

## ***Interactive comment on “A new multi-gas constrained model of trace gas non-homogeneous transport in firn: evaluation and behavior at eleven polar sites” by E. Witrant et al.***

**E. Witrant et al.**

emmanuel.witrant@ujf-grenoble.fr

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The authors would like to thank the referee for his/her constructive remarks. We tried our best to answer all the questions raised, as detailed below.

*RC: General: This paper could be an important contribution to the firn air modeling efforts. However the work is presented in a cumbersome style and not easy to follow even for a scientist with good background knowledge in this subject. I suggest to focus on the new aspects of this study, to use references for established ideas and results (e.g. it is not necessary to derive the barometric formula) and to discard unnecessary/unused sections.*

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AC: Assessing the detailed remarks below, it appears that there is a difficulty associated with the multidisciplinary aspect (which is encouraged by ACP) of the paper and its proper presentation for the intended audience. The revised version of the paper will follow the general review suggestions of logical structuring, including only relevant material and writing concisely towards goals. As a solution to this problem we propose to shorten and clarify Section 2, which has been partially considered in the review process, as detailed below.

2.1 The firn network. Revised version of 2.1 focused on the 1D case, including a simplified version of 2.2 and with an appropriate reference to Rommelaere et al. 1997 for the solution of equations (1a, ice lattice) and (1c, gas concentration in closed porosity). The following subsection will focus only on Eq. (1b), the concentrations in open porosity.

2.2 Trace gas transport in the open pores network.

- a The Advective Diffusive Model. The flux term is composed of the airflow speed directly obtained from Rommelaere et al. 1997 (which allows to remove 2.6) and molecular diffusion (simplified 2.5). Limitation: it does not allow to capture the fractional gravitation effect (case  $\zeta = 0$  in former Section 2.9 and Table 1/Figure 1);
- b The quasi-steady state approach. We introduce the gravity effect (Darcy-like behavior) and the fact that the fluxes have to compensate each other at steady-state, which provides the generic definition of  $\mathcal{A}_{ss}$ . This leads to the gravitational fractionation (case  $\zeta = 1$ ). A simplified expression can be obtained by neglecting the difference between the trace gas and airflow speeds (it captures the deviation from the gravitational slope and flattening of  $\delta^{15}N$  and other isotopes of inert gases in the LIZ,  $\zeta = 2$ );

2.3 Turbulent flows in the upper firn. Past 2.7 + definition of the convective region.

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We will make our best efforts to improve the next sections of the manuscript, although precise comments from the referees could have been helpful.

The writing style will be revised and Copernicus copy-editing for English service could be used if necessary after revision.

Derivation of the barometric formula (p 23038 l.24 to p 23039 l.2) will be suppressed in the revised manuscript.

*RC: It is too lengthy and contains substantial misconceptions.*

AC: The length will be reduced as mentioned above. The authors do not agree with the stated misconceptions and hope that the more detailed explanations/proper definitions provided below will help to clear some misunderstandings.

*RC: After struggling through the first 12 pages I decided to stop my review, because I found the status of the paper unacceptable. I do not question the value of the results at this point, but I would wait for such judgment until the paper is in a better condition.*

AC: Our presentation of a multi-disciplinary based mathematical analysis of the modeling problem appears unadapted to the scientific community it wishes to reach. Thus the model description will be simplified and focused on new developments with respect to the existing models. However having both referees refusing to comment the overall manuscript, especially Section 4 (main results) which does not require a full understanding of the model equations, comes as a strong surprise. The authors acknowledge the referee efforts to contribute to the paper improvement and will gladly discuss any further difficulty.

*RC: It should be relatively easy for the many expert co-authors to give good advices.*

AC: Please note that this remark could easily be interpreted as personal criticism of the lead author. We would also mention that the manuscript has been circulated among co-authors, who contributed significantly to the first submission of our work. It will be improved for wording in this revision.

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#### Remarks (1)

*RC: One main problem is that the authors do not present the work in a goal-oriented way. Rather the first 12 pages contain a number of effects, thoughts and statements, subjects partly presented with completely wrong weight, while relevant parts are mentioned in a sub-clause.*

AC: The above proposed re-organizing of Section 2 intends to solve these issues.

*RC: For example there is a lengthy discussion about hydrostatic equilibrium that is not reached due to viscous flow. There is no discussion that the velocities discussed here are in the order of cm per year, which should be compared to time constants of viscous flows.*

AC: This remark may result from a misunderstanding as the term "viscous flow" is not used in the paper. The fact that hydrostatic equilibrium is not reached is introduced l.3-7 p 23039: it is due to firn sinking and gas trapping. The model implementation is described p 23044 l 23 to p 23045 l.10 and illustrated by model tests in section 2.9.

The "time constants of viscous flows" would need a more precise definition to be considered, as viscous flows formally appear (e.g. in the classical Navier-Stokes equations) in the momentum and energy conservation, which are not directly considered here. Considering the time constants associated with Darcy's law would require the knowledge of the firn permeability, which we do not have precisely, and wouldn't really fit with its use as a steady-state constraining case, as it is done in this model.

The LIZ flattening effect appears when the trace gas velocity is equal to the air velocity (see p 23045) and is thus important in the deep firn to avoid a manual tuning of the LIZ (an interesting contribution of this model). The revised manuscript will clarify and simplify our (quasi steady-state) use of Darcy's law.

*RC: There is no discussion on the effects of temperature and surface pressure varia-*

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tions that are neglected.

AC: These fundamental assumptions are classically done in models of gas transport in firn (e.g. Buizert et al. 2011), based on the fact that temperature and surface pressure (and wind) variations act on a faster time-scale than the modeled processes, affecting the upper part of the firn.

A simplified representation of these processes is introduced p 23032 l.10-12, discussed in section 2.7 and illustrated by tests in section 2.9. They can be represented by an “eddy” component of the diffusivity, possibly explaining the fact that the calculated values of the effective diffusivity in the upper firn can exceed the molecular diffusivity in free air.

At longer time scales (seasonal) temperature also affects trace gases in the upper firn through thermal diffusion. The fact that our model does not include this process is stated e.g. p 23059 l. 10-11. These processes will be discussed in our revised Section 2.

RC: *On the other hand on page 23041, first line, the most important trapping rate of air bubbles is given in an in-line equation, without any comments. This trapping rate is very debatable.*

AC: The trapping is described p 23041 l. 1-6 where the reference model (Rommelaere et al. 1997) is quoted as well as a possible other model. Additional references on the trapping rate are welcome. We did not identify a bias in our model results that could be attributed to an incorrect formulation of the trapping in Rommelaere et al. 1997.

As the trapping rate is not an original development of this study and to fulfill the simplification objectives, the section on air transport will be removed.

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Remarks (2)

RC: *Reaching eq. 3 after 10 lengthy pages, the discussion on viscous non-equilibrium is suddenly terminated by “A direct approach to this problem would require a knowledge of the firn permeability (scaling laws such as those proposed by Schwander (1989) or Freitag et al. (2002) could be used for this purpose) and would require solution of a nonlinear BVP, which would necessitate a dedicated mathematical analysis (our tests using classical commercial solvers failed to provide satisfactory results).” I guess most readers will be confused here if not earlier.*

AC: Introduction and sections 2.1 to 2.3 do not specifically focus on Darcy’s law (eq. 3, resulting from the 3 previous lines).

The discussion is not terminated here but introduces our alternative approach described in Section 2.8 (see e.g. p 23044 starting l. 11) and illustrated in Section 2.9.

Our simplified use of Darcy’s law (quasi steady-state), which aims at introducing external forces (gravity) rather than representing viscous flows will be clarified in the revised manuscript.

Remarks (3)

RC: *Then follows a variation approach (deviation from hydrostatic equilibrium). Does the superscript o mean hydrostatic equilibrium? (it is not well explained!).*

AC: The subscript o refers to open pores, as defined p 23035. The hydrostatic equilibrium is denoted as  $\bar{p}$ , as defined p 23038, l.25

RC: *If so, is the almost trivial solution for w(air) given at the bottom of page 23041 the outcome of the first 10 pages?*

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AC: No, it comes from the equation I. 14 p 23041 (10 lines above). Section 2.6 will be removed in the revised manuscript, as explained before.

Remarks (4)

*RC: There are a number of misconceptions and errors: Page 23031, line 12: "...and an almost-stagnant behavior described by Darcy's law (gravity effect)." A stagnant behavior is not described by Darcy's law.*

AC: The Darcy-like behavior is used here to introduce external forces, and more precisely the gravity effect (as done in the main cited reference on poromechanics: Coussy p 59 for quasi-static flows). The stagnant behavior, as defined p 23038 l. 11-13, implies that the different fluxes are at equilibrium. For nonisobaric conditions, Darcy's law needs to be combined with a diffusion model to include the gravity effect. This is described for example at p 17 in:

Scanlon, B. R., J. P. Nicot, and J. W. Massmann, 2002, Soil gas movement in unsaturated systems, in Warwick, A. W., ed., Soil Physics Companion, p. 297-341. [http://www.beg.utexas.edu/enviro/qly/vadose/pdfs/webbio\\_pdfs/soilphysicscompanion2002.pdf](http://www.beg.utexas.edu/enviro/qly/vadose/pdfs/webbio_pdfs/soilphysicscompanion2002.pdf)

*RC: Page 23037, line 11: What is the purpose of the filtration vector? It is not used further in the manuscript. It is rather unusual to speak of a flow speed  $w(x)$  in a diffusive mixing environment.*

AC: The purpose of the filtration vector is to separate trace gas transport due to air transport  $\phi_{air}$  from trace gas transport within the air column  $(\phi_{\alpha_i} - \phi_{air})$ . This allows to separate the analysis of the phenomena associated with the flow speed (sections 2.4 and 2.6 for  $\phi_{air}$  only) from those associated with molecular diffusion  $((\phi_{\alpha_i} - \phi_{air})$  in section 2.5).

In the paper, the filtration vector is mostly used to explain the structure of the classical

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advective-diffusive model (ADM). It will be removed from the new version, where we can start from the ADM and simply note the associated limitations.

*RC: line 16: Eq. 2 does not follow from the previous equation. This can be check e.g. by substituting the variables with suitable numbers. The final eq. is not a solution for  $\phi_i$  as suggested by "The specific filtration vector is then.." ( $\phi_i$  is still on both sides of the equation).*

AC: Eq. 2 does follow from the previous equation and the mole fraction definition, except for a sign mistake (which does not change the result obtained after the approximation but has to be corrected indeed). To see this, first note that the sum of the mole fractions is one:

$$\chi_{air} + \sum_{j=1}^{N_g} \chi_{\alpha_j} = 1$$

which implies:

$$\begin{aligned} \phi_{\alpha_i} &= \left( \chi_{air} + \sum_{j=1}^{N_g} \chi_{\alpha_j} \right) \phi_{\alpha_i} \\ &= \left( \chi_{air} + \sum_{j \neq i} \chi_{\alpha_j} \right) \phi_{\alpha_i} + \chi_{\alpha_i} \phi_{\alpha_i} \end{aligned} \quad (1)$$

Eq. p 23037 l.13 writes as:

$$\begin{aligned} \phi_{gm} &= \chi_{air} \phi_{air} + \sum_{i=1}^{N_g} \chi_{\alpha_i} \phi_{\alpha_i} \\ &= \chi_{air} \phi_{air} + \sum_{j \neq i} \chi_{\alpha_j} \phi_{\alpha_j} + \chi_{\alpha_i} \phi_{\alpha_i} \end{aligned}$$

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Substituting the term  $\chi_{\alpha_i} \phi_{\alpha_i}$  with its value in (1) gives:

$$\phi_{\alpha_i} = \left( \underbrace{\chi_{air} + \sum_{j \neq i} \chi_{\alpha_j}}_{\approx \chi_{air}} \right) \phi_{\alpha_i} + \underbrace{\phi_{gm}}_{\approx \phi_{air}} \underbrace{-\chi_{air} \phi_{air} - \sum_{j \neq i} \chi_{\alpha_j} \phi_{\alpha_i}}_{\approx -\chi_{air} \phi_{air}}$$

Our goal is not to make  $\phi_{\alpha_i}$  disappear but to separate the advection with air  $\phi_{air}$  from the gas movement within the air expressed by  $\chi_{air}(\phi_{\alpha_i} - \phi_{air})$

*RC: Page 23039, line 12: This equation is only valid for a single trace gas in air. This should be stated, since previous equations and the title of the paper suggest a multi-trace gas discussion.*

*AC: The multi-trace gas approach refers to Section 3, which describes the use of multiple gases to constrain the inverse diffusivity model rather than physico-chemical interactions between trace gases. The hypothesis that trace-gases do not affect each other is stated p 23037 l. 18. The hypothesis of non-interacting diffusions is stated and discussed p 23038 l. 6-10.*

*RC: Page 23040, line 6: "Note that, while Fick's law is reasonably accurate to describe the molecular diffusive flux for a nonstagnant gas, it was found to be totally inadequate for a stagnant gas (considered as a gas without source or sink, thus at steady state), as discussed by Thorstenson and Pollock (1989). Note also that Fick's Law is adequate for a trace gas, but for a major gas the Stefan-Maxwell equations (diffusion in a multicomponent 10 system) must be used. This implies, in the framework of trace gas transport in firns, that Fick's law is suitable to describe the dynamics of gases with large atmospheric variations in nonstagnant firn regions but should be used with care otherwise (small concentration gradients or limited airflow in deep firn)".*

*There is a serious misconception here. Stagnant gases described in Thorstenson are*

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*not the same as stagnant firn regions. Fick's law is precisely the physics to describe (trace gas!) fluxes in a stagnant environment. This has been proven valid in many studies.*

*AC: Scanning the manuscript for our use of the expression "stagnant", we realized that it has two different meanings. This is confusing and will be corrected. Stagnant gas as defined p. 23038 l. 11-13 refers to a null flux as in Thorstenson, which implies  $[\rho_{\alpha}^o f]_t = 0$  (steady-state), the reference frame being the Eulerian coordinate. On the other hand, a "stagnant firn region" refers to slowed gas transport processes in deep firn. This is incorrect in the frame of a moving ice lattice and needs to be clarified. As an additional interesting reference, Scanlon et al. define a stagnant gas as "zero flux, no sources or sinks, no reactions", which excludes the deep firn due to the sink induced by gas trapping. In the above extract, we refer to a stagnant gas behavior defined as in Thorstenson. The fact that Fick's law is not sufficient is due to the non-isobaric condition, it applies to the diffusive zone of the firn for gases at steady state (e.g. isotopes of inert gases) for which concentration gradients are dominated by the effect of gravity (as illustrated p 23047 l. 16-18 along with Tab.1 and Fig. 1).*

*RC: If Fick's law is inadequate e.g. in the close-off zone then it is not due to stagnant conditions but rather due to flows induced by compaction of almost close firn layers.*

*AC: The authors agree with that, and the hypothesis  $w_{\alpha} = w_{air}$  is of prime importance to capture the LID (see p 23045 l.6-9, p 23046 l.1-3 and p 23047 l. 24-29 along with Tab. 1 and Fig. 1). This point will be emphasized.*

Remark (5)

*RC: The manuscript is loaded with "empty phrases", which is another reason that makes the paper too long. E.g. page 23034, line 16: "Several issues associated with the transport characterization are detailed, as well as simplifying hypotheses that*

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*allow the use of this model for multigas diffusivity optimization.”???*

AC: This transition sentence will be removed. Efforts will be made to suppress other unnecessary sentences such as p 23032 l.22-25, p 23033 l. 17-19, p 23034 l. 12-18, etc.

Remarks (6)

*RC: By checking further sections of the manuscript this cumbersome style unfortunately carries on. E.g. page 23046, line 7: “The choice of  $Ass = 0$  below  $z_{lid}$  implies that the internal and external forces have a negligible impact on trace gases in the vertical direction, which is the case if the surface stress tensor or non-vertical pressure gradients dominate the gravitational force in the LIZ. The stress tensor is induced by the firm deformation gradient (Coussy, 2003) and local pressure gradients that may appear if thin walls enclosing a pressurized bubble break, two phenomena associated with the bubble closure process.” It is difficult to guess the meaning of this if one does not already have a good background in the matter.*

AC: This remark was intended to pinpoint neglected processes that would require the use of momentum equation (to include the stress tensor). As no existing firm model takes these processes into account, these two sentences will be suppressed.

When put in parallel with the comment about neglected effects of temperature and surface pressure, the choices related to commenting neglected processes while preserving conciseness appears as not trivial.

The “cumbersome style” in the above two sentences also reflects different writing styles of different scientific communities and is adapted to the mathematical/physical community of the lead author. Efforts will be made to adapt the writing style to the ACP reading community.

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Interactive comment on Atmos. Chem. Phys. Discuss., 11, 23029, 2011.

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