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Atmospheric
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Discussions

Interactive comment on "Modelling and intepreting the isotopic composition of water vapour in convective updrafts" *by* M. Bolot et al.

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Answer to referee #2

We thank referee #2 for his comments and suggestions and for his very thorough review of our manuscript. We agree with the reviewer's identification of areas where we should better clarify the rationale behind or use of model assumptions. We also agree that the assumption of a specific path of saturation adjustment has complex implications that should be more carefully presented. Our purpose with this simplified model was to explore parameter space and exmaine the isotopic consequences of different scenarios. We have now tried to better articulate when model assumptions are made out of convenience/necessity, with the understanding that reality is captured within a

parameter sweep, rather than directly representing exact physical processes.

We respond to individual referee comments below. (Comments by the referee are in boldface, followed by our answers in regular font.)

1. ***Consider equations A5 and B2 as they apply to the deposition of the standard and heavy isotopes (respectively) onto liquid water. If saturation adjustment is assumed, one is asserting that the coefficient of*** $(1 - S_l)$ ***in A5 is essentially infinite, so that departures of*** S_l ***from one are quickly removed on whatever timescale is considered. As the coefficient in B2 is similar in size, isotopic equilibrium of cloud liquid and vapour (as implied by the term in square brackets in B2 set to zero with*** $(S_l = 1)$ ***should also be assumed for consistency.***
Above 0C, all of this is clear... However, below 0C, mixed-phase saturation adjustment ... introduces issues that must be thought about carefully for their isotopic implications.
[...]
With all of that said, the assumption of mixed-phase saturation adjustment used in the paper is a reasonable starting point for a model of its complexity.

   The referee begins the discussion of a saturation adjustment with an argument that vapour in liquid-only clouds, and possibly in mixed-phase clouds, should be close to saturation over liquid: $S_l \sim 1$. We agree with much of this argument and have added further discussion to the manuscript. (We also agree that our assumptions were a reasonable starting point for our work.) We do not adopt the form of the mathematical representation proposed by the reviewer to explain intermediate values of vapour pressure in mixed-phase clouds, for reasons outlined below, but the arguments laid out in this careful review have helped us greatly clarify our explanations.

The referee's argument is similar to that in appendix B3 of Blossey et al. 2010, which is based on the assumption that the number density of droplets $n(a)$ scales as $1/a_c^3$, where a_c is the characteristic size of droplets (see Fig. 1). The time evolving equations for total mixing ratios then contain a factor $\int an(a)da = O(1/a_c)$ where the a in the integral arises from the geometric factor in our equations A5 and B2 (C in our notation). Because the $1/a_c$ contribution is large, it can only be balanced when the other factors coming from equations A5 and B2 are small. This result would in turn imply that water vapour is close to liquid saturation in much of the mixed-phase regime, whenever these droplet size distribution holds. A qualitative explanation for these results is that the assumed droplet distribution effectively means that droplets are densely packed within the available volume and therefore the relaxation to liquid saturation is fast.

The assumption of liquid saturation further implies that environmental water vapour isotopic ratio equilibrates with the surface of the droplet, and, if the diffusive time scale within droplets is of the same order or faster as relaxation to liquid saturation, with entire liquid droplets. For this reason Blossey et al. 2010 assume no kinetic effects would arise on evaporation of liquid in mixed-phase clouds, and their model does not produce the isotopic enhancements during the WBF process that we suggest are possible in certain conditions (Figures 5, 6, and 8).

We agree that the above argument is compelling and would hold in the mixed-phase regime when the system is dominated by liquid water. At some point during continuing evolution of a mixed-phase cloud, however, the argument must break down, because vapour pressure must transit from close to saturation over liquid to saturation over ice (or saturation over ice times a pre-defined supersaturation factor). Our varying saturation adjustment is intended to capture this transition.

The referee suggests that the saturation adjustment could be thought of as due to patchiness in the system, with some parts of the system close to liquid saturation

and others close to ice saturation, resulting in an intermediate average saturation. The referee suggests that this effect could be captured by replacing the factor $(1 - S_l)$ in A5 and B2 with something like $(f - S_l)$, where f represents the value to which spatially averaged vapour pressure tends in this regime. (Since $f < 1$, vapour would be subsaturated over liquid at droplet surface). Both we and the referee are somewhat bothered by the physical interpretation of this representation, which would suggest that isotopic equilibration no longer holds across the surface of droplets. The suggested representation would imply that the local physics on the surface of the droplets is influenced by some macroscopic constraint, something that cannot hold in reality.

We suggest that a better explanation for intermediate values of saturation in the mixed phase is that the droplet distribution changes. First, as some droplets freeze, the droplet distribution effectively becomes less dense, so that the density scaling $1/a_c^3$ would be replaced by $1/a_c^s$ where $s < 3$ is a fractal dimension which decreases as the ice phase develops within the mixture. Second, as freezing progresses, the mean droplet size a_c may increase, as small droplets are more easily frozen. The result is that the factor in front of the mixing ratio equations that was originally large ($O(1/a_{c0})$ in liquid-only clouds above 0 C becomes increasingly smaller as glaciation proceeds. That lessening means that vapour pressure can depart from liquid saturation without the need of an unphysical $(f - S_l)$ factor.

Although we have not adopted the referee's proposed representation, these very useful comments have led us to amplify our discussion of the saturation adjustment process in mixed-phase clouds, including articulating the differences between our representation and that of Blossey et al. 2010 and discussing conditions that our simple saturation adjustment parameterisation cannot reproduce. We cannot, for example, capture persistent vapour pressure close to saturation over liquid that transits abruptly to saturation over ice during the last stages of glaciation, a case that may be physically plausible in conditions of slow uplift. We intended this work to be an exploration of parameter space, and the saturation adjustment representation necessarily involves some restriction of that space. We have added both a longer discussion of the relevant physics and an explanation of the limitations of our parameterisation. As the reviewer points out, it is a useful first step, but should be followed by a prognostic representation (also the subject of work in progress). As a last comment, we note (and have now noted in the revised manuscript) that if vapour pressure remains close to liquid saturation through much of the mixed-phase lifetime, the resulting reduction in kinetic isotope effects during the WBF process makes isotopic composition a better, rather than worse, tracer for some science. The less sensitive vapour isotopic composition is to the details of glaciation, the better an indicator it is of glaciation altitude.

2. ***461/15 (p. 461/line 15): re-equilibrate on what timescale? In numerical models, one can usually assume equilibrium if the re-equilibration time is smaller than or comparable to a model timestep. What kind of timescale is being considered here? You might mention that appendix B (and in particular figure B1) supports the use of an equilibrium assumption for cloud liquid. Even the largest cloud droplets (~50 um) have a ten second equilibration timescale at -25* C**. ***Equilibrium seems a good assumption in most any models (even high-resolution models with timesteps of only a few seconds) except at temperatures below -20* C*, where the largest cloud droplets would equilibrate more slowly.***

   We realise that the model criteria for removing a droplet from the “actively exchanging” bin and assigning it to the “nonexchanging” bin was not made as clear in the manuscript as it could have been. The model autoconversion parameterisation is a simple ad hoc constant fractional removal of a factor C_l of the cloud liquid. That removal represents both precipitation and growth of droplets to a non-exchanging size. Note that our figures 7c and 7f show that the parameterisation of liquid removal makes a relatively small difference to vapour isotopic

   profile over a wide range of potential values. In the liquid-only regime, profiles are unchanged for different values of C_l as long as enough liquid is retained to dominate the vapour content. The removal parameterisation matters most in the mixed-phase regime, but still has a smaller effect than does the height of glaciation (Figures 7b and 7e).

   We have moved much of the discussion of timescales for equilibration to the appendices and have added additional details and a new figure, to provide detail for those readers wishing to see it without making the text confusing.

3. ***465/12: The text states that irreversibility effects are neglected. Perhaps, it could be useful to add that this means that the source terms of*** θ_{il} ***on the RHS of equation 4 are neglected.***

   We had attempted to convey this with our text of (465/11-12), but apparently did not do as good a job as we had hoped to. We wrote “the tendency in θ_{il} arising from irreversibility has small effect on the temperature profile, as shown by Bryan and Fritsch (2004), and we therefore neglect it.” We have reworded following the referee’s suggestion.

4. ***For the sake of generality, I wish that the equations would be written in terms of*** $d(.)/dt$***, rather than*** $d(.)/dz$***. Constructing the model in terms of time variation seems more generally applicable to me, so that others could use the model more directly. At the end of the model development, the equivalence of time and height variations could be noted and the required assumptions mentioned.***

   We agree with the referee that a model expressed in terms of time variation is more generally applicable and would permit its use within a larger numerical model of the atmosphere. That more complex representation is beyond the scope of a model like ours, however, whose intent is to explore parameter space and determine relationships between microphysical processes and isotopic tracers.

The governing equations of the model presented here describe the vertical evolution of a system that is quasi-balanced at each grid step (with the exception of supersaturation in the glaciated part of the cloud, as will be seen in item 6).

It is true that writing equations in terms of $d(.)/dz$ bypasses an explicit incorporation of an updraft speed. The updraft speed is implicitly included by the choice of parameter values, especially the saturation parameter zeta. Following the reviewer's suggestion, we have in the revised manuscript tried to better describe the relationship between updraft velocity and the parameter values that would follow.

5. ***466/16: The font for ACP makes the lowercase letter "e" nearly indistinguishable from "theta". Would it be possible to use a special form of theta or e, so that one can discriminate between them more easily.***

   This is an unfortunate typographical issue that we also noticed on publication of the ACPD version of the manuscript. Since the ACP format seems to differ from that of ACPD, we will wait to see the final ACP layout (presuming acceptance), but will definitely request a font change if the confusion persists.

6. ***467/equation 6: What is the justification that deposition onto ice should not affect the supersaturation with respect to ice? Would not one expect deposition to draw down the supersaturation to a more modest value? While the GCM parameterizations of supersaturation are rather simplified and unrealistic, they seem to represent that deposition onto ice will occur over a range of supersaturations. The supersaturation assumed by GCM parameterizations is suggestive of a weighted average of the supersaturations over which deposition has occurred.***
   Thinking about the balance between vapor uptake and the generation of supersaturation through adiabatic cooling is one place where the use of height as a vertical coordinate is unsatisfying. The updraft speed is quite

important in thinking about the balance of these processes, and that disappears in the d/dz formulation of the parcel model.

The supersaturation described by our ζ parameter represents the quasi-steady state to which supersaturation relaxes once glaciation is sufficiently advanced and liquid water no longer controls the value of supersaturation. While saturation adjustment over liquid water is rapid, that over ice is slow enough that updrafts in nature regularly show persistent supersaturation over ice (see also Korolev and Mazin, 2003). As the reviewer points out, the speed of the updraft is important in setting this quasi-steady supersaturation level: in situations with no uplift or residual uplift, it is expected that cloud ice draws saturation down to ice saturation, but when the speed of updraft is higher, the quasi-steady value of supersaturation has no reason to decrease upon glaciation. (Adiabatic cooling of the rising parcel provides a source of supersaturation). Varying ζ allows us to span the possible values taken by saturation over ice during ascent of a glaciated cloud.

This parameterisation, which has the virtue of simplicity, does have some limitations. We cannot capture the case that the quasi-steady value of saturation over ice varies with the phase of the cloud life cycle. And, as the reviewer points out here, because the pathway for mixed-phase saturation adjustment and the final value for saturation over ice are are both set by a single parameter ζ in our model, a slow updraft rising with minimal supersaturation is also constrained to reach ice saturation at the onset rather than at the completion of glaciation. We have no particular defence for this other than that the simplification seemed acceptable for the purposes of this exploration. The insight our model provides on mixed-phase processes is not affected by assumptions of subsequent supersaturation conditions, and vice versa. We now state more explicitly that our parameterisation of saturation over ice is ad hoc. We have also added a discussion on equilibration timescales in the appendix and provided a figure that relates the time of phase relaxation over ice to updraft speed.

7. ***A related issue is my puzzlement at a comment on p. 468/line 6-8, "We will however rule out liquid growth below 0 C, as we rule out vapour pressure above liquid saturation in the mixed phase region." My impression is that supersaturation with respect to liquid is ruled out because liquid readily takes up any available vapor in excess of liquid saturation. At least when $\zeta = 1$, the vapor uptake should be mainly on liquid, I would think, rather than onto ice as seems to be the case for the model in the paper.***

   We agree with the referee that as long as the cloud is not glaciated, vapour saturation should range from a few percent below liquid saturation if droplets evaporate to a few percent above liquid saturation, and that in cases of vapour pressure in excess of liquid saturation, both ice and droplets may grow simultaneously. The word choice that we made ("rule out") is ambiguous and may have produced the impression that we meant to imply that nature cannot produce water uptake on liquid at temperatures below 0C. We meant instead that our model was limited to permitting no deposition on droplets. Unfortunately, we do not have a physical basis that allows us to compute the fraction of condensate growth occurring as ice deposition, in conditions of simultaneous growth of droplet and ice, because we do not treat deposition onto the condensed phases prognostically. We could have handled this as an extra free parameter, but decided instead to restrict our sensitivity study to cases where deposition onto liquid does not occur below 0 C. As shown by Korolev and Mazin 2003, this is equivalent to limiting the assumed updraft velocity in the mixed phase zone (below a value that they denote u_z^*). We now acknowledge more explicitly that limitation at the beginning of our sensitivity study.

8. ***468/3: Make distinction between model and reality: "As an air parcel is lifted adiabatically, condensed phases -are assumed- to take up vapor so as to maintain the ..."***

   Suggestion taken in this an other places throughout the text. A persistent theme in the reviewer's comments has been to request a clearer distinction between model assumptions (some ad hoc or simplified from necessity) and physical constraints, and we have attempted throughout to provide this.

9. ***468/23-25: suggested re-wording: "Unlike WBF, freezing does not involve diffusive exchanges of isotopologues with vapour, so that the two processes have very different isotopic signatures."***

   Suggestion taken.

10. ***468-469: Why should WBF and homogeneous/heterogeneous freezing have the same functional dependence on altitude/temperature? If $\zeta > 0$, then both supersaturation with respect to ice and droplet supercooling increase with decreasing T. However, should they have the same form? It seems a somewhat strong assumption, though I do not think that I can offer a better one short of making deposition onto ice prognostic.***

    Yes, there is indeed no reason why WBF and homogeneous/heterogeneous freezing should have the same functional dependence on altitude/temperature. Our representation of WBF and freezing is ad hoc and adopted because merging the two processes into a single conversion rate (φ) with a partition coefficient (b) was a way to reduce the number of free parameters. We have now made the ad hoc nature of this assumption more clear in the text.

11. ***470: Would riming (which might be quite important in convective updrafts) change the way that auto-conversion is specified? Perhaps, at the least, the parameterization could be said to contain tendencies due to both "auto-conversion and accretion".***

    Riming in our model is effectively incorporated in the freezing parameterisation. (See response 13). In both case, ice grows at the expense of cloud liquid water, reducing the exchanging reservoir, and the effects on latent heat are the same. We have now made this point in our revised manuscript.

12. ***472/eqn 22-23: Equation 22 seems not so useful from my point of view. I would suggest that equations 22-23 be replaced with two equations: vapor-liquid equilibrium on 463/24 (deserves an equation number) and an evolution equation for $r_v' + r_l'$, which looks something like:***

    $$\frac{d}{dz}\left(r_v' + r_l'\right) = r_v\frac{dR_v}{dz} + r_l\frac{dR_l}{dz} + R_l\left.\frac{dr_l}{dz}\right|_{\mathsf{frz+acv}} - R_i^{(\mathsf{s})}\left.\frac{dr_i}{dz}\right|_{\mathsf{wbf+ajs}}$$

    I would further encourage that equations 22-26 be written so that the balances between sources and sinks are easily spotted. For example, the source and sink of the droplet freezing should be written the same way (i.e., $R_l\frac{dr_l}{dz}$) in both equations, not with $\frac{dr_l}{dz}$ in one and $\frac{dr_i}{dz}$ in the other. This also would make it easier to eliminate typos. For example, my guess is that the acv term in equation 24 should have R_i, rather than $R_i^{(\mathsf{p})}$.

    We have followed the referee's excellent suggestion and have rewritten equations 22 to 26 so that the balance between production and loss terms is more easily spotted. We have also added an equation number for the equilibration equation on 463/24. After some though we think that combining equations 22 and 23 as the referee suggests might be more confusing for the average reader less familiar with the field. The acv term in equation 24 is correct in having $R_i^{(\mathsf{p})}$: this is the isotopic ratio of the subspectrum of ice that precipitates, and it is not expected that $R_i^{(\mathsf{p})} = R_i$ unless precipitation operates non selectively across particle sizes. The terms having $R_i^{(\mathsf{p})}$ disappear from equation 27 though. Adjusting the notation to make sources and sinks more clear will hopefully make these equations more naturally understandable.

13. ***473/7-15: Since these updrafts seem to be strong enough to loft condensate, is there any mechanism for freezing of raindrops? This would affect temperature and buoyancy if these were allowed to freeze. I imagine that***

many raindrops will rain out and that having all of the passive liquid freeze would likely overestimate latent heating, but this seemed something that could be an issue in convective updrafts. On the subject of freezing in convective updrafts, a paper with a similar spirit but different focus appeared recently in JAS (Kumjian et al. 2012, doi: http://dx.doi.org/10.1175/JAS-D-12-067.1).

Droplet freezing is represented as one route of the conversion from liquid to ice via a parameter $1 - b$ described on p. 473/24-25: "The parameter b is the fraction of liquid to ice conversion occurring via the WBF process (see Sect. 3.2.3) as opposed to via direct freezing." Since we are assuming conservation of total energy and do not actually allow the droplets to rain out, we are necessarily overestimating the contribution of latent heat release from droplet freezing for a given b, as the reviewer points out. Our scenario yields the upper bound on updraft temperature and buoyancy. We have now noted this in the text, and now explicitly reference the work of Kumjian et al. in section 4.2.

14. ***474/6/eqn 30: Why not define Λ as the current Λ minus one? Later in the paper, $\Lambda - 1$ appears everywhere. It might make the notation less cumbersome to define it with the "-1" included. The only argument against this that I can see is if someone else defined it in the past.***

    We define Λ as we did and term it the "generalised fractionation coefficient" because Λ naturally reduces to the equilibrium definitions α_l and α_i in conditions of equilibrium deposition over liquid and ice, respectively. We assume that our readers are so familiar with fractionation factors in the conventional notation that attempting to alter that notation would detract from understanding rather than providing additional insight. In our representation, effective fractionation factors continuously transition from α_l or α_i to Λ in disequilibrium conditions.

15. ***474/16: The "-1" after α_{kl} seems superfluous.***

Suggestion taken.

16. ***474/23: "at length" rather than "in length".***

    Suggestion taken.

17. ***477/17-18: Again, is there really no vapor deposition onto liquid water below 0 C? If so, would not this speed up the WBF process a lot by providing lots of surface area for vapor deposition onto the ice?***

    See our answer to item 7. Simultaneous deposition onto supercooled droplets and cloud ice can occur in nature, but we did not permit it in the sensitivity study (although our equation for the evolution of R_v is sufficiently general to handle this situation). We now make it clearer that this limitation is a choice on our part rather than a constraint of nature. As the reviewer notices, the absence of deposition onto liquid water below 0C means that droplets and ice crystals do not compete for available vapour, which should speed the process of droplet evaporation and redeposition onto ice. However, our representation of transfer from liquid water to ice is ad hoc and the associated rate cannot be made dependent upon saturation conditions short of treating diffusive exchanges in a prognostic way.

18. ***481/16: Likely typo: "no precipitation" and "full condensate retention" sound more similar than different to me. Perhaps, the limiting cases were Rayleigh (immediate condensate removal) and "full condensate retention".***

    Corrected to: "[...] immediate condensate removal and full condensate retention."

19. ***483: Would the isotopic impact of WBF be exaggerated because of the assumption of no autoconversion in the base case?***

    Yes, in our model autoconversion would reduce the reservoir of cloud liquid water available to evaporate in the WBF process. Our computed values for η (and the corresponding assumed magnitude of the WBF process) should be considered as upper bounds. We have now noted this in section 4.3.

20. ***483/15: The WBF is driven by non-equilibrium processes that result in kinetic fractionation effects ? it is not itself driven by kinetic fractionation effects.***

    That is correct. We rephrase to: "[...] since the WBF is driven by non-equilibrium processes that result in stronger kinetic fractionation effects for the system $H_2^{18}O/H_2O$ than for HDO/H_2O."

21. ***487/15-21: Could not some CRYSTAL-FACE data from Webster et al. be used to address the questions raised here?***

    In theory CRYSTAL-FACE could have provided the type of observations we are describing, but the data presented in Webster et al. has particular systematic artefacts that make us cautious about its use for this purpose. The CR-AVE and TC4 missions produced some observations of convective plumes but not the comprehensive sampling of CRYSTAL-FACE. We suspect that future measurement campaigns will provide relevant sets of measurements, especially given the increasing availability and capabilities of commercial instrumentation.

22. ***489/2-7: Units might be useful here.***

    We have added the units that were missing.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 22451, 2012.

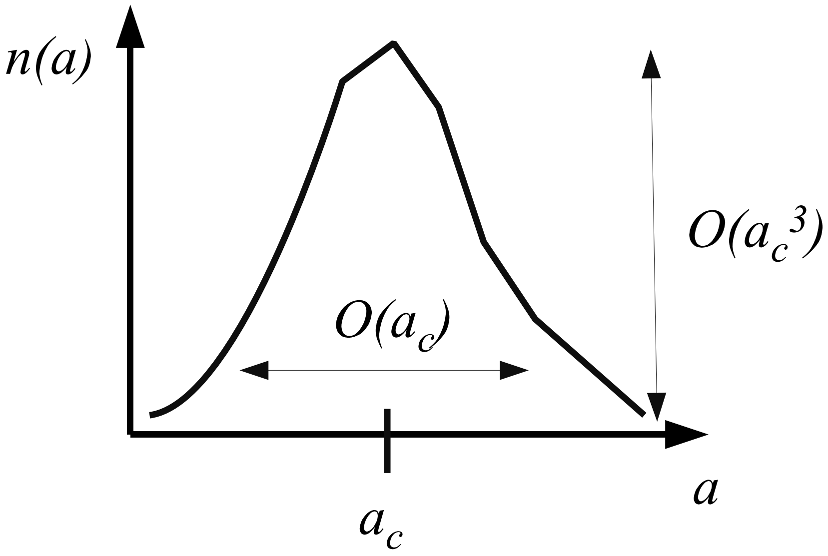

Fig. 1. number density of droplets $n(a)$ (see answer to item 1)