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Atmospheric
Chemistry
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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.

Anonymous Referee #2

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Paper: Modeling and interpreting the isotopic composition of water vapour within convective updrafts// Authors: Bolot, Legras & Moyer// Manuscript #: acp-2012-363

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Summary:

The authors develop an idealized model for the evolution of water isotopic composition within non-entraining, adiabatic convective updrafts. Much attention is devoted to the subtleties of microphysical effects on isotopic composition, such as glaciation temperature, the WBF (Wenger-Bergeron-Findlayson) process and autoconversion rates. It is suggested that information can be inferred about levels of supersaturation with respect to ice and glaciation temperature from observations of isotopic composition at cloud

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base and in the upper troposphere.

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Recommendation:

Publish subject to minor revisions after taking account of the comments offered here.

The manuscript is interesting in developing a model for the evolution of water isotopes in convective updrafts and looking at how one might infer information about microphysical processes from isotopes at the bottom and top of a convective storm. The study is very specialized in that it assumes a non-entraining, adiabatic convective updraft. Additionally, its base case assumes that cloud liquid is not removed by autoconversion and accretion, though this condition is relaxed for the sensitivity study at the end of the paper. Most of my queries below relate to the assumptions embedded in the study. Those assumptions are reasonable for a study of this kind and allow for an exploration of the sensitivity of the model to parameter variations. However, one would expect that the real atmosphere would behave in a slightly different and more complicated fashion. Still, the paper is well worth publishing for its exposition of isotope physics, application of them to the study of convective updrafts and demonstration of how microphysical parameters might be retrieved from observations of isotopic content at the top and bottom of an updraft.

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General comment regarding saturation adjustment, isotopic equilibration of cloud droplets and alpha_kl:

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 vapor (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. In cloud, S_l–>1 unless the updraft is very strong and/or few CCN are available, and vapor and cloud liquid are in isotopic equilibrium (not pseudo-equilibrium). However, below 0C, mixed-phase saturation adjustment, such as that implied by equation 6 and also used in older GCM and mesoscale model bulk microphysical parameterizations, introduces issues that must be thought about carefully for their isotopic implications. If cloud liquid drops persist in subsaturated conditions, as implied by zeta<1 in equation 6, this implies that equation A5 shoud not have (1-S_l) in parenthesis, but rather (fac-S_l) where fac<1 is a function of zeta and the ratio of rho_vˆs for liquid and ice. This seems non-physical as there is no reason that equation A1 should lose validity across T=0C, but is perhaps a justifiable assumption given the complexity involved in representing mixed-phase cloud microphysics faithfully. My justification for mixed-phase saturation adjustment would be to imagine a sizeable grid box filled with patches of liquid or mixed-phase clouds and patches of ice-only cloud, with conditions close to liquid saturation in the patches of liquid or mixed-phase cloud (as suggested by Korolev & Isaac, 2006, JAS 63:2865) and close to ice saturation in the patches of ice cloud. Korolev & Isaac also suggest that mixed-phase clouds are not patchy in the sense described above, at least on smaller scales, as I understand their paper. The work of Korolev et al (2003, doi: 10.1256/qj.01.204) and Field et al (2004, doi: 10.1256/qj.03.102) suggest that such patchiness does exist on larger length scales (on the order of kilometers). The mean saturation vapor pressure in the large, heterogeneous grid box might be intermediate between that of liquid and ice, so that a weighted-average of the two saturation vapor pressures seems appropriate. Such a view seems to be justified by the work of Fu & Hollars (2004, JAS 61:2083). In figure 8 of Korolev & Mazin (2003, which you cite), their parcel model remains close to liquid saturation – within a couple of percent – until all of the liquid is evaporated. Colder temperatures (-30C < T < -40C) might allow a bigger drift from liquid saturation. I would imagine that the drift from isotopic equilibrium and from liquid saturation would be similar at these temperatures, since the coefficients in equations A5 and B2 are quite similar except for A_l, and A_l should probably not be too much smaller than one at these temperatures because of the factor of rho_vˆ(sat,liq).

If one is to treat the isotopic exchanges between cloud liquid and vapor consistently in the setting where zeta<1, one should derive the expression for alpha_kl using the modified version of A5 rather than the standard version. This would, I think, differ from the expression for alpha_kl in the paper. A second option, taken in Blossey et al (2010) and mentioned on 483/29, is to assume for isotopic purposes that equation A5 remains valid across T=0C and that cloud droplets evaporate in conditions close to S_l=1. This implies that cloud liquid is always in isotopic equilibrium with vapor, rather than pseudo-equilibrium. Based on the observations of Korolev & Isaac and the analysis in Korolev & Mazin, assuming exchanges between cloud liquid and vapor occur at equilibrium seems valid to within a couple of percentage points of super- or sub-saturation. In my view and as noted in appendix B, isotopic equilibrium is more likely to be violated in strong updrafts due to deposition in supersaturated conditions with respect to liquid than in subsaturated conditions. A recent paper, Lebo et al (2012, ACP, doi:10.5194/acp-12-9941-2012, especially figs 12-13) finds supersaturated conditions in strong updrafts where vapor uptake by liquid cannot keep up the production of supersaturation by adiabatic cooling. The alpha_kl derived in the paper is entirely appropriate in this situation. My suggestion above is that the mixed-phase saturation adjustment with zeta<1 implies a new form for equation A5 that would in turn imply a different form for equation B1-B2 and hence for alpha_kl.

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. However, I would encourage the authors to think about the above comments regarding alpha_kl in the context of the mixed-phase saturation adjustment scheme used in the paper. The main outgrowth of the point raised above is that isotope physics is simpler in more complex and realistic microphysics schemes that treat vapor uptake onto ice prognostically and

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is more complicated when saturation adjustment with respect to ice is assumed. Note that the Lebo et al study focuses on the effects of assuming saturation adjustment with respect to liquid on their simulations: all of their microphysical schemes treat vapor deposition onto ice prognostically.

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Specific recommendations (all page numbers are prefixed with 22):

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 ($\sim$50 um) have a ten second equilibration timescale at -25C. 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. As noted above, it seems that if one allows cloud droplets to drift from isotopic equilibrium with vapor, one should also relax saturation adjustment with respect to liquid and allow S_l to drift from one inside clouds.

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 theta_il on the RHS of equation 4 are neglected.

465/14: 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.

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.

467/equation 6: What is the justification that deposition onto ice should not affect the supersaturation with respect to ice? Wouldn't 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.

A related issue is my puzzlement at a comment on p. 468/line 6-8, "We will however rule out liquid growth below 0C, 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.

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 ..."

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."

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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 don't think that I can offer a better one short of making deposition onto ice prognostic.

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".

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 rv' + rl', which looks something like:

d/dz(rv' + rl') = rv d(Rv)/dz + rl d(Rl)/dz + Rl*d(rl)/dz|_(frz + acv) - Ri(s)*d(ri)/dz|_(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*d(r_l)/dz) in both equations, not with d(rl)/dz in one and d(ri)/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_iˆ(p).

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).

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474/6/eqn 30: Why not define \Lambda as the current \Lambda 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.

474/16: The "-1" after alpha_kl seems superfluous.

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

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

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

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

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.

487/15-21: Couldn't some CRYSTAL-FACE data from Webster et al be used to address the questions raised here?

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

figure order: Do the figures appear in the order in which they're referenced in the text? If they aren't, please re-order them appropriately.

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