

## **Response to reviewers' comments**

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Title: Re-volatilisation of soil accumulated pollutants triggered by the summer monsoon in India

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We would like to thank the reviewers for their thoughtful reading, comments and questions, which considerably helped to improve this manuscript. We have addressed all comments below and have indicated the corresponding **modifications** in the revised version of the manuscript which is **attached to this response (changes highlighted)**.

### **Colour coding below:**

- (1) comments from Referees,**
- (2) author's response,**
- (3) author's changes**

### **Anonymous Referee #1**

Does modeling study point a specific region as a source of re-volatilised pesticides in the study area?

Under on-going primary emissions (which applies for PCBs and DDT until present day, and for  $\alpha$ -HCH until 1980s), the highest absolute air-soil exchange fluxes,  $F_c$ , are simulated for southern India, somewhat lower for northern, and least for central India (see also Fig. 4). The  $\alpha$ -HCH air-soil exchange flux, which remains long after the ban, is found higher in the northernmost zone than in southern and central India, corresponding to a somewhat higher soil burden in the north (Fig. S3a). (Multi-decadal simulation using the 1D multimedia mass balance model, Sections 2.3 and S1.4.2).

The exchange fluxes are resulting from a combination of the historical emission distributions and fate, the latter latitudinal dependence being dominated by differences in climate across latitudinal zones.

### **Anonymous Referee #2**

The authors discussed the re-volatilisation of the nowadays-banned POPs accumulated in soils. The abstract is too simple; there was not description on the data, the methods, and models used. The way of the method validation is also not very clear.

Abstract now extended, including quantification of concentrations in air and drop of these, identification of models used. sentences to abstract. **Clean air masses from the Indian Ocean, advected with the onset of the summer monsoon, are found to reduce concentrations of hexachlorocyclohexane (HCH), dichlorodiphenyltrichloroethane (DDT) and its derivatives, endosulfan and polychlorinated biphenyls (PCBs) in air at a mountain site (all in the range 5-20 pg m<sup>-3</sup>) by 77, 70, 82 and 45 %, respectively. The analysis of fugacities in soil and air suggest that the arrival of summer monsoon triggers net volatilisation or enhance on-going re-volatilisation... (after the first sentence of the abstract) The response of the air-soil exchange was modelled using a regional air pollution model, WRF-Chem-PAH/POP. The results suggest that... (before the last sentence) Using a multidecadal multi-media mass balance model, it is found that... (beginning of the last sentence of the abstract)**

Quantified the drop in air concentration in the text (Section 3.1, lines 168-169) using the same matrix: **concentration changes i.e., 77, 70, 82 and 45 % for  $\Sigma_4$ HCH,  $\Sigma_6$ DDX,  $\Sigma_3$ Endosulfan and  $\Sigma_7$ PCBs**

Specific Comments:

Line 18: Clean air masses from the Indian Ocean, advected with the onset of the summer monsoon, are found to trigger or enhance re-volatilisation of the nowadays banned chemicals hexachlorocyclohexane (HCH) and polychlorinated biphenyls (PCBs) from background soils in southern India.

Comments: Re-volatilisation of the chemicals in soil always exists at the boundary between soil and air, and cannot triggered by clean air masses from the Indian Ocean.

True. rephrased accordingly (line 19-20)

Line 29: Semivolatile substances (i.e., vapour pressure at 293 K in the range  $10^{-6}$  – $10^{-2}$  Pa) tend to re-volatilise from land and seasurfaces to which they had previously been deposited, once a level of contamination in chemical equilibrium with air pollution is reached.

Comments: This statement is not correct since both the deposition and re-volatilisation co-exist regardless if the equilibrium is reached or not.

Thnaks, corrected accordingly (line 29-30)

Line 56: Thus far, studies on environmental exposure of the Indian subcontinent have been mostly limited to urban areas (Chakraborty and Zhang, 2012; Sharma et al., 2014; Chakraborty et al., 2015), while the continental background was scarcely addressed.

Comments: The following three published papers should be addressed in the manuscript, since these three papers studied the SVOCs in Indian air and soil on a national scale.

Li, W. L., Ma, W. L., Zhang, Z. F., Liu, L. Y., Song, W. W., Jia, H. L., Ding, Y. S., Nakata, H., Minh, N. H., Sinha, R. K., Moon, H. B., Kannan, K., Sverko, E., Li, Y. F., 2017, Occurrence and Source Effect of Novel Brominated Flame Retardants (NBFRs) in Soils from Five Asian Countries and Their Relationship with PBDEs, *Environmental Science & Technology*, 51, 11126-11135

Li, Wenlong, Ma Wan-Li, Jia Hongliang, Hong Wenjun, Moon Hyo-Bang, Nakata Haruhiko, Minh Nguyen Hung, Sinha Ravindra Kumar, Chi Kai Hsien, Kannan Kurunthachalam, Sverko Ed, Li Yi-Fan, (2016). Polybrominated Diphenyl Ethers (PBDEs) In Surface Soils across Five Asian Countries: Levels, Spatial Distribution and Source Contribution. *Environmental Science & Technology*. 2016, 50 (23), 12779–12788

Hong, Wen-Jun, Hongliang Jia, Wan-Li Ma, Ravindra Kumar Sinha, Hyo-Bang Moon, Haruhiko Nakata, Nguyen Hung Minh, Kai Hsien Chi, Wen-Long Li, Kurunthachalam Kannan, Ed Sverko, and Yi-Fan Li, 2016, Distribution, fate, inhalation exposure and lung cancer risk of atmospheric polycyclic aromatic hydrocarbons in some Asian countries, *Environmental Science & Technology*. 2016, 50 (13), 7163–7174

Thanks, included results and citation Li et al. 2016 (lines 156-160). The other two papers address pollutants not studied here.

Page 205: The differences in concentrations before and during monsoon are significant ( $P < 0.05$ , t-test) in south, central and parts of northern India (Fig. 3b,c).

Comments: In Fig. 3, there is no Panels b and c indicated!

Corrected (line 209)

Line 225: but soil concentrations only decreased for p,p'-DDT, while they have levelled off for  $\alpha$ -HCH, or are even still on the rise (PCB153, Fig. S4).

Comments: There is no Fig. S4!

Corrected (line 229)

Line 260: Both, field measurements and modelling results reveal a thus far overlooked mechanism of pollutant cycling over the Indian subcontinent, i.e. monsoon-driven mobilization from previously contaminated soils.

Comments: Rewrite the sentence.

Followed for better clarity, and corrected “The results of both the field measurements and modelling results of this study indicate a so far overlooked mechanism of pollutant cycling over the Indian subcontinent, i.e. monsoon-driven mobilisation of POPs from previously contaminated soils.”