

Response to Reviewer 2:

Rev 2

### **General Comments**

**This manuscript reviewed literatures for the atmospheric concentrations of several classes POPs in the Antarctica from 1988 to 2021. Temporal trends were evaluated for DDT, DDD PCBs, HCHs and HCB according to the effective ban of SC. Atmospheric half-life times of these POPs were estimated using characteristic decreasing times (TD).**

**The results showed that the ban of SC significantly influenced the levels of HCHs and PCBs, while HCB showed increasing concentrations in some publications, and longer half-life time than other POPs.**

**The impact of climate change on the POPs levels was discussed. Increasing temperature can cause remission of POPs from the surface, and other biogeochemical processes. Overall, the manuscript have been well documented, and addressed to the emerging concern for POPs in the Antarctic. I would suggest it can be accepted with some revision.**

We agree with the reviewer in their comments provided below and we corrected the manuscript following their valuable comments that we think improved the text in a significant way.

### **Specific comments**

**L116-118, including only articles written in English; excluding from the analysis references that do not refer to a good quality assurance and quality control during the chemical analysis, or if the levels of field blanks were not reported.**

**Please give more detail description for “good quality assurance and quality control during the chemical analysis” applied for literature selection**

We agree with this comment, English language is not a decisive issue to check the quality criteria for selection of the studies thus we expanded our criteria in the text giving more details.

Now the text says:

“An exhaustive search was performed in the Web of Science and Scopus databases using the words "Persistent Organic Pollutants", "atmospheric" and "Antarctica", including only articles written in English; excluding from the analysis references that do not refer to a good quality assurance and quality control measures. Thus, studies not reporting information about blank samples, limits of detection, limits of quantification, and/or instrumental detection limits, and referring to previous works reporting the quality criteria used were not included in the present study. This is important because the reported concentrations are at very low levels and to avoid bias it is necessary to be sure about the quality measures of sample collection and analysis”

### **L121-122, data obtained from active and passive sampling**

**As data from both active and passive sampling were collected in this work, although the authors stated no clear variation between these two data sets, I guess it is worth to compare the data between active sampling and passive sampling in this review, and give a suggestion for future monitoring program.**

We agree with this comment, so we decided to include a GLM in the analysis considering the terms time (year) and method (Active vs passive). The results of the analysis indicated that the atmospheric POPs concentration obtained, and the variation was only related to time and no influence of the sampling methodology was identified. indicate that the variability of atmospheric POP concentrations is mainly due to the years and not to the type of sampling used (Table S.5.). On the other hand we could suggest that the sampling strategy should be selected according to

the objectives of the study design. For example, if our focus in a study is a high-resolution monitoring and no logistic restriction is found (i.e. electricity) then the sampling strategy for monitoring should be active but this will limit the spatial cover of the study. On the other hand, if a highly geographical coverage is needed, less time resolution and logistics are complicated (no electricity available) then passive sampling will be the correct option. However, we think that this is far from the objectives of the present study. But we included a comment on this aspect in the source of bias section.

New text could be found in

Methodology section, page 5, line 130-133.

“Finally, a Generalized Linear Model (GLM) was performed to elucidate whether the variability in the atmospheric POP levels reviewed is due to the different types of sampling used by the different studies (active or passive sampling) or the time variable.”

OCP results, page 7, lines 200-202.

“2,4'-DDD isomers, because all levels reported in East Antarctica were below the detection limit. In addition, the results of the Generalized Linear Model indicate that the variability of atmospheric OCPs is mainly due to the year variable, with no significant differences ( $p > 0.05$ ) between the atmospheric levels obtained from active and passive sampling (see Table S.5).”

PCB results, pages 9 and 10, lines 263-266.

“. On the other hand, it is essential to highlight that the variability of PCBs reported in this study is substantially due to the time variable ( $p < 0.05$ ), with no significant differences ( $p > 0.05$ ) between the atmospheric levels of PCBs obtained from active and passive sampling (see Table S.5).”

And Potential source of bias section, page 10-11, lines 322-326

“Therefore, recent data are generated by more sophisticated techniques and modern laboratory QA/QC criteria. On the other hand, we also included studies using active and passive sampling, but no major differences in the values obtained were observed (See Tables 1, 2, and 3) for the whole compounds which agrees with intercalibration experiments conducted in other areas comparing passive and active sampling together (Prats et al., 2022).”

Also in the section conclusions we commented out thoughts regarding the use of active and passive sampling for future monitoring networks.

Now the text says:

“There is increasing evidence of the presence of emerging compounds in different environmental matrices in Antarctica, however, the current surveillance of atmospheric pollutants is related to specific research groups, instead of coordinated efforts between countries with Antarctic presence, where continuous monitoring networks could be generated with the inclusion of various persistent toxic chemicals, as analogous to the efforts done by the Arctic Monitoring and Assessment Program (AMAP), or the Integrated Atmospheric Deposition Network (IADN) in the Great Lakes. In this sense to establish a monitoring program for assessment of POPs levels in Antarctic Atmosphere will depend on the capabilities and the facilities, since active sampling strategy will benefit a higher resolution in the assessment of POPs trends in the monitoring points but on the other hand the use of passive sampling strategy could represent a high spatial coverage to monitor trends but lower time resolution. But there is need to establish in the future a bigger monitoring network coordinated following the previous experience gained from AMAP.”

**L189-197, spatial distribution of HCB, a-HCH, b-HCH, and g-HCH isomers was discussed in this section. Please give more discussion for the significant spatial differences of HCB and a-HCHs between East and West Antarctica.**

We discussed this in the text however is difficult to discuss this topic due to the lack of information we included a paragraph trying to summarize the information. However we only could argue that the proximity of South America to Western Antarctica and the data on historical usage and emissions from continents in the bibliography could suggest an explanation to these differences:

Now the text says:

“The detected concentrations of HCB,  $\alpha$ -HCH, and 4,4'-DDT indicate significant spatial differences ( $P < 0.05$ ), with higher atmospheric concentrations in West Antarctica than in East Antarctica (Table S.4). The  $\alpha$ -HCH, and 2,4'-DDT isomers did not show spatial differences between the two zones ( $P > 0.05$ ) (Table S.4), but the usage of these compounds decreased in a similar way from 1990 to 2000 (Vijgen, 2006). This can be explained by two causes together, the first is the greater proximity of South America to the Antarctic Peninsula. The proximity itself has to do with the possibility of transport of these compounds from southern South America where it is suggested that air samples influenced by the continent are capable of transporting pollutants from South America to Antarctica (Dickhutt et al., 2005) such as Heptachlor epoxide. However, when looking to usage reported in South America compared to Africa (Li, 1999). Thus, this could influence the abundance of  $\alpha$ -HCH in the western Antarctic area. Examining previous information for both HCB and 4,4'-DDT there is not a great deal of information about the use of these compounds in areas near Antarctica but the proximity to South America could explain these variations in conjunction with the paucity of data in Eastern Antarctica. On the other hand, the U-Mann Whitney variance analysis was not performed for the  $\alpha$ -HCH and 2,4'-DDD isomers, because all levels reported in East Antarctica were below the detection limit. In addition, the results of the Generalized Linear Model indicate that the variability of atmospheric OCPs is mainly due to the year variable, with no significant differences ( $p > 0.05$ ) between the atmospheric levels obtained from active and passive sampling (see Table S.5).”