

## 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 #2

**General comments:** This manuscript developed the emission inventory in Chinese mainland for 12 dioxin-like UP-PCBs congeners from 66 sources with a resolution of  $0.1^{\circ}\times 0.1^{\circ}$  from 1960 to 2019. The characteristics of geospatial and temporal variations in total, per capita emissions and emission intensities were also investigated. This UP-PCBs emissions provides an essential data support for the assessment of human health risks from exposure to PCBs and for policy-makers to optimize PCBs mitigation strategies, and key input data for environmental modeling of PCBs. In general, the paper is well written, although some places require modification and clarification. 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.** This study established emissions inventories of 12 dioxin-like UP-PCBs for various sources by using the economic activities and corresponding EFs. But the EFs listed in Table S1 are total EFs for the sum of 12 dioxin-like UP-PCBs. How can obtain the EF for each UP-PCB from various sources? The detail EFs for individual dioxin-like UP-PCB should be presented.

**Response:** To estimate emissions for each PCB congener, emission profiles of 12 dioxin-like UP-PCBs were acquired by a thorough literature review. For source without data, emission profile of similar source was applied. Table S2 for final choices of emission profile for each source and corresponding references can be seen in supplement.

**Comment 2.** In the Section 2.3, for other sources, gridded industrial coal consumption from PKUFUEL was used as surrogate. This surrogate data for spatial allocation is not appropriate for some emission sources. I suggest gridded field fire data can be used to

allocate the emissions from open biomass source. The surrogate data for waste incineration should use the spatial distribution of incinerator in different provinces.

**Response:** Open biomass including wildfire and agricultural waste burning were included in PKU-FUEL. The proxy used for allocation of these sources in PKU-FUEL was actually gridded field fire data from GFED v4. We made corresponding description as below.

“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.”

However, information about spatial distribution of incinerator was hard to acquire, so industrial coal was used as proxy to disaggregate emission in space in current inventory and also in PKU-FUEL. We noted that this would result in spatial bias of inventory. We made corresponding description in conclusion section.

Line 253-254. “Large uncertainties still exist in current emission inventory due to insufficient EF data (e.g. source emission profile) and lack of in-depth source information (e.g. geolocation of point source).”

Line 256-258. “Future works might focus on acquiring detailed home-made EFs and developing more accurate method in allocate emission across space and time to estimate emissions more realistic and reduce temporal and spatial biases.”

**Comment 3.** Line 148, Fig. 3 shows UP-PCBs emission profiles for 9 source categories in Chinese mainland. But Fig. 3 shows the emission profiles for 12 dioxin-like UP-PCBs. The results (line 152-155) for emission profiles for 12 UP-PCBs from various sources also can not be seen from Fig. 3. The authors should add one figure to show the 12 UP-PCBs emission profiles for 9 source categories.

**Response:** Sorry for the mistake and it was revised as “Fig. 3 shows composition profiles of 12 dioxin-like UP-PCBs emissions in Chinese mainland.” In addition, we added one figure showing emission profiles for 9 source categories (Fig. S1) and a table of detailed composition profile for each source (Table S2).

**Comment 4.** Line 214-215, Cement production was the 215 major contributor across Chinese mainland from 1960 to 2019, followed by waste burning and industry boilers sector. This result and be seen from Fig. 4A, not Fig. S5.

**Response:** We apologize for this careless citation. We revised this sentence as your comment.

“Fig. 4A shows temporal trends of emissions from different regions in Chinese mainland. Cement production was the major contributor across Chinese mainland from 1960 to 2019, followed by waste burning and industry boilers sector. In general, cement production emissions were increasing from 1960 to around 1995 except for Northwest and Northeast China. From 1995 to 2019, the contribution of iron & steel sector increased a lot in North China. In the same period, the emission contribution from industry boilers sector was substantial increased in East, Central, Southwest and Northeast China (Fig. S6).”

**Comment 5.** The spatial resolution of Fig. 2 should be given.

**Response:** Thank you for the comment. The spatial resolution of Fig. 2 was  $0.1^{\circ} \times 0.1^{\circ}$ .

We added it in the caption of Fig. 2.

“Fig. 2. Spatial distribution of emission densities across Chinese mainland (the resolution is  $0.1^{\circ} \times 0.1^{\circ}$ ) and sum of emissions from various emission sources in different regions in 2019.”

**Comment 6.** Line 50, H. et al., 2004? Please check it.

**Response:** We apologize for this incorrect reference. The correct reference was revised in this sentence.

“key input data for environmental modeling of PCHs (Gluge et al., 2016; ter Schure et al., 2004)”

**Comment 7.** Line 105. “Total population” should be gridded population density, and the data source should be given.

**Response:** “Total population” has been replaced with “Gridded population density”.

“Gridded population density (Oak Ridge National Laboratory, 2020)”

**Comment 8.** The representative meaning of  $\sigma$  in eq. 1 should be given.

**Response:**  $\sigma$  is the standard deviation of log-transformed EFs. Explanation for meaning of  $\sigma$  was added in the main text accordingly.

“ $\sigma$  is SD of log-transformed EFs”

**Comment 9.** The year should be given in Fig.2 and Fig. 3.

**Response:** The captions of the two figures were revised as follows.

“Fig. 2. Spatial distribution of emission densities across Chinese mainland (the resolution is  $0.1^{\circ}\times 0.1^{\circ}$ ) and sum of emissions from various emission sources in different regions in 2019.”

“Fig. 3. Emission profiles for 12 dioxin-like UP-PCBs in Chinese mainland in 2019. Pie chart for PCBs congeners in terms of mass (A) and TEQ (B), respectively.”

**Comment 10.** The unit of y-axis in Fig. S5 should be given.

**Response:** The title (UP-PCBs emissions, g WHO-TEQ) for y axis was added for Fig. S5 (now is Fig. S6). Please check Fig. S6 in supplement.