Review of Trudinger et al. ACPD 2012

This is an excellent paper that makes an important contribution to better understanding of the true constraints that observations of firn gases place upon the effective diffusivity of the firn profile. Particularly helpful are the findings that some gases are more useful than others, which could guide future sampling campaigns. In general the paper is well written, however it could be more tightly focused to improve readability. Some points need clearer and better explanation. Other points could possibly be dropped to conserve space. On the whole it is a welcome contribution to the field of firn air studies and the closely allied field of ice core studies of trapped gases.

Specific remarks:

Page 17779, line 24

The choice the authors make, to restrict their search for diffusivity profiles to those with a monotonically decreasing diffusivity with depth, begs the following question.
A monotonically decreasing diffusivity with depth may be easier to deal with, but is it realistic? One can easily imagine a melt layer creating a local minimum in diffusivity. Not that the authors should abandon the monotonic choice, but at least they could say a few words about the drawbacks of this choice and why it is potentially unrealistic.

Also, it is a bit confusing to the reader to say first that "We specify diffusivity as a function of open porosity", and then to describe a choice regarding "diffusivity as a function of depth". Does this imply that open porosity must also be a monotonically decreasing function of depth? Perhaps it would be clearer if you make the implicit explicit here?

It is not clear if you insist that the diffusivity-porosity relation be the same at all sites, or if it is allowed to be site-specific. Please clarify.

Another, perhaps related, issue is that plate-like grain microstructures have much lower diffusivites than spherical-grain microstructures, for a given open porosity. Is it a wise choice to limit the diffusivity to being a function of only open porosity? What if there are discrete layers of wind-packed plate-like grains, separated by layers of spherical grains? Forcing the diffusivity to be a function of only open porosity will cause an error in this case. I suppose one could make the diffusivity be a function of two independent variables, the open porosity and a shape factor, which is an adjustable parameter found through fitting to data. [Theoretical relationships then could be employed to explore the relation between the shape factor and the tortuosity].

Somewhere in this discussion it should be mentioned that we use the term "diffusivity" for convenience, and it is better described as the "effective diffusivity". For the diffusive part of the column, we really mean the "inverse tortuosity multiplied by the free-air diffusivity of CO_2 at the temperature and pressure of the site at the time of sampling", and so the physically meaningful parameter we seek is really the tortuosity, a site-specific property of the porous medium that should be independent of gas type, temperature, and pressure. [The tortuosity's physical meaning can be thought of as the distance actually traveled by the average molecule, divided by the distance it would travel in free air, and the tortuosity is thus a dimensionless parameter]. Making this distinction will be helpful to non-specialist readers.

In the convective zone, the effective diffusivity cannot easily be related to the tortuosity alone because mixing is greatly enhanced via macroscopic stirring. The impact of stirring can be described in a one-dimensional model by an "eddy" diffusivity (a concept well developed by physical oceanographers – see Young, 1999). The effective diffusivity is best thought of as the sum of the molecular and eddy diffusivities, with the molecular part related to the tortuosity, and the eddy part related to the vigor of stirring. Importantly, eddy diffusion affects all gases similarly (because macroscopic air movement advects all gases together in the viscous flow regime), whereas molecular diffusion does not, so the separation of the two components of effective diffusivity is needed for maximum relative accuracy in model comparisons of different gases.

At present, the recommended pragmatic approach for firn air studies at sites with substantial convection (e.g. Severinghaus et al., 2010) is to estimate the tortuosity in the convective zone from empirical tortuosity-porosity relationships (e.g. Schwander et al., 1988; Fabre et al., 2000) and the observed porosity in the convective zone. Then, $\delta^{15}N$ observations in the deep part of the diffusive zone and model tuning are employed to estimate the eddy diffusivity profile at the site (which is influenced by both firn structure and wind speed; Kawamura et al. 2006). The reason that $\delta^{15}N$ is useful for this purpose is that it is nearly insensitive to tortuosity, because it is a quasi-steady-state tracer with constant atmospheric value, yet it responds strongly to the destructive effects of eddy diffusion on molecular separation due to gravitational and thermal fractionation. [As a check on the realism of the tortuosities found in this fashion, the tortuosity should never be less than 1 (which is unphysical) and the molecular diffusivity should never exceed the free-air molecular diffusivity.]

The fundamental physical reason that molecular diffusion can separate gas mixtures, whereas eddy diffusion cannot, is that eddy diffusion involves macroscopic turbulence. The typical length scales of the air filaments and parcels involved in this process are of order 10^{-3} to 10^{+1} m. In contrast, the molecular scale is of order 10^{-7} m (mean free path at typical firn temperature and pressure). Thus turbulent mixing involves parcels at least 10^4 times larger than the molecular scale. These parcels contain $>10^{16}$ molecules, all moving more-or-less together in a bulk (viscous) flow.

Minor comments:

Table 1: the measured firn temperature at Pole was -51˚C (Severinghaus et al., 2001). You have -49˚C here; perhaps that was the air temperature?

References cited in this review:

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