

Editor Dr. Galmarini, Atmospheric Chemistry and Physics

Regarding review of manuscript acp-2015-12

Dear Dr. Galmarini,

Please find enclosed the revised manuscript acp-2015-12. I apologise for the delay in submission of the final revised version of the manuscript, but I have been ill for three weeks with a severe pneumonia and just returned to work this week.

We thank the two anonymous referees for their comments to the manuscript. Following the comments and suggestions we have made two major changes to the manuscript. We have followed the suggestions of referee #1 and included an additional model scenario in the manuscript. This has resulted in added discussion of the new results as well as changes in all figures in the manuscript and in the supplement. In addition both referees suggested changes in the results and discussion sections, and as a result we have re-organized these sections completely so they appear more reader friendly. For these reasons we have not prepared a marked-up manuscript version showing the changes made. We have however included below the response to each of the individual comments from the two referees that were also uploaded to the manuscript webside on April 30.

We hope that you are satisfied with the changes made to the manuscript following the suggestions of the referees.

Kind regards

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Hansen et al.: "Modelling impact of climate change on atmospheric transport and fate of persistent organic pollutants in the Arctic"

Reply to anonymous referee #1

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Referee:

The paper describes a model to predict effects of climate changes on the long range transport of POPs. The model is not new. The paper apply an existing model to specific scenarios suitable to describe transport and fate patterns to the Arctic in present and future climatic conditions.

The topic is of relevant interest and the modelling approach is sound. Therefore, the paper may be suitable for publication.

However, it is not always clear and reader friendly. Some information is incomplete or not reported. Some major and minor weaknesses need to be revised before publication.

Referee:

The model is applied to a present (1990-2000) and a future (2090-2100) temporal interval. In both cases the same emission hypothesis is made, with (W) and without (Wo) initial concentration. These scenarios should allow assessing distribution and fate of new emitted chemicals (the Wo scenario) and a combination of new and already present chemicals (the W scenario). Why not considering a scenario with initial concentration and no emissions? It would allow assessing disappearance patterns of POPs in a realistic condition, considering control and phase-out measures in act, according to international agreements (e.g. Stockholm Convention).

<u>Answer</u>: We thank the referee for the suggestion to make another model scenario with only initial environmental concentrations and no emissions. We have done this and included the results in the manuscript. As a result we have renamed the model scenarios to 'E' – Emissions only, 'S' – spin-up concentrations only, and 'ES' – emissions and spin-up concentrations.

<u>Referee</u>: The scenarios are insufficiently described. A detail of the climatic conditions in the two temporal intervals should be reported, at least in the supplementary material. The initial concentrations of the chemicals in the different environmental compartments should also be reported. Table S2 shows total emissions of the chemicals. They are emissions for the whole 10 years period? Emission is constant and continuous? In which compartment emissions occur?

<u>Answer</u>: We have described the major differences between the two simulated periods for the SRES A1B climate scenario in the introduction section on page 5, line 19-26. The initial concentrations applied in the 'ES' and the 'S' scenarios have been added to table S2 as well as more detailed information on the temporal resolution of the emissions and to what compartment they enter.

<u>Referee</u>: Section 3 is a detailed description of the results. This section is too long, in some parts unnecessary (describing some quite obvious issues), without focusing on some relevant issues that are not immediately understandable from the figures. For



example: (Figures S1, S6, S10) why some chemicals show strong seasonal variability and other are almost stable? Why chemicals with comparable properties (e.g. α -, β - and γ -HCH) show a completely different behavior?

<u>Answer</u>: We have rearranged the results and discussion section. We have removed the unnecessary parts of the results section and as a result the section has become shorter and more focused.

As for the comment on the seasonality this is a question of the scale of the plots, where seasonal signals can be 'hidden' for the compounds with high mass. We have strengthened the discussion of the difference of the HCHs following the Referee's suggestion.

<u>Referee</u>: A more schematic, synthetic and less dispersive description of what happens (highlighting differences between: present and future; total domain and arctic; light and heavy chemicals) would be more reader friendly.

<u>Answer</u>: We thank the referee for the comment. We have produced a table displaying either positive or negative differences in mass in total, in air, water and soil, in the entire model domain and in the Arctic for the three model scenarios. The new table presents the major results in a schematic way that is more reader friendly. We have furthermore rearranged the results and the discussion sections following comments from Referee #2 with a more strict discussion of the individual compounds.

<u>Referee</u>: Moreover, many statements in the description does not correspond to the figures. For example:

Page 6, lines 11-12: "The total mass of α -HCH decreases over the modelled decades for the 'W' simulations with a more rapid decline for the 2090s than for the 1990s" I can not see any substantial difference. The two trends are practically identical. Page 6, lines 13-14: "The mass of β -HCH and the intermediately chlorinated PCBs (PCB101 –PCB118) display the same pattern". Not true. β -HCH shows a decrease of about 40% in the W scenario and negligible increase in the Wo. PCB 101-118 show much smaller decrease in W and substantial increase in Wo.

Many other inconsistencies are present in the discussion.

<u>Answer</u>: We have thoroughly revised the result and discussion section and the inconsistencies mentioned (as well as others) have been removed.

<u>Referee</u>: In conclusion, it is my opinion that the paper should be rewritten, checking for inconsistencies in the description, eliminating un-necessary parts, synthesizing the most relevant outcomes and providing clear interpretations of the most relevant differences.

<u>Answer</u>: Thanks to the Referees suggestions we have revised the manuscript thoroughly. Apart from adding the information the Referee has requested on the climate scenario, on the initial concentrations and the emission as well as data for the additional model scenario, we have revised the results and discussion section thoroughly as mentioned above, and the resulting manuscript is now much more reader friendly.

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Reply to anonymous referee #2 Anonymous Referee #2 Received and published: 31 March 2015 General comments

The authors present the research on the impact of climate change on atmospheric transport and fate of 13 persistent organic pollutants in the Arctic by a comprehensive DEHM model with relatively higher resolution and complex chemical and physical processes compared to the peers.

The research results would enrich the current understandings of POPS distributions in the Northern Hemisphere and in Arctic and their future changes under future climate change scenarios.

However, the paper is not well organized and hard to follow the results. Suggest to reorganize the paper with following points:

Major comments

- As to "the results"

<u>Referee</u>: 1) The paper presents a rich set of modeling data and their presentation style is a little confusing. There are more than three categories for discussing the results: (1) regions from northern Hemisphere to the Arctic, (2) components from HCHS to PCBs, (3) compartments from air, soil, water to vegetable for the variations of total mass, difference, relatively difference and monthly means of POPS in two climate periods. The authors should select ONE category as the main pillar and then describe results of other categories within the main category.

<u>Answer</u>: We thank the referee for the comments on the organization of the manuscript. As a result the result and discussion sections have been re-arranged so the main category now is the individual compounds under where all other subjects are discussed. This has improved the readability of the manuscript significantly.

<u>Referee</u>: 2) A lot of results or discussions came or were based from the supplements materials. Suggest that the authors select major plots from the Figure sX to the manuscript figures to make the paper more readable.

<u>Answer</u>: The referee requests an improvement of the readability of the manuscript. We have carefully considered the suggestion of the Referee to move plots from the supplement to the main manuscript, and we have decided not to follow the suggestion but to improve the readability in other ways. As a result of comments from Referee #1 the references for the supplement figures have been reduced to a minimum and only in cases where the plots are essential for the discussion. In this way the manuscript has been made more reader friendly as the referee requests.

<u>Referee</u>: 3) The authors should give more quantitative conclusions or discussions instead of those subjective words of 'more rapid decline'

<u>Answer</u>: We have strengthened the discussion and conclusions and as a result the subjective words have been removed and we only discuss results that are statistically significant.



<u>Referee</u>: 4) The key word of this paper is on the transport. The impact of climate changes on the transport pathways of the POPs should be discussed. Scientifically, we would like to know how the climate changes will influence the pathways such as the "grasshopper" and "cold condensation" effects

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<u>Answer</u>: While we agree with the Referee that it is scientifically interesting to know how climate change will influence the 'grasshopper' transport and the 'cold condensation' process this is not possible to quantify with the model set-up applied in this study. The model applied in the study is a very complex model and it is not possible to separate the effect of one single process with the applied model set-up. It is out of the scope of the study but could be the subject of a future study.

- As to "Test of statistics significance"

<u>Referee</u>: The authors should add this content to the results and discussion to support the discussion instead of simply description alone just like it does in 4.3 for r-HCH. <u>Answer</u>: We have combined the 'Results' and the 'Discussion' sections and moved the section about test of statistical significance to the beginning of the new 'Result and Discussion' section. We furthermore only presents and discusses the results that are statistical significant.

- As to the" comparison with previous results"

<u>Referee</u>: The authors should focus more on the result same to or different from previous works and the reasons why. No need to describe the previous work one by one. <u>Answer</u>: We have shortened this section according to the suggestion of the referee.

Technical corrections

Referee: The first (Fig. S9) should be Fig. S7 in 3.2 The Arctic

Answer: We have corrected this error.