

## ***Interactive comment on “Atmospheric deposition of polybromodiphenyl ethers in remote mountain regions of Europe” by L. Arellano et al.***

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Title: Atmospheric deposition of polybromodiphenyl ethers in remote mountain regions of Europe

Summary: The atmospheric Deposition profile for polybromodiphenyl ethers (PBDE) were investigated for remote alpine regions in Europe. Four characteristic sites in Europe were selected representing four different alpine regions in Europe. Characteristic site-specific deposition fluxes were estimated based on concentrations levels as well as meteorological conditions. The predominant contribution of trans-continental transfer of PBDEs from North American sources into Europe were assumed for the Scottish as well as the Pyrenean location. Secondary emission sources were found, dependent

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on ambient temperature, particulate distribution as well as precipitation events. Photochemical transformation is considered an important degradation pathway throughout the transport and deposition of PBDEs into the here investigated high-alpine remote locations.

General comments In line 17/ introduction it is stated that the environmental levels of the indicator PBDE are still increasing. This postulate is based upon references from 2000 – 2007). As stated in a more recent paper (deWit et al. 2010, Sci. Tot. Environ. 408: 2885-2918, Tanabe & Ramu 2012, Mar. Poll. Bull. 64/7: 1459-1474 and others), PBDEs are on a downward trend now. Please revise your introduction. In general, the first paragraph of the introduction dealing with PBDE and their relevance as environmental pollutants in the biotic compartments is based upon older references, up-date should be considered. At line 20 it is stated, that atmospheric deposition data on PBDE is scarce. During the recent years atmospheric deposition information on selected PBDEs is reported frequently. Please refer to Newton et al. , 2014, Environmental Sci. Proc. & Impacts. 16: 298-305, Tian et al. 2001, Env. Sci. Technol. 45/11: 4696-4701., Sofuoglu et al. 2013, Environ. Poll. 177: 116-124, Tlili et al. 2012, Water Air Soil Poll. 223/4: 1543-1553, Li et al. 2010, Sci. Tot. Environ. 408/17: 3664-3670 and many more. P22854/L4/5: “The fractions were vacuum-evaporated to 1mL” . Rotary evaporation based volume reduction requires usually a vacuum controlled treatment in order to reduce potential loss of target chemical. Please provide information on the procedures for vacuum control during rotary evaporation of extracts.

In order to assess accurate deposition fluxes as well as selective distribution processes in the deposition samples, parallel quantification of atmospheric samples (high vol. air sampling and/or passive sampling) is usually done. Previously, air samples have been taken and analysed for POP analysis at these respective sampling sites (in the frame of a finalized EU-project). A re-examination of this sample material is advised in order to assess accurate deposition pathways. The pearsons distribution usually assumes a normal distribution (Gaussian) of the data to be compared. Has the statistical distribu-

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tion for the PBDE groups been verified and tested for this type of statistical comparison? Please add information about the statistical procedures in the methods section.

#### Detailed Comments

Page 22852/line 25: "Whatmann glass fiber filters (GF/B, 45mm diameter, 1 $\mu$ m retention size": 1.) Typo - change name to Whatman. 2.) Please add information about city and country of origin for the providing company (as done below for Ehrendorfer) P22852/L26: "total particle mass," This term is misleading and refers to the mass of a single particle. I, therefore recommend to change the term into "total mass of the collected particles" P22853/ L4: "Trace analysis solvents, isooctane, n-hexane, chloromethane, cyclohexane, methanol, and acetone, were from Merck" Please provide information about the quality of the used solvents P22853/L6: "aluminium oxide were cleaned by Soxhlet extraction with dichloromethane:hexane (1:1, v/v , 24h) and were activated by overnight heating" 1.) Aluminium is not an English term – change to Aluminum 2.) Aluminum oxide is usually referred to as Alumina. 3.) The usage of a total activated alumina will ultimately lead to complete retention of all PBDE compounds (almost irreversible). Therefore Alumina is usually deactivated with a defined amount of deionized water. Please provide this information or refer to an earlier method description. P22853/L10: "wrapped into aluminium foil" 1.) Typo – change to Aluminum 2.) usually the aluminum foil is pre-cleaned before usage. Please provide information on this procedure. P22853/L13 . . . . "The standard solution contains 14 PBDEs congeners being two tribromo BDEs . . ." It is recommended to summarise the target PBDEs in table form. Please provide structural information, IUPAC name and CAS numbers for the interested reader. P7 P22853/L21: "from the freeze-dried filters" Provide information on the freeze-drying method including instrumentation. P22854/L3: "aluminium oxide" Please replace aluminum oxide with alumina throughout the text. P22855/L7: "The three-day back trajectories were calculated by the Hybrid Single-Particle Lagrangian Integrated Trajectory" Please provide information about the HSPPLIT version used for this study. P22857/L20: "The mean deposition PBDE fluxes were

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similar at all studied sites with differences of two-fold at the most. The most abundant PBDE congener was BDE209, with fluxes, ranging from 71 ngm<sup>-2</sup> mo<sup>-1</sup>" No information about the calculation method is provided. Please describe in detail (in the methods section) the calculation method for the flux estimation or refer to an earlier paper. P22859/L2: "the limit of quantification" The Limit of quantification (LOQ) calculation is not sufficiently explain the "Quality control" section. Please add information on this QC criterion under "quality control" P22860/L27: "BDE153/BDE154 ratios which should be lower than 1 to indicate significant contributions." Please provide evidence, that this ratio is such an important threshold value. Refer also to a previous paper to strengthen your argumentation. Figure 8: R = 0.476 and 0.453 (r<sup>2</sup> = 0.2265 and r<sup>2</sup> = 0.205) are not considered as a significant ratio based on this regression calculation. Be careful with interpretation of this relationship!

Recommendations The Manuscript has a few severe shortcomings on the method description and the data interpretation. In addition the relationship between the air mass trajectories and the PBDE distribution patterns in the total deposition samples is weak because the deposition process selectively washes out PBDEs (mainly bound to particles) from the atmosphere. For this type of assessment, high volume air samples would be better suitable for a LRT based evaluation. However, if the authors are able to respond to the above raised concerns, suggestions and comments. I am willing to review a potential resubmitted paper. The here presented manuscript is thus not publishable in the present form and need major revisions before accepted for publication at ACP.

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Interactive comment on Atmos. Chem. Phys. Discuss., 13, 22847, 2013.

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