Review of "The role of the global cryosphere in the fate of organic contaminants" by Grannas et al.

This review article is one of a number emanating from the Air-Ice Chemical Interactions effort, which has produced a suite of really good papers. The topic of organic contaminants is important, and this review fits well within the aims of AICI and well within the scope of ACP. It should certainly be published once reviewer concerns are addressed. There is certainly some really interesting stuff in the paper, but there are many areas that could (and need to) be improved.

I have reviewed mainly presentation, as the work itself has already been through the review process in the individual papers referred to.

The sections themselve vary in quality. Some are just what a review needs – a distillation of key results from a wide number of papers, linked together in a logical way so that the thread of what matters is easy to follow and understand, both by experts and non-experts in the field. Other sections are more like a list of results from different papers, that just leaves the reader struggling to figure out what point the review article is trying to make. Overall, the paper looks a bit as if sections of text have been moved around, and in doing so some information now appears in the wrong place, and phenomena are referred to before they are properly explained. Also, there is a tendancy to use jargon, and assume prior knowledge that many atmospheric chemists/physicists will not have. It's not hard to explain terms/concepts so as to make the issues in the paper accessible to a wider audience (which is the purpose of a review paper).

Overall, the manuscript would have benefitted from a more thorough and critical look over before it was submitted, as many of the issues I raise below are pretty fundamental. This list of suggestions is not in order of importance, but generally follow the order in which they appear in the paper:

Introduction

The Introductionwas not well written, trying to link disparate ideas together by making vague statements. It could (and should) be much more interesting! I have a whole bunch of editorial suggestions/comments that I will include here, but to me it would have benefitted enormously from a proper critical read by co-authors. To do the review justice, the Introduction needs some attention.

P16925 line 13: pose "a" severe risk; line 16 (and throughout): "polar" needs lower case "p" – it's an adjective; line 24: Domine needs an é, "Dominé"

P16926 line 9: if you're trying to present a comprehensive list, then also include ice shelves; line 11: please spell out what this involvement is and how it works; line 12: yes, snow has a high albedo, but does ice... surely this varies depending on the number of air bubbles e.g. can't lake ice be transparent..??; line 13 (and throughout): surface of what..? Given that this paper is from the Air/Ice Chemical Interactions community, it is critical (and obvious that you need) to be clear what surface you are talking about – the surface of the Earth, the surface of a snow grain... or what...; line 13: I don't understand what you mean by "decoupling the surface-air interface", please explain better; line 18: please give a reference to support your statement that ice sheets hold nearly 80% of the world's freshwater; line 22: we sort of know that the cryosphere is in the high altitude, boreal and mid-latitude regions... if you want to make this point, do it earlier in the paragraph (maybe where

you list various component of the cryosphere on line 8, e.g. It thus includes... which can be found in polar, mountain, boreal, mid-latitude and urban etc...);

P16927 line 2: what is a "fate process"? This is just poor English; line 4: what is variable about the cryosphere..? I guess you mean coverage of the Earth, but whatever you mean, please spell it out; lines 6-8: "complex cold ecosystems" are not the same as "cryosphere-dominated environemnts" so there is no evidence that cryosphere-dominated environments would be influenced by all three feedback mechanisms – at least the link is not convincing to me from what you've written.

Section 3.1 P16931 line 25 to P16932 line 13 – this section is vague and reads just like a list of published results with insufficient detail and no clear linkages. Would be helped by some "for example..."s and some explanations of "why" the authors found what they did.

P16933 line 6: "increased snowfall on the east side of the Rockies..." as opposed to west (but doesn't it rain/snow more on the west?)? Or increase relative to what..?? Please clarify.

Section 3.2 P16934 line 12: "firn" core rather than "snow"? line22: what post-depositional processes might occur..?

Section 3.3 P16938 line 1 - please explain Darcy's Law, even in brief, for those not familiar with it;

Section 3.4 P16938 Lines 22 to 27 sit oddly in a section about seasonal snowmelt. I'd move them to section 3.2, about ice cores. I would then include an introductory section about seasonal snowmelt instead, describing features (early vs late), explaining what is "freshet" etc.

Section 3.4.1 P16940 lines 10-12: could you explain why this is happening? i.e. why enriched in early or late melt water fractions? Line 17: now I'm confused – just above you're written that enrichment can be in early or late fractions, but here you write that highest loads are in early melt... Really, this section needs to link to what is written in section 3.4.3, which explains all this in detail. To me, the information in section 3.4.3 should appear ahead of what's written here.

P16942 line 4 to end of section 3.4.2: I don't see why this text is included in Section 3.4.2 when it is really about amplification and then dilution. These two paragraphs would sit much better within section 3.4.1 which is specifically about amplification.

Section 3.4.2: link this section to section 3.5 as both are about the cryosphere as storage and release, the critical difference being timescalse. To me it would make sense to move section 3.4.2 to just ahead of section 3.5, or at least refer them to each other.

P16942 line 6 onwards: this information from Bergknut et al was already presented in part on P16932 (although the explanation here is better!). Find some way of merging these sections, or at least link/refer to each other.

P16942 line $15 - \gamma$ -HCH is not defined.... Which brings me to a bigger point. The review would benefit from having a section about the contaminants themselves... i) spell out the abbreviations, ii) explain what they are used for, iii) where are they used, globally or in specific areas, iv) are they likely to have a global or regional influence (would depend on transport as well), v) are they phased out or still in use? Vi) what is their level of toxicity... This could be put into a table, or just a short

section, but there is so much prior knowledge assumed that many readers will not have, so ought to be addressed somehow.

P16944 line 3: particle-bound

Section 3.5, P16946 line 25: what is a "pro-glacial" lake..?

P16947 line 22: lower case "p" for "polar"

Section 3.6: there is a noticeable jump in the level of technical and highly specific language used at the start of this section. If you want to take the reader with you, explain things... e.g. "enantiomer", "chiral", "racemic"... yes, many readers will understand this, or could look it up, but they shouldn't need to, and it's not hard to explain. Plus, EF, E1 and E2 are not defined by this stage, this happens later (P16952 line 24) – the explanation needs to appear before the terms are used. Also, aren't you missing brackets in your equation, which surely should read: EF=E1/(E1+E2)..?

Figure 4 needs a reference of some sort

P16951 line 4: another example of things appearing in a strange order. Statements appearing here about brine enrichment before any explanation of why this should happen. All the explanation (that makes all this clear) appears on P16953 and P16954.... Need to re-order or link the sections.

Figure 5 – can't read it... needs better quality

P16953 line 7: define K_{OC} and K_{OW} properly; line 9: again, discussion about enrichment in ice brine, with no explanation as to why. I'd suggest explaining brine enrichment issues much earlier in this section.

Figure 7 I'd suggest presenting much earlier in this section. In fact, it would help the reader to have an overall description of the physical system, highlighting the relevant processes, near the start of section 3.6.

P16953 line 4: beneath-ice

Section 3.8 – reads more like the influence human populations have on the polar regions, rather than the effect of thawing permafrost... Is there any thawing permafrost in Antarctica..?? Again, information that just sits oddly.

Section 3.9 – if you are writing about photodegradation of "contaminants" then why are the Jacobi references here..?? The only organic molecule these papers deal with, as far as I'm aware, is HCHO... And the Jacobi and Hilker paper has no mention of any organic molecule at all.

Section 4 – a fairly comprehensive section, the one thing that is missing is some comment on links/interaction with policymakers and industry... I think a short piece about this would be interesting and relevant.

Other minor comments/typos:

P16927 line 25: once-pristine;

P16928 line 2: convention doesn't need a capital C on this line;

P16929 line 15: Is ATME the right reference here..? You're not thinking of Antarctic Climate Change and the Environment (ACCE)..? line 22: above-mentioned; lines 21-23: long-term monitoring does not measure long-term records, it creates them... do you mean instead long-term monitoring to measure long-term changes..??

P1630 line 3: change "make up" to "determine"

Figure 1: define LRAT in the caption