nantes.
- 477 Besbes, E., Jury, V., Monteau, J.-Y., Le Bail, A., 2013. Water vapor trans-
478 port properties during staling of bread crumb and crust as affected by
479 heating rate. *Food Research International* 50 (1), 10–19.
- 480 Besbes, E., Jury, V., Monteau, J.-Y., Le Bail, A., 2014. Effect of baking
481 conditions and storage with crust on the moisture profile, local textural

- 482 properties and staling kinetics of pan bread. *LWT - Food Science and*
483 *Technology* 58 (2), 658–666.
- 484 Datta, A., 2005. Modelling heat and mass transfer in food systems: cur-
485 rent state and needs. In: Pagliarini, G., Rainieri, S. (Eds.), *Heat and*
486 *Mass Transfer in Food Processing - Eurotherm Seminar 77*. Università
487 degli Studi di Parma, Parma, pp. 3–8.
- 488 Datta, A. K., 2007a. Porous media approaches to studying simultaneous heat
489 and mass transfer in food processes. I: Problem formulations. *Journal of*
490 *Food Engineering* 80 (1), 80–95.
- 491 Datta, A. K., 2007b. Porous media approaches to studying simultaneous heat
492 and mass transfer in food processes. II: Property data and representative
493 results. *Journal of Food Engineering* 80 (1), 96–110.
- 494 Hug-Iten, S., Handschin, S., Conde-Petit, B., Escher, F., 1999. Changes in
495 starch microstructure on baking and staling of wheat bread. *LWT - Food*
496 *Science and Technology* 32 (5), 255–260.
- 497 Jury, V., 2007. Transferts couplés masse chaleur d'une matrice alvéolée. ap-
498 plication à la décongélation-cuisson du pain précuit surgelé. Ph.D. thesis,
499 Université de Nantes.
- 500 Ronda, F., Caballero, P. A., Quilez, J., Roos, Y. H., 2011. Staling of frozen
501 partly and fully baked breads. Study of the combined effect of amylopectin
502 recrystallization and water content on bread firmness. *Journal of Cereal*
503 *Science* 53, 97–103.

- 504 Roussel, P., Chiron, H., 2002. Les pains français. Évolution, qualité, produc-
505 tion. MAE Editeurs ERTI, Vesoul.
- 506 Thorvaldsson, K., Janestad, H., 1999. A model for simultaneous heat, water
507 and vapour diffusion. *Journal of Food Engineering* 40, 167–172.
- 508 Whitaker, S., 1977. Simultaneous heat, mass, and momentum transfer in
509 porous media: a theory of drying. *Advances in Heat Transfer* 13, 119–203.

Figure captions

Figure 1: Summary of assumptions of each model

Figure 2: Geometry and Mesh. a: 1st Model, b: 2nd Model. The results were taken at the points shown on the figure. Points represent different zones taken to analyse local profiles, i.e. core, crumb, interface crumb-crust, crust, surface

Figure 3: Comparison of experimental and computed water content profiles: D0 after 2 h, D3 after 3 days, D7 after 7 days and D10 after 10 days of staling

Figure 4: Local liquid water content variation

Figure 5: Water vapour content variation

Figure 6: Dry air variation

Figure 7: Evaporation flow variation

Figure 8: Pores water saturation variation

Figure 9: Overall water content variation in a 1 cm-thick bread slice

Figure 10: Pressure profiles along the line (0, 0) - (55, 55) at $t = 10.2$ days

Figure 11: Comparison of the liquid water content given by models 1 and 2

Figure 12: Comparison of the water vapour content given by models 1 and 2

Figure 13: Comparison of the dry air content given by models 1 and 2

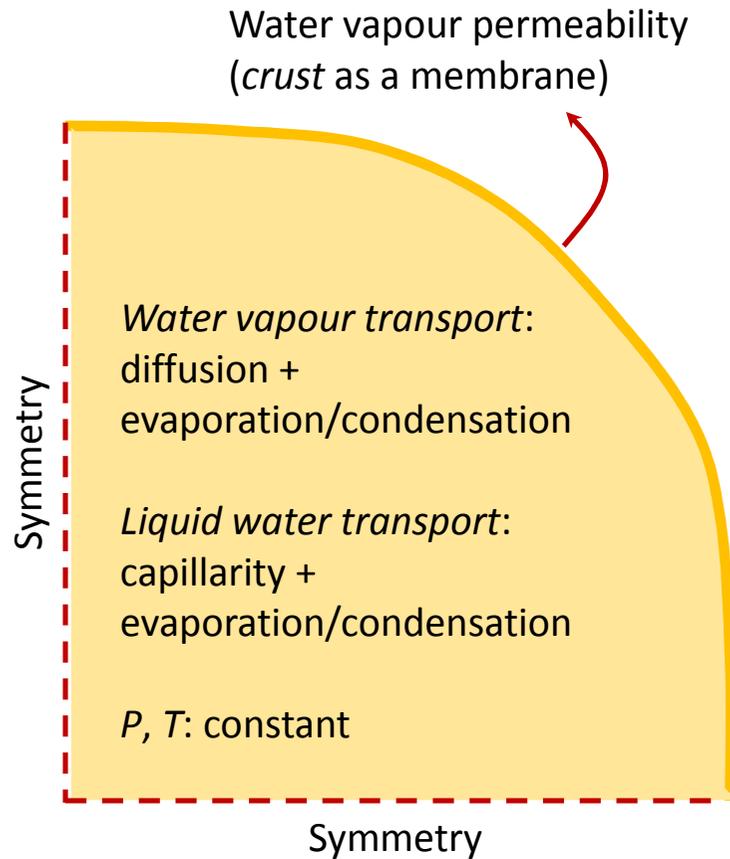
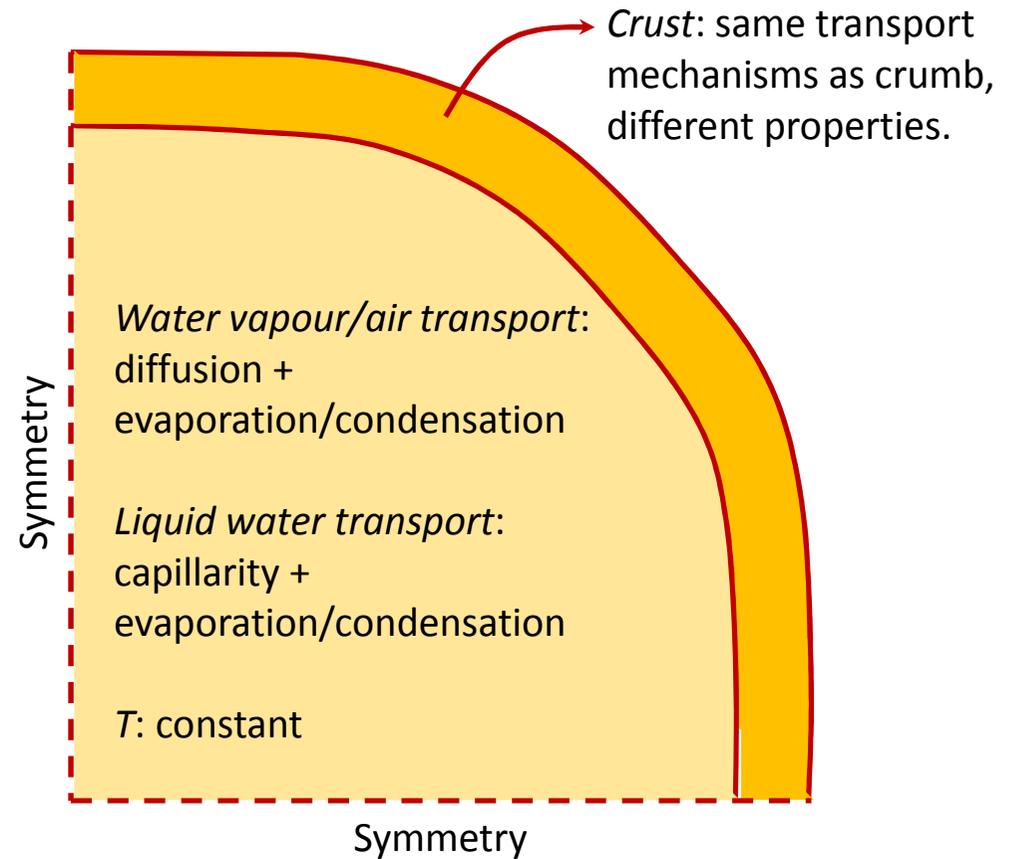
Figure 14: Comparison of the evaporation ow given by models 1 and 2

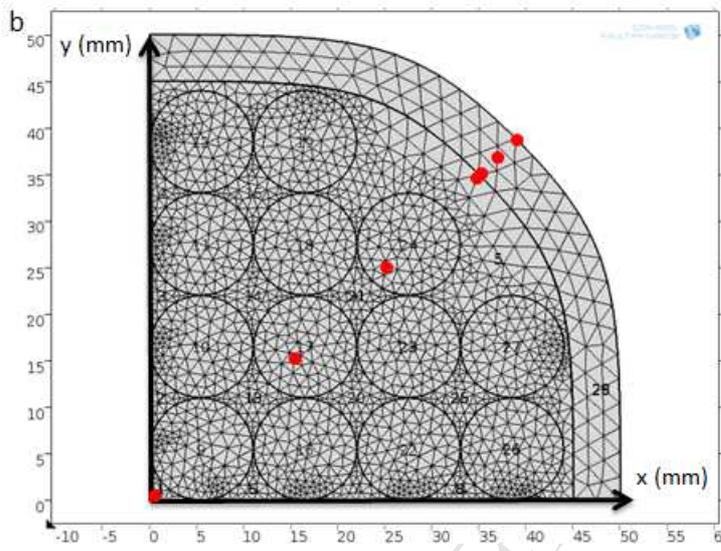
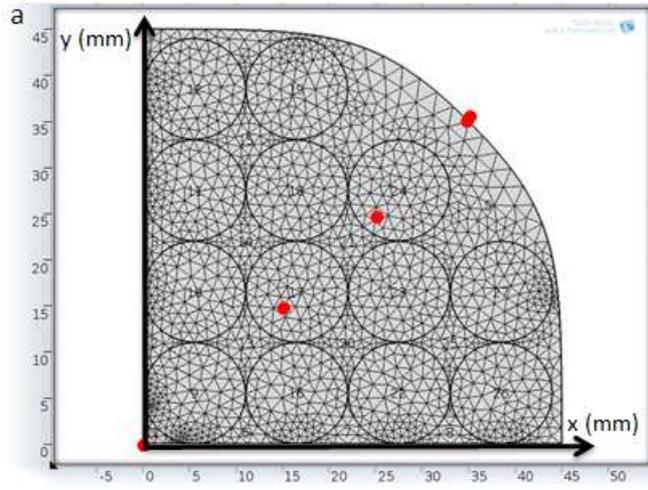
Figure 15: Comparison of overall water content given by models 1 and 2

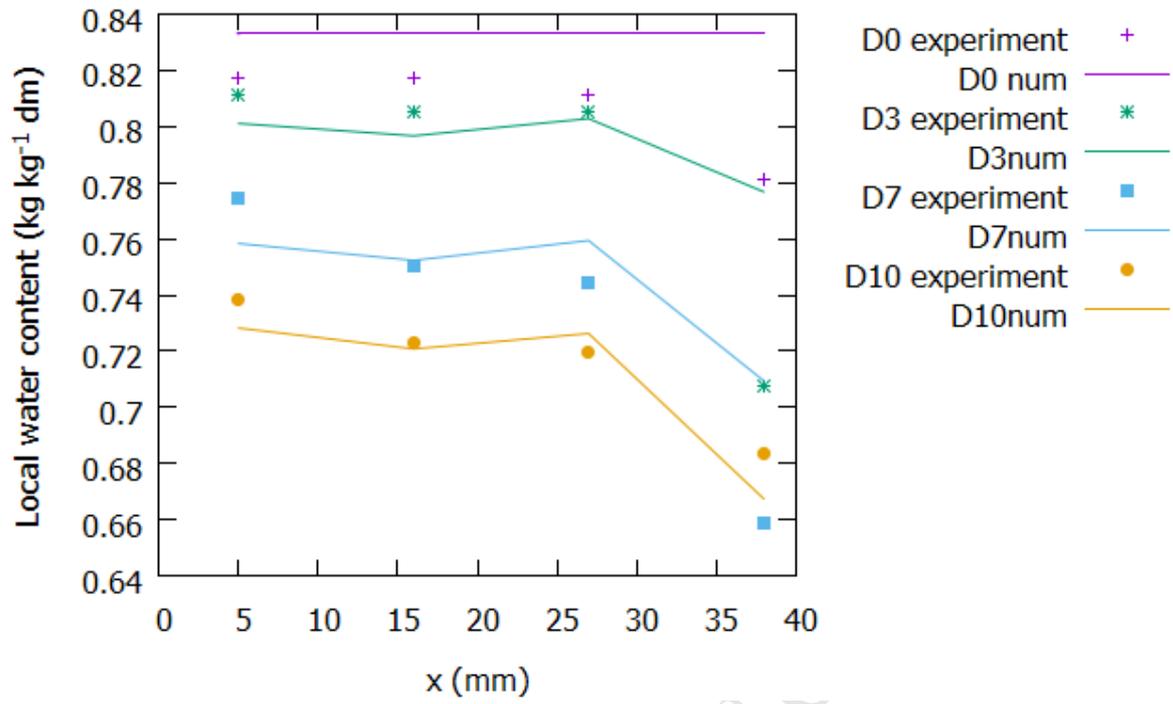
Table 1 : Models parameters

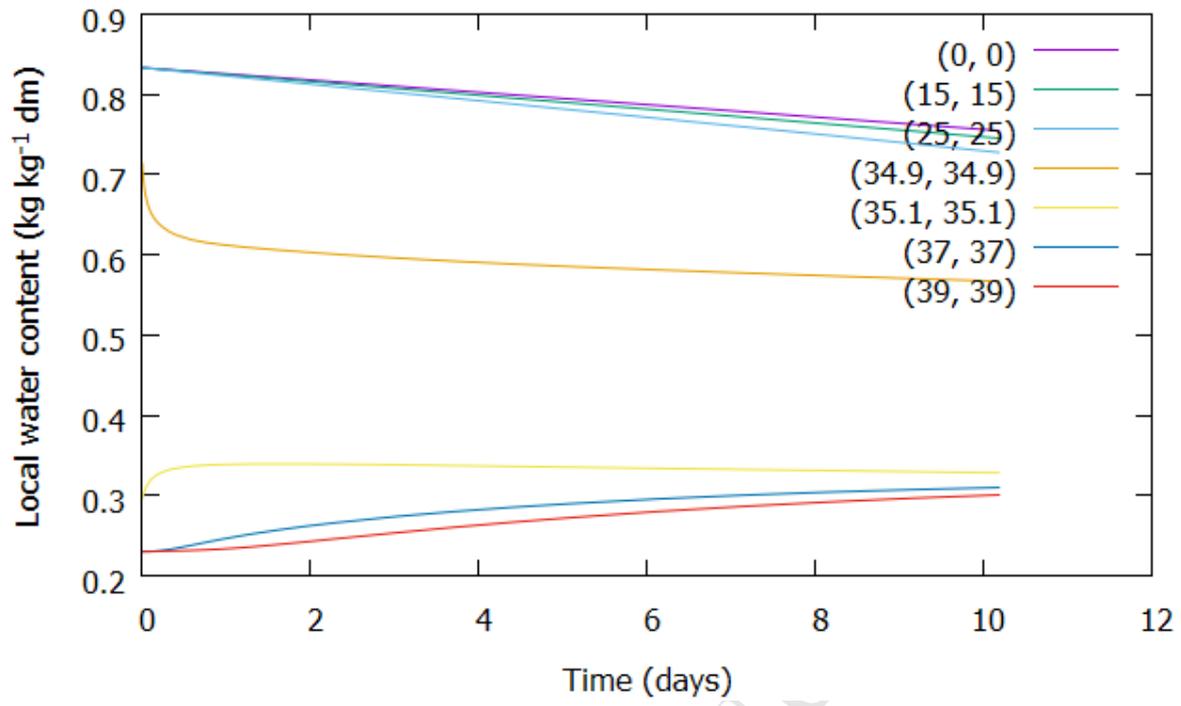
Physical constants		
D_{av}	($m^2 s^{-1}$)	2.34×10^{-5}
R	($J mol^{-1} K^{-1}$)	8.314 5
M_{vap}	($kg mol^{-1}$)	18.02×10^{-3}
M_{air}	($kg mol^{-1}$)	28.96×10^{-3}
ρ_{liq}	($kg m^{-3}$)	998.98
μ_g	(Pa.s)	$1,72 \times 10^{-5}$
Product constants		
D_{liq}^a	($m^2 s^{-1}$)	1.34×10^{-10}
Φ_{crumb}		0.811
Φ_{crust}		0.750
C	($s m^{-2}$)	5×10^{-4}
$\rho_{app,s,crumb}$	($kg m^{-3}$)	192
$\rho_{app,s,crust}$	($kg m^{-3}$)	330
WVP	($kg m^{-1} s^{-1} Pa^{-1}$)	4.67×10^{-11}
e	(m)	0.005
Environmental parameters		
T	($^{\circ}C$)	15
P_{atm}	(Pa)	101 325
RH		0.9

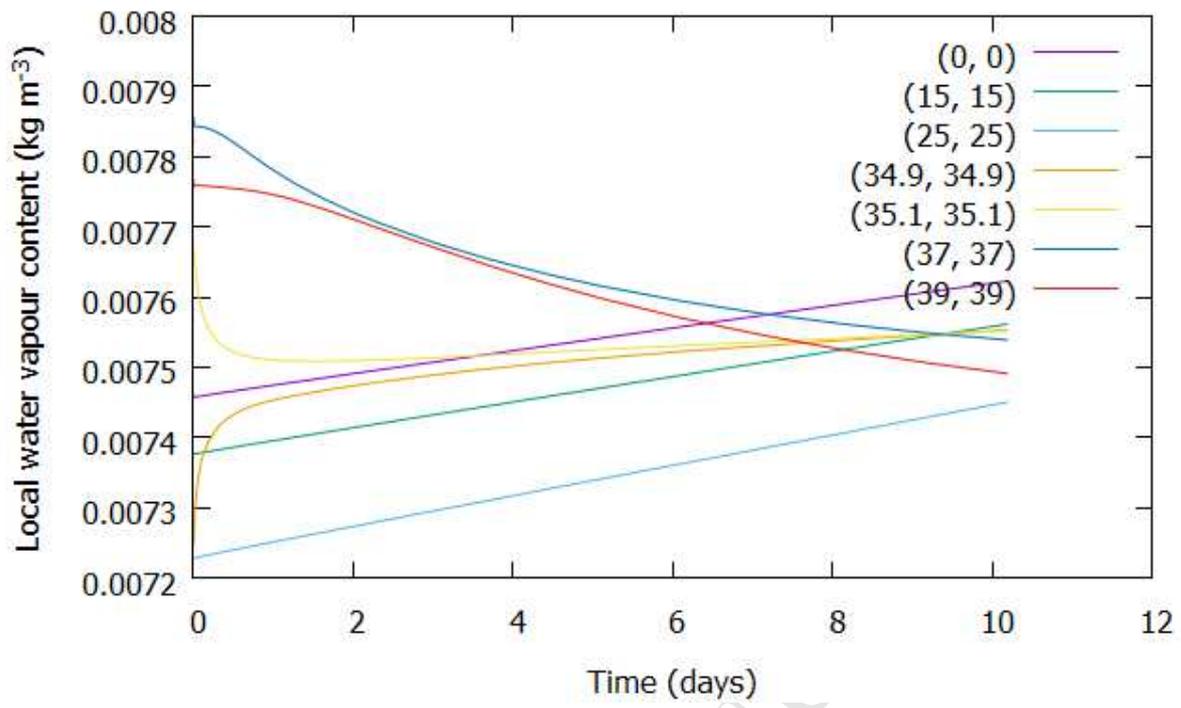
^a 1st model

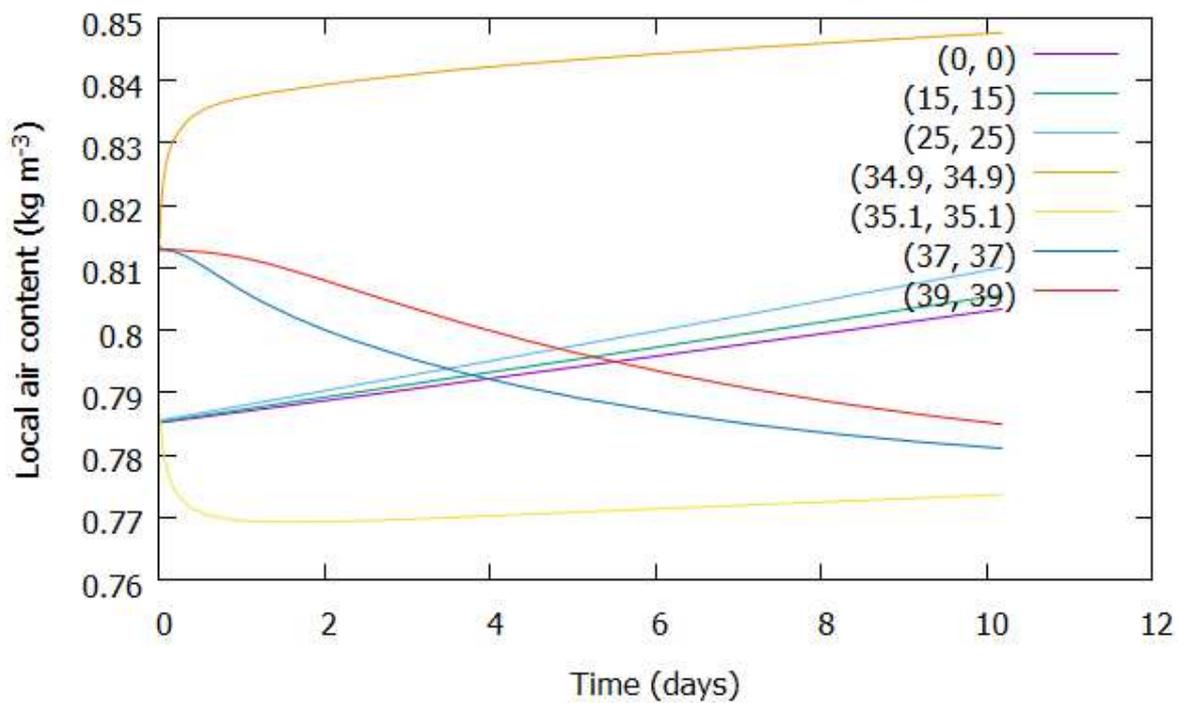
**(a) Model 1****(b) Model 2**

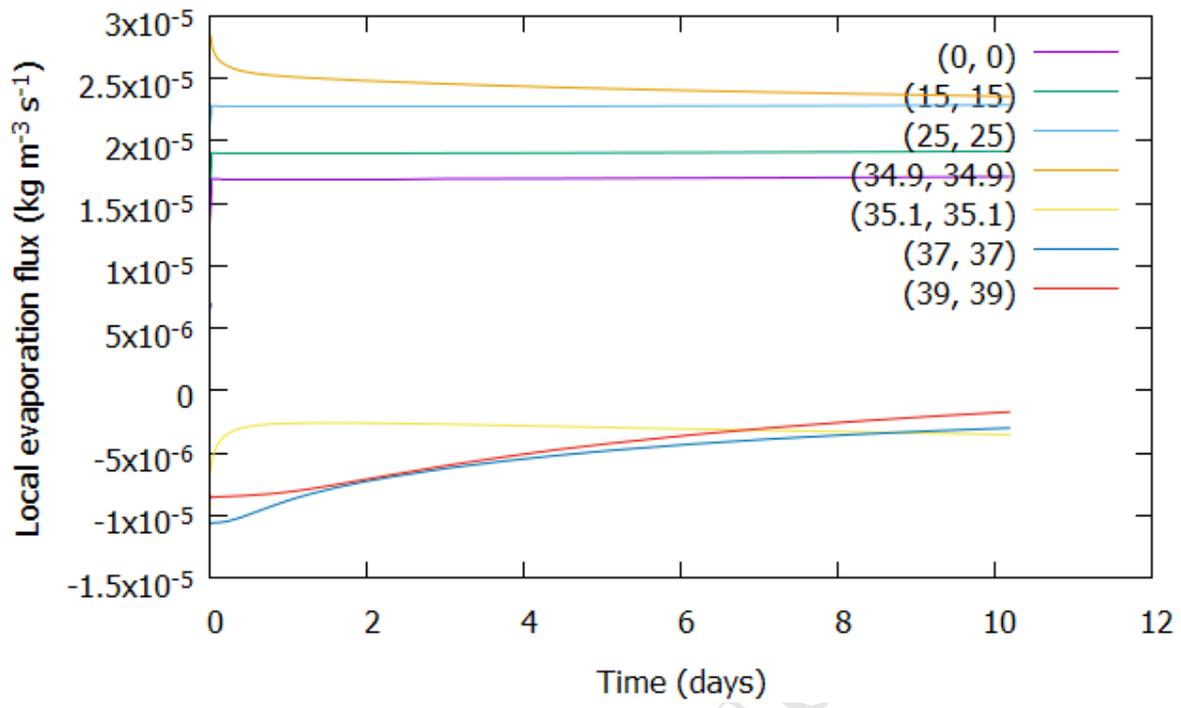


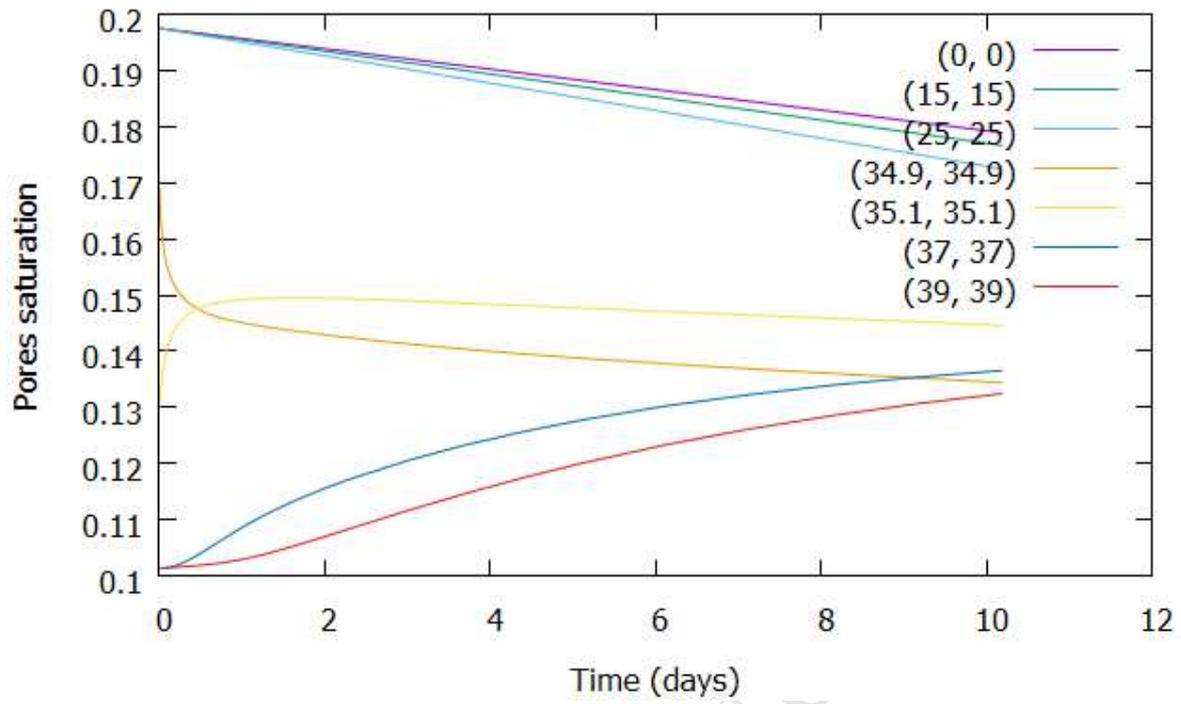


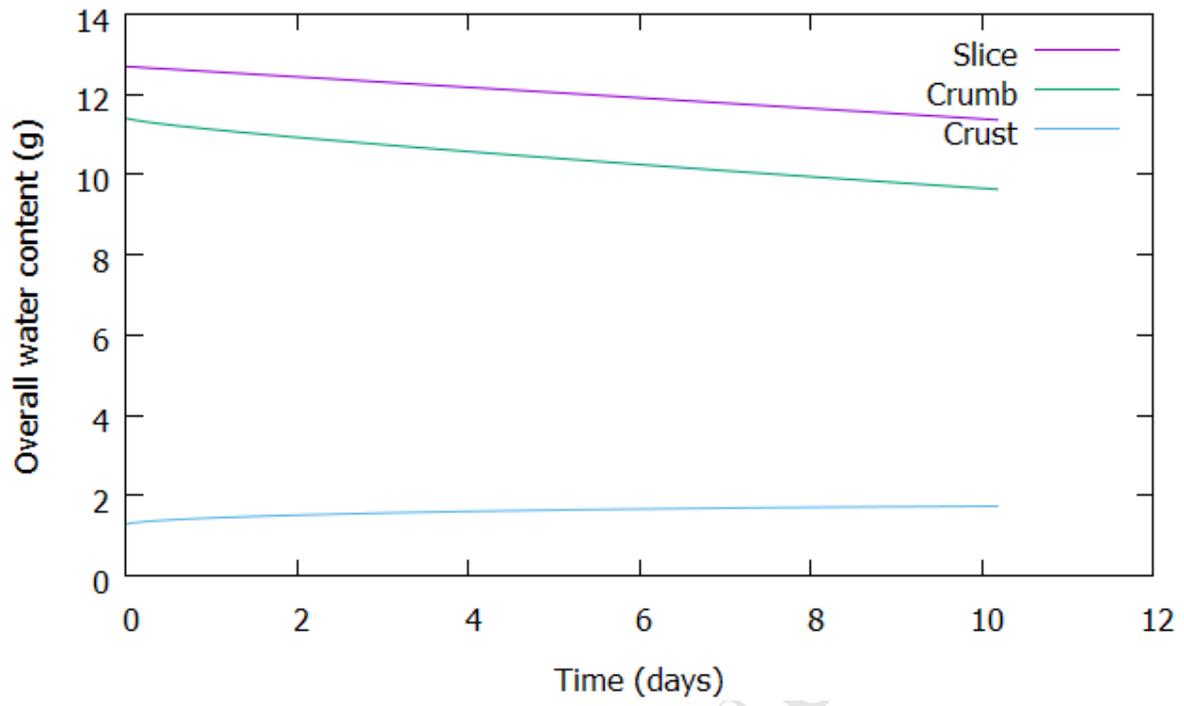


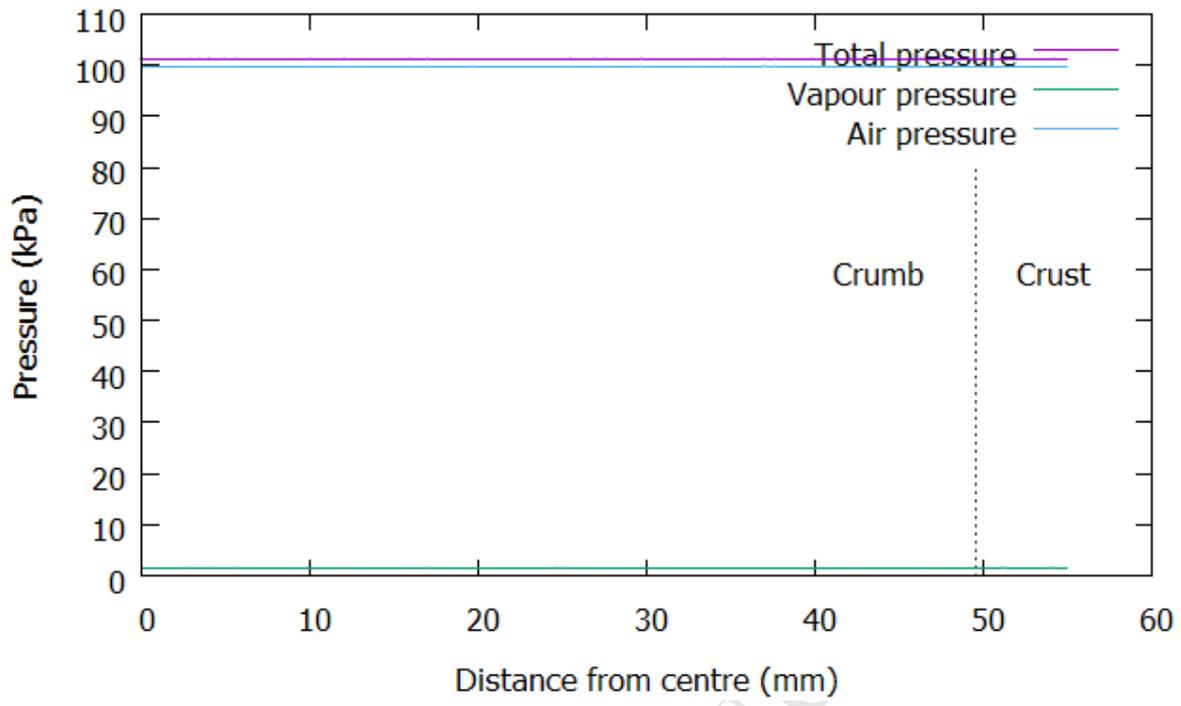


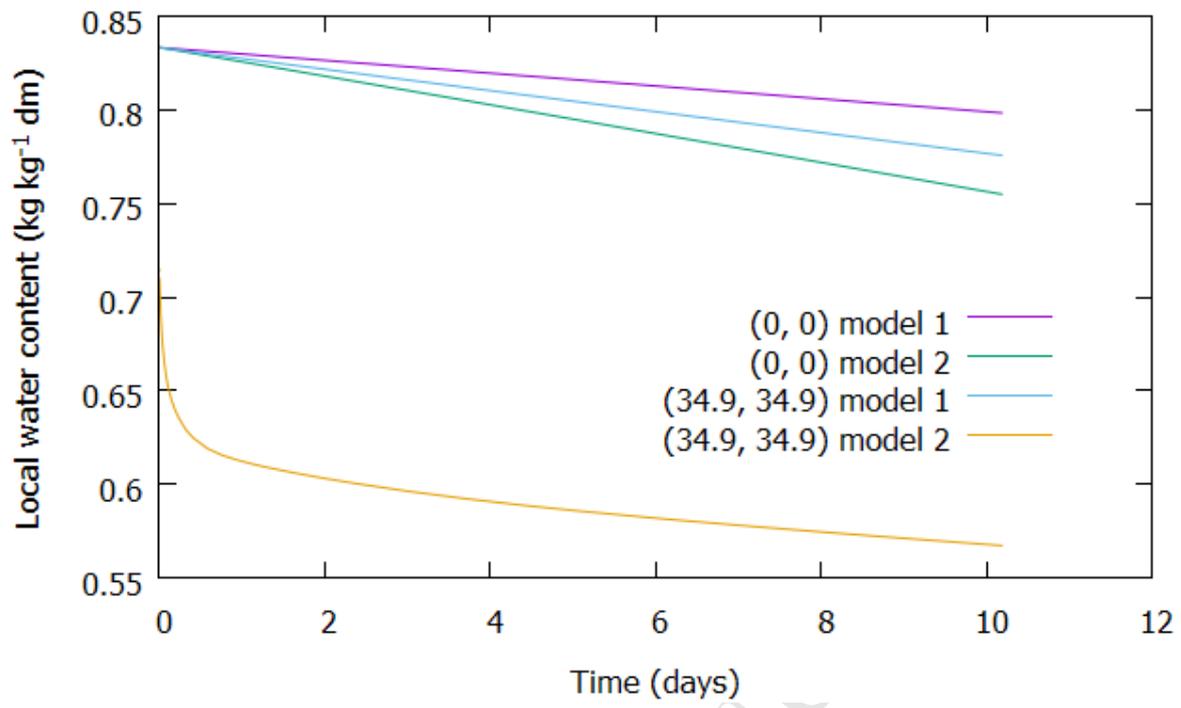


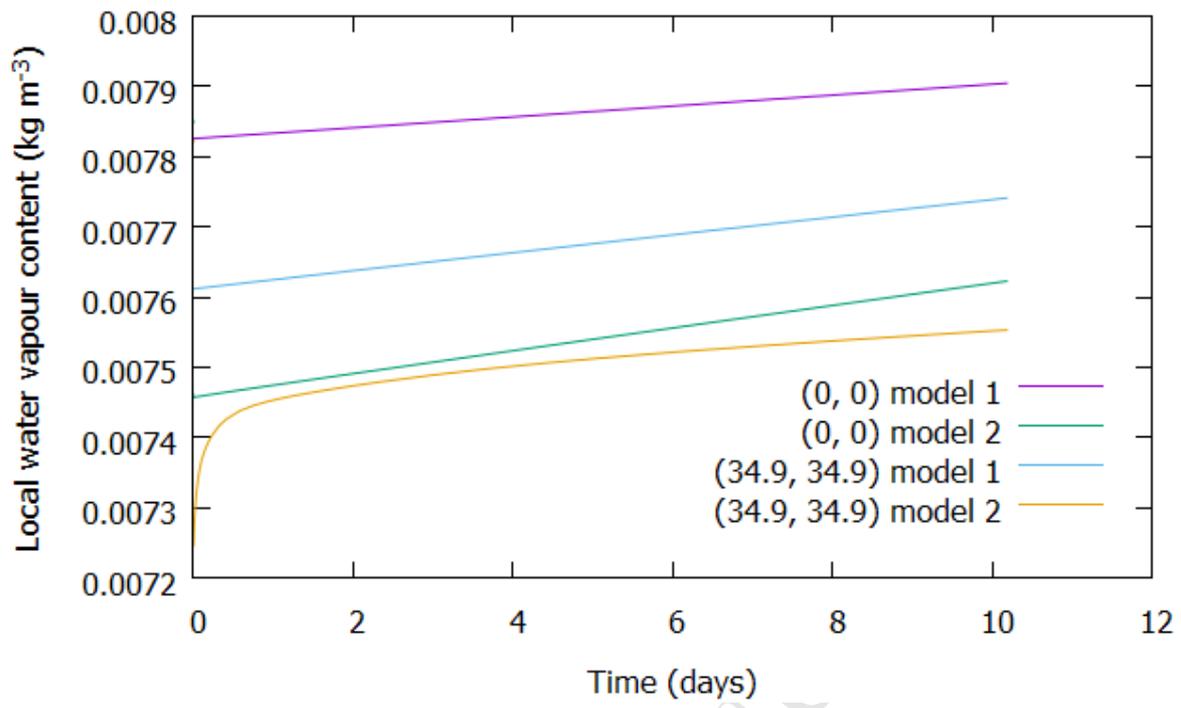


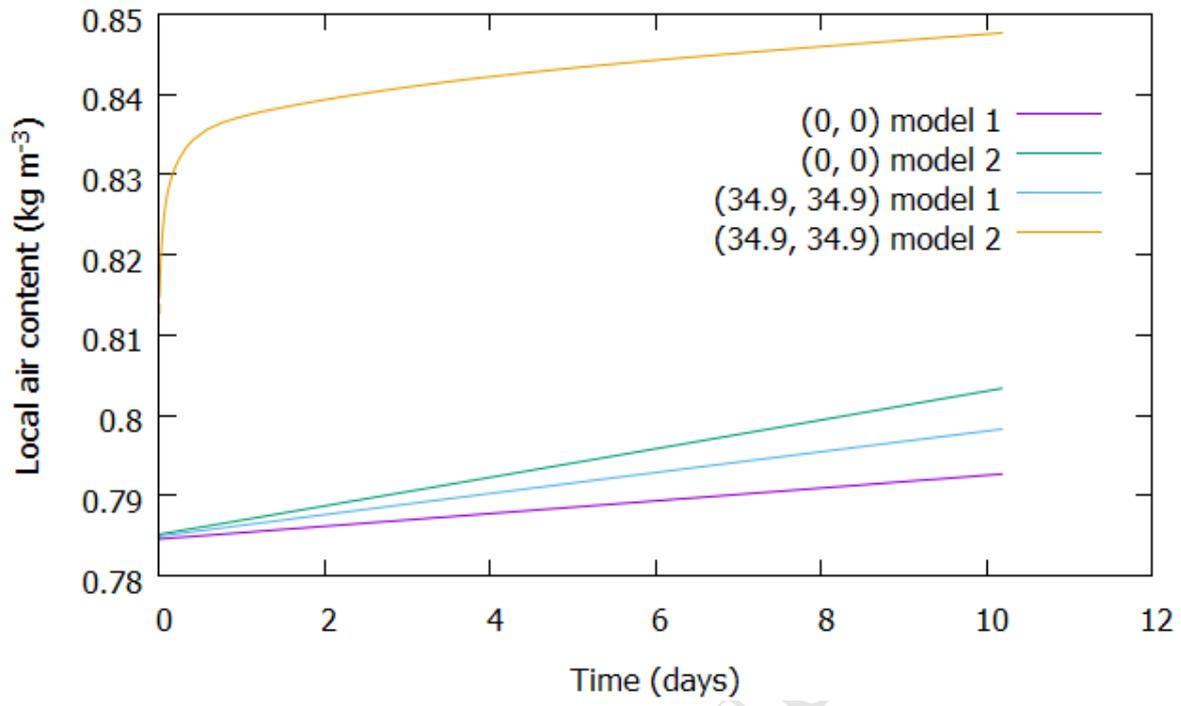


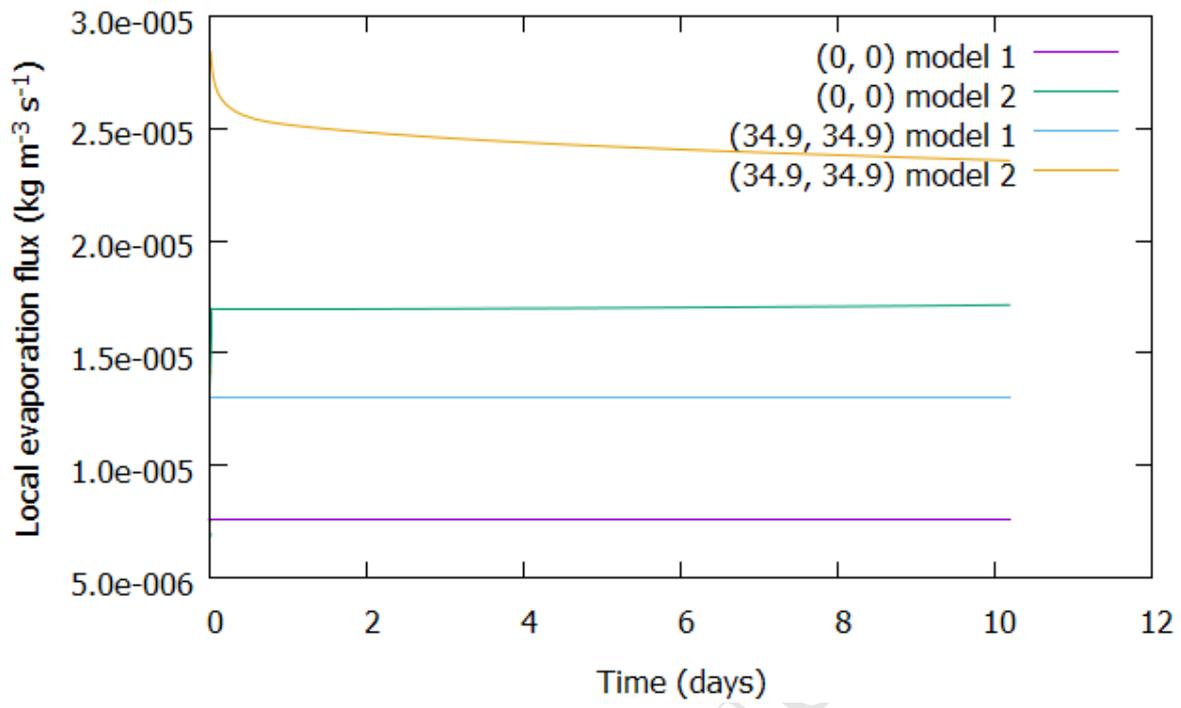


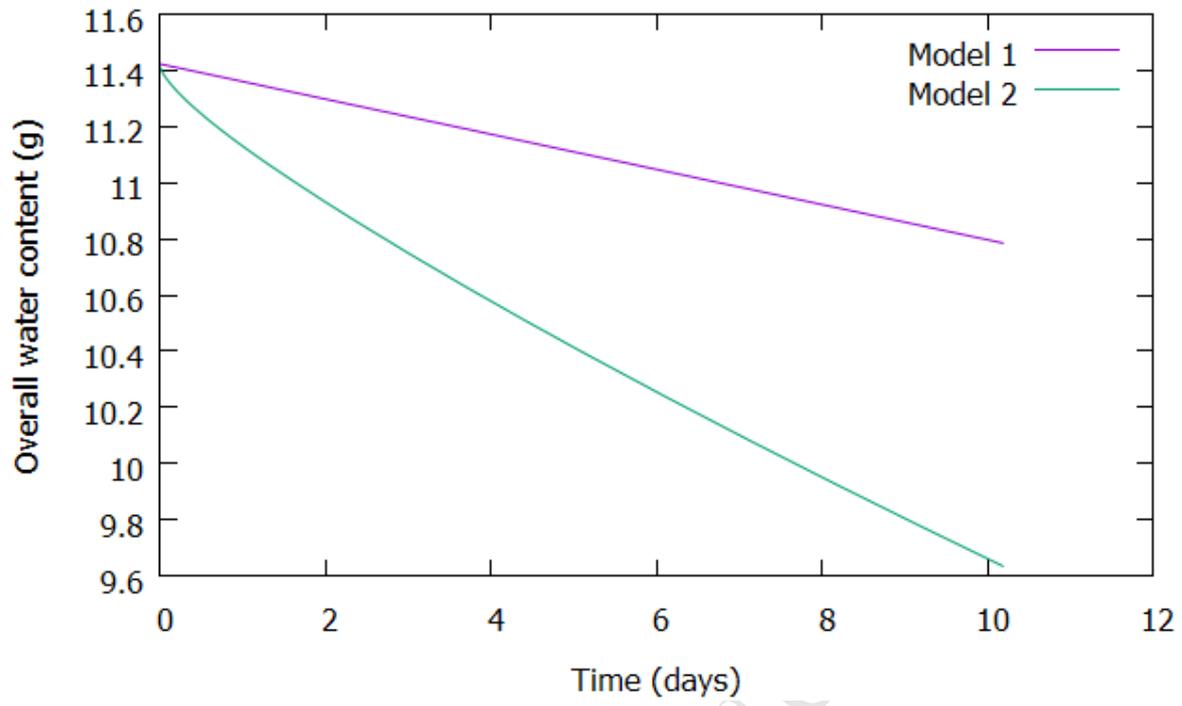












- Liquid water is transferred by capillary diffusion and water vapour migrates by molecular diffusion; Darcy's flow can be neglected.
- There is rebalancing of water from crumb to crust during staling.
- To model the water transfer, considering the crust as a membrane permeable to water vapour is not sufficient.
- Evaporation and condensation also take place during isothermal storage of bread.

ACCEPTED MANUSCRIPT