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Comment

## ***Interactive comment on*** “Theory of isotope fractionation on facettted ice crystals” *by* J. Nelson

**Anonymous Referee #1**

Received and published: 28 July 2011

Paper: Theory of Isotope Fractionation on Facettted Ice Crystals

Authors: Nelson

doi: 10.5194/acpd-11-17423-2011

Summary:

The author proposes that surface processes on facettted ice crystals could influence their isotopic fractionation and also (though to a weaker extent) the isotopic fractionation of precipitation records in ice and snow cores. These effects have not been previously considered to my knowledge and represent a significant contribution, even if some aspects of these processes are still unknown at the present time.

Recommendation:

Minor revisions. I would suggest that the paper is nearly ready for publication, and

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would ask the author to consider the comments below during revisions. There will be a lot of people reading this paper who will not be familiar with the author's previous work (especially Nelson and Baker, 1996) and will want to parameterize this effect approximately in their models. As a result, most of my suggestions ask for increased exposition (probably only a sentence or two in each case), so that these readers better understand the context of the mechanism proposed in the paper and how to approximate its effect in a model.

Major comments:

1. The main weakness of the paper is that the surface impedance of the water isotopologues is unknown and can only be guessed at in the present manuscript. I think of the present manuscript as framing the issue of surface fractionation effects and leaving the determination of its sign and magnitude to future work. While this is a limitation of the present paper, I still believe that it is worthy of publication in ACP.

It would be useful if the author could give some intuition as to the sign of this effect and its likely magnitude, if this is possible. Could one make an argument that the surface impedance of the heavy water might be lower than that of standard water because the heavy molecules would be less likely to leave the face of the crystal before attaching to a surface step? Any such arguments/intuition would be appreciated, including a justification for thinking on p. 17438, line 5, that the heavy water migration distance curve would be similar to that of standard water but shifted in temperature from it.

Minor comments/suggestions (all page numbers start w/174):

A note on language: The word "isotope" refers to atoms, as in deuterium is an isotope of hydrogen. "Isotopologue" refers to a molecule which combine different isotopes, so that HDO or H<sub>2</sub>O<sup>18</sup> are properly spoken of as isotopologues of water. See, e.g., the discussion at the beginning of section 2 of:

Nassar et al. JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 112, D21305,

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The adjective is "isotopic", so that the title should have "isotopic fractionation", rather than "isotope fractionation".

p. 25, line 7: Could a reference be given that states/shows that "surface processes ... are crucial to the growth of faceted crystals." Perhaps, this sentence could be re-worded as: "But the KF coefficient ignores surface processes that have been shown to be crucial to the growth of faceted crystals by XXX."

p. 26, line 9: Could an explanatory/expository sentence or two about beta be added after "... or some other orientation." to give the reader some intuition for this parameter that is rather central to the paper? Perhaps, "While beta  $\rightarrow$  1 for non-faceted crystals, beta can be much less than one for faceted crystals. In particular, beta  $\rightarrow$  0 as  $\sigma_S \rightarrow$  0 for faceted crystals." or something of the sort.

p. 26-27, section 2: Some kind of cartoon or sketch, along the lines of figure 2 in Nelson and Baker (1996) might be helpful in illustrating to the reader where surface effects might be important and how they compare to the vapor impedance. One aspect that might be emphasized (and isn't considered in NB96's figure 2) is that small ice particles – even at very cold temperatures – would not be subject to these effects as long as they remained roughly spherical. I am thinking of tropical tropopause layer cirrus as an example.

p. 28, line 7: Even if using the values for alpha from Jouzel (1986), it seems worthwhile to cite the original sources:

M. Majoube (1970). Fractionation factor of  $^{18}\text{O}$  between water vapor and ice. *Nature*, vol. 226, 27 June 1970, p. 1242. <http://dx.doi.org/10.1038/2261242a0>

L. Merlivat and G. Nief (1967). Fractionnement isotopique lors des changement d'état solide-vapeur et liquide-vapeur de l'eau a des temperatures inferieures a 0degC. *Tellus*, vol. XIX, p. 122-127. <http://dx.doi.org/10.1111/j.2153-3490.1967.tb01465.x>

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Note that the Merlivat-Nief paper has slightly different coefficients in the HDO formula. I'm not sure why this would differ from Jouzel (1986), but I suppose the latter reference may be more reliable.

p. 30, line 4: Could some motivation or a reference be provided for the power law approximation for beta? In particular, it seems worth noting explicitly that  $\beta=0$  for the case of no surface supersaturation and so that the vapor flux onto the surface increases rather slowly for small surface supersaturations.

p. 30, lines 13-14: Part of this sentence confused me. Specifically:

"... the former [surface impedance ratio,  $z$ ] ... decreases with increasing crystal size, as shown in figure 1."

When I look at figure 1, I see crystal size increasing to the right, marked along the top of the figure. Each of the curves show  $z$  increasing to the right, which suggest to me that  $z$  increases with crystal size. Am I missing something here? The " $z$ " label on the left axis is partly cut off, but the caption does suggest that this is " $z$ " and not " $Z_S$ " or something else.

p. 30, line 16: Should this be re-worded as: "Deviations from the KF prediction can occur at large  $z$  or when  $x$  differs from the KF limit of  $d$ '."?

p. 32, line 13: Perhaps, the word "Results" could be replaced here by "The resulting analysis" or "This analysis", so that it is more clear where the results are coming from.

p. 35, line 22-24: This is a nice idea and is similar in some respects to the estimation of source region humidity as in, for example:

S. J. Johnsen, W. Dansgaard and J. W. C. White (1989). The origin of Arctic precipitation under present and glacial conditions. *Tellus B*, Vol. 41B, p. 391-486. <http://dx.doi.org/10.1111/j.1600-0889.1989.tb00321.x>

p. 38, appendix C: Why is everything here in terms of  $\sigma_{\infty}$ , while equation 5

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is written in terms of  $\sigma_S$ ? I can understand that there is a probably a good reason for the switch, but it is a bit confusing to the reader. It would also be useful to have a bit more detail in this appendix, specifically:

- a reference for computing the correct value of  $z$  (or, equivalently,  $Z_S$  or  $\beta$ ),
- explicit mention of the assumptions about crystal habit/size, and
- a range of values that  $\sigma_1$  could take on.

Only a sentence or two in each case would probably be enough.

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Interactive comment on Atmos. Chem. Phys. Discuss., 11, 17423, 2011.

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