The role of the global cryosphere in the fate of organic contaminants
Grannas et al.
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Response to Reviewers

We appreciate the timely and detailed reviews of our manuscript submitted to Atmospheric Chemistry and Physics Discussions. The reviewers provided a number of helpful suggestions that have allowed us to improve the content and quality of this manuscript. We have made significant changes and hope that this revised version is now suitable for publication. Please find below detailed responses to reviewer comments, including line numbers in the current manuscript where we have made specific additions or changes. The comments of the reviewers are included below in black. Our responses follow each comment/set of comments in blue.

Reviewer #2:

The Introduction was 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.

We have significantly revised the introduction with an eye to better descriptions so that material is clear and more interesting to the reader.

P16925 line 13: pose “a” severe risk

This change has been made.

line 16 (and throughout): “polar” needs lower case “p” – it’s an adjective

This change has been made.

line 24: Domine needs an é, “Dominé”

We communicated directly with Professor Dominé regarding this. He has chosen in publications from recent years to spell his surname without the accent, so it is actually correct to write his name as Domine, and he has asked us to change this accordingly.

P16926 line 9: if you’re trying to present a comprehensive list, then also include ice shelves

This addition has been made.

line 11: please spell out what this involvement is and how it works

We have expanded this description of the role of the cryosphere in Earth’s energy balance (lines 90-104).

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..??

This is certainly true, but on the global scale, the role of snow and ice (e.g. polar ice caps) is as a far greater contributor to reflectivity.

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...

Throughout the paper, in instances where surfaces are discussed, we have made efforts to clarify what exact surface is being referred to.

line 13: I don't understand what you mean by "decoupling the surface-air interface", please explain better

We have revised the introduction extensively, in order to better explain cryospheric processes, and the role of snow/ice related to energy balance is explained in greater detail now in lines 90-104.

line 18: please give a reference to support your statement that ice sheets hold nearly 80% of the world's freshwater

A reference has been added to IPCC, 2007.

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...)

We now discuss this earlier, in the second paragraph of the introduction.

P16927 line 2: what is a "fate process"? This is just poor English

This has been reworded (line 131) to read: "Not only is pollutant fate controlled by a wide range of physical and chemical properties of contaminants that are strongly temperature dependent, but the nature and extent of the cryosphere itself may be highly variable under different climate change scenarios."

line 4: what is variable about the cryosphere..? I guess you mean coverage of the Earth, but whatever you mean, please spell it out;

Here we are referring to the nature and extent of the cryosphere, which is discussed in the sentence just before this statement (e.g. Arctic sea ice decline, glacier melting, lines 125-131).

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.

For clarity, we have removed the statement regarding the three feedback mechanisms and now refer only to contaminant cycles, as integrated parts of a complex environmental system of cold regions, being prone to change as a result of a changing climate (which we then indicate will be discussed in more detail in the following sections) (lines 134-139).

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 3.1 has been significantly revised (as also discussed above in response to reviewer #1 comments). We have better organized the section to link the topics clearly and to provide specific examples and better explanations of observations.

Section 3.2 P16934 line 12: "firn" core rather than "snow"?

This change has been made.

line22: what post-depositional processes might occur..?

We have added a few examples of what we mean by post-depositional processes at line 966.

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

For clarity we have removed this reference to Darcy's Law, as it did not add crucial information to the discussion.

Section 3.4 P16938 Lines 22 to 27 sit oddly in a section about seaonal 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.

As suggested we have moved the section that referenced ice core formation into the ice core section (now Section 3.6). We have also added a paragraph about seasonal snowmelt characteristics (lines 467-475).

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.

The particular sentence at lines 10-12 is now deleted from the revised section. Our revised section 3.3 was written to better explain what processes are happening as the snowmelt

progresses. At the previous line 17, the statement that "the melt of a seasonal snow cover causes highest contaminant loads to occur in early spring, when organisms are at a particularly vulnerable stage of development" meant that the contaminant loads were high in the early spring *season* (not the early *snowmelt* stages). This is problematic for aquatic species as they are in a crucial developmental stage in many cases in the early spring season (when the snowmelt process is occurring as a whole). We believe the revised section 3.3 now better explains these processes.

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.

Again, because we have revised this section, we believe this is now more clearly described and better organized.

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.

We have made this suggested change, and a revised section on snowmelt delivery of contaminants (section 3.3) now occurs just before glacier melt delivery of contaminants (section 3.4).

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.

Again, because we have revised this section, we believe this is now more clearly described and better organized.

P16942 line 15 – γ-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.

We have followed this advice (and the advice of reviewer #1) and included an introductory list of abbreviations. We have included in Section 2 a table of the Stockholm Convention POPs (and those under consideration for inclusion), their main usage, and references to key physicochemical parameters (Table 1). Toxicity is difficult to quantify in a way that could be presented concisely in a table. We discuss this in lines 173-179 of the revision and refer to a toxicology review there.

P16944 line 3: particle-bound

This change has been made.

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

This refers merely to a lake fed by glacial waters (and thus influenced by glacier melt). We have defined this in the caption of Figure 3.

P16947 line 22: lower case “p” for “polar”

This change has been made.

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)..?

We have significantly revised this section (now section 3.5), taking into account the suggestions of the reviewer. We have also corrected the equation for EF to include the brackets.

Figure 4 needs a reference of some sort

We have included a reference in the figure caption.

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.

We have significantly revised this section (now section 3.5), and taking the reviewer suggestions feel this is now more well organized.

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

We have improved figure 5.

P16953 line 7: define KOC and KOW properly

Given the revision to this section, we no longer refer directly to Koc and Kow.

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.

We have significantly revised this section (now section 3.5), taking into account the suggestions of the reviewer. Brine enrichment is described earlier in the section (beginning line 756), and accompanied by a figure to help describe the process (Figure 4).

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.

As suggested, an altered Figure 7 (now Figure 4) is presented much earlier in the section (line 710).

P16953 line 4: beneath-ice

This change has been made.

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.

We felt it was important to consider the role of thawing permafrost in the potential mobilization of local pollution. This certainly is attributable to human influences, but the climate change implications nonetheless indicate that changes in contaminant distribution or enhanced mobilization may occur at these localized dumpsites and sewage lagoons.

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.

As suggested, the Jacobi references have been removed (except for the reference to photosensitizing activity of nitrate, peroxide, etc; in this context Jacobi et al. 2006 is still relevant).

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.

As suggested, this addition has been made (lines 1407-1412).

Other minor comments/typos:

P16927 line 25: once-pristine

This change has been made.

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

This change has been made.

P16929 line 15: Is ATME the right reference here..? You're not thinking of Antarctic Climate Change and the Environment (ACCE)..?

This change has been made.

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..??

This change has been made.

P1630 line 3: change “make up” to “determine”

This change has been made.

Figure 1: define LRAT in the caption

This change has been made.