Response to Reviewer

Dear Reviewer:

Thanks for your comments concerning our manuscript entitled "**Temporal and spatial variations of atmospheric unintentional PCBs emissions in Chinese mainland from 1960 to 2019**" (Ms. Ref. No.: EGUSPHERE-2022-977). Those comments are very valuable and helpful in improving our paper. We have studied the comments carefully and made corrections correspondingly. The main corrections in the paper and our point-by-point responses to reviewer's comments are presented below.

Responses to Reviewer #1

General comments: This manuscript presents an estimation on emissions of 12 dioxin like UP-PCBs from China. Historical provincial emissions from 66 sources in China were estimated. Furthermore, a 0.1º × 0.1º gridded emission inventory for 2019 was developed. This study provides key information about temporal and spatial characteristics of UP-PCBs emissions in China. The inventory could be used as key input in modelling the environment behaviours of UP-PCBs and support the assessment of health and ecology risks due to UP-PCBs pollution. The paper is overall well organized and properly documented. It can be published after minor revisions.

Response: We really appreciate your valuable comments and suggestions. We have made corrections correspondingly according to your comments. Please check point to point response as below.

Comment 1. PCBs have many congeners, but only 12 congeners were considered in this study, a brief explanation of choosing these 12 congeners might add in the introduction.

Response: Thank you for the comment. Our study mainly focused on unintentionally produced dioxin like PCBs. In 2005, WHO evaluated the toxic equivalence factors of dioxins and dioxin-like compounds and 12 PCB congeners were identified with potential significant adverse impacts on human health and environment. Also, most of studies only focused on these 12 PCB congeners. As a result, emission factors for PCBs other than these 12 PCB congeners were scarce, making it hard to estimate their emissions. Thus, 12 dioxin-like PCBs were included in this study. The description of 12 PCBs congeners was added in Line 37 in preprint.

"other sources of these compounds (UNEP, 2001). In 2005, WHO evaluated the toxic equivalence factors of dioxins and dioxin-like compounds and 12 PCB congeners were identified with potential significant adverse impacts on human health and environment. Globally, PCBs were mainly used commercially".

Comment 2. The first sentence in the abstract is about PCBs, but the following contents are about dioxin like UP-PCBs. This sentence should be reorganized.

Response: Thanks for the comment. The sentence was revised as "Polychlorinated biphenyls (PCBs) are mainly unintentionally produced with the ban of their use and manufacture, and they, especially those dioxin-like compounds have been proven to be harmful to ecosystem and human health."

Comment 3. In section "2.3 Emission estimation and spatial allocation", spatial allocation of fuels in PKU-FUEL might be briefly described here

Response: Thank you for the comment. This section was revised as: "A bottom-up procedure was applied to compile the inventory. Provincial emissions of each PCB congener for each source were calculated as products of the source strength and corresponding EFs. Provincial emissions of 2019 were further allocated into $0.1° \times 0.1°$ grids using various surrogates. For fuel combustion sources (including agricultural waste burning and wildfire), gridded fuel consumption data from PKU-FUEL (Wang et al., 2013) were used to disaggregate emissions. In PKU-FUEL, county level fuel consumption data were acquired and were further disaggregated into $0.1°\times0.1°$ grids using GDP, rural population, urban population or total population depending on fuel categories. For fuel consumed in power plants and production of lime, coke, as well as aluminum, the geolocations of the sources were obtained to allocate emissions. For agricultural waste burning and wildfire, emissions were firstly allocated into $0.25°\times0.25°$ grids based on dry matter burned from GFED4 (van der Werf et al., 2017) and were further disaggregated into g $0.1° \times 0.1°$ using vegetation density (Friedl et al., 2002). For cremation of corpse, which was a PCBs source included in this study, gridded population density (Oak Ridge National Laboratory, 2020) was applied to allocate emissions. For other sources that were not included in PKUE-FUEL, gridded industrial coal consumption from PKU-FUEL was used as surrogate."

Van der Werf, G., Randerson, J., Giglio, L., van Leeuwen, T., Chen, Y., Rogers, B., Mu, M., van Marle, M., Morton, D., Collatze, G., Yokelson, R.: Global fire emissions estimates during 1997–2016, Earth Syst. Sci. Data, 9, 697-720, https://doi.org/10.5194/essd-9-697-2017, 2017.

Friedl, M.A., McIver, D.K., Hodges, J.C.F., Zhang, X.Y., Muchoney, D., Strahler, A.H., Woodcock, C.E., Gopal, S., Schneider, A., Cooper, A., Baccini, A., Gao, F., Schaaf, C.: Global land cover mapping from MODIS: algorithms and early results, Remote Sens. Environ., 83, 287-302, https://doi.org/10.1016/S0034-4257(02)00078-0, 2002.

Comment 4. Line 66: resulting in potential underestimation of

Response: Thank you for the comment. This sentence has been revised to "resulting in underestimation of".

Comment 5. Line 74: Provincial atmospheric emissions of 12 dioxin-like UP-PCBs.

Response: Thank you for the comment. This sentence has been revised to "Provincial atmospheric emissions of 12 dioxin-like UP-PCBs".

Comment 6. Line 94: due to lack of in-depth data.

Response: Thank you for the comment. This sentence has been revised to "due to lack of in-depth data".

Comment 7. Line 131: Due to variations in
Comment 8. Line 132: land use/cover

Response: Thank you for the comment. This sentence has been written to "Due to variations in development status, industrial structure, energy structure and landuse/cover,".

Comment 9. Line 157: should be the priority concern

Response: Thanks for your comment. This sentence has been revised to "should be the priority concern".

Comment 10. Line 176: change "represented" to "were"

Response: Thanks for your comment. This sentence has been revised to "the per capita emissions were seriously high".

Comment 11. Line 182: change "extremely" to "much"

Response: Thanks for your comment. This sentence has been revised to "was much higher".

Comment 12. Line 205-206: were rising until 1990, but have been decreasing ever since

Comment 13. Line 206: was similar to that of other pollutants

Response: Thank you for the comment. This sentence has been written to "This trend was similar to that of other pollutants".

Comment 14. Line 224-226 is not clear, should rewrite

Response: The two sentences were revised as "For these provinces except Zhejiang, cement production was the largest source and their relative contributions to national cement production were 1.3-3.2 times of those for national GDP."

Comment 15. Line 245: "which indicated … regions" should be removed as there is no necessary connection

Response: Thanks for your comment. This sentence was removed.

Comment 16. Line 253: exist

Response: We apologize for this careless writing. This sentence was revised to "Large uncertainties still exist in current emission inventory".

Comment 17. In Fig. S6, "UP-PCBs emissions, g WHO-TEQ" in blue should be removed

Response: We apologize for this layout. This sentence was removed in supplement.