

We appreciate the comments and suggestions made by Dr. Kallenborn. They will be taken up in the revised manuscript. To facilitate the review process, we include our answers in blue italics under the comments of the reviewer.

Sampling and methods: Please provide detailed information on sampling period, type of deposition sampler (type, version, provider, city and country of origin) and all consumables/chemical's used for field- and laboratory work (incl. type, quality, provider, city and country of origin) or refer to an earlier peer-reviewed paper where the unmodified methods is described in detail as required for scientific descriptions. The here cited Carrera et al 1998 paper is only describing the SPE sample preparation method but not the detailed deposition sampling procedures. The dimensions, sampling schedule and origin/type of sampling units were not adequately described.

In the revised version of the paper, we are now including a complete description of the consumables/chemicals used. In addition we are also referring to the paper published by Carrera et al, ES&T 36, 2581-2588 (2002), in which more details on sampling procedures are reported.

Concerning sampling, as stated in the manuscript, "Monthly samples were taken at all sites from May 2004 to August 2006 except at Lochnagar where sampling was performed biweekly from June 2004 to March 2007".

The deposition samplers were made in the lab using stainless steel or Teflon coated containers of different volumes and sizes. Now, in the manuscript we are providing more information on the devices used.

Please explain in detail, why 12C instead of 13C standards (for recSTD as well as ISTD) were applied for the GC/MS quantification. There are plenty of commercial 13C labeled PBDE standards available for this type of quantification methods.

We began the analysis of the deposition samples in 2004 following the method described in the manuscript. Meanwhile, we optimized a new method that used ¹³C standards and took into account possible BDE 209 degradation in the injection system during the chromatographic analysis. The new method was described in the reference Vizcaino et al. (2009) included in the manuscript. Some aspects of this new instrumental method were followed for the analysis of the rest of deposition samples (injection system, ions used for quantification). We did not find significant quantitative differences in PBDE deposition flux data when using the initial or ¹³C standards. Nevertheless, to avoid possible influences on data variability all deposition samples were quantified using the same standards.

PBDE209 (Deca) was analysed in parallel with 13 other PBDE congeners. Usually the analysis of PBDE 209 is done differently compared to less brominated congeners. Short-columns must be used for a reliable quantification due to well known analytical restrictions. In addition, PBDE 209 is highly photosensitive and tend to decompose in standard solution after rel. short time. Please reflect on these specific challenges in the introduction as well as the QC section.

We took into account all these issues when analyzing these contaminants. We used a short column (15 m) with less film thickness (10 um) in order to avoid all these problems. These issues are already described and discussed in the paper from Vizcaino et al. (2009) which is cited in the manuscript. The instrumental methodology described in this paper demonstrated to be suitable for the analysis of all BDE congeners (including BDE209) in only one single injection.

In order to provide a reliable threshold for the flux calculations (the entire discussion is based upon these values), the application and active usage of a limit of quantification (LOQ), usually defined as 5 x individual blank is advised (discuss this in the QC section).

A paragraph on MQL has been included in the QC section.

Detailed comments:

P22852/L25: "pre-weighed Whatmann glass fiber.." 1.) Type, please change to "Whatman".

Done

2.) Please provide information on the pre-cleaning procedures for the GFFs as well as the C18-disks and our sampling devices including the bulk samplers (cleaning/rinsing procedures etc).

The pre-cleaning procedure for C18-disks and bulk deposition samplers have been included (for GFFs it was already reported in page 22853 L9). In addition, references of previous work reporting more details on these procedures have been added (Quiroz et al., 2008).

P22853/L25: "...dried over anhydrous sodium sulfate." Add information on the drying procedure as well as the quality/ origin of the Na₂SO₄ applied.

We have added the information on the drying procedure. Information on quality/origin of the Na₂SO₄ was already reported in the 2.2. Chemicals section (page 22853, L6).

P22854/L12: "SGE-BPX5 MS" Please, provide information on the origin of this capillary column (including film thickness, provider, city and country of origin).

This information has been included.

P22854/L24: "PBDE determinations were performed by the internal standard method."

This sentence is difficult to understand. The expression "Performed by the internal standard method" is easy to be misinterpreted. I suggest changing the sentence to "The target PBDEs were quantified with the above mentioned internal standards".

Done

P22854/L26: "...was filtered and solid-phase extracted as a real sample" . This sentence is difficult to understand. The sentence should be changed to "filtered and solid-phase extracted like a real sample."

Done

P22855/L4: " 0.66 to 47 ng m⁻² mo⁻¹" This dimensions imply flux-calculations. I don't see that such a dimension make sense for LOD determinations! I assume that this is a typo and you mean ng total or ng/m², If not! Please recalculate the LOD values in ng/m²

Yes, it was a typo, they are ng/m², we also include LOD values in ng total in the new version.

P22860/L3:" Highest photodegradation is expected to occur at highest altitude and the results of the above.." Please Discuss why PBDE209 is still found in considerable levels at the high-altitude sites although the compound is known to be highly photosensitive.

BDE209 in the environment cannot be considered highly photosensitive. It is true that it can be degraded by photooxidation but this degradation does not take place at 100%. Several laboratory studies indicate that BDE209 half lives are longer in environmental conditions or when BDE209 is adsorbed in natural supports (sediment, soils or particles) than BDE209 adsorbed on artificial supports or in organic solvent solutions (see for example Ahn et al, Environm Sci Technol, 40, 215-220 (2006); Lagalante et al., Environ International, 37, 899-906 (2011); Bezares-Cruz et al., Environ Sci Technol, 38, 4149-4156 (2004)).

Raff and Hites (Environ Sci Technol, 41, 6725-6731 (2007)) indicated that photolysis of BDE 209 in the gas phase is very high, but BDE209 is mostly associated to particles which makes this compound more resistant to photochemical transformation. Similar processes have been described for other organic pollutants associated to atmospheric particles like PAHs. In

addition, BDE209 has been found in environmental samples from other remote sites than those considered in the present study (see for example: Bartrons et al., Environ. Pollut. 159, 1816-1822 (2011); Wang et al, Environ Sci Technol, 39, 7803-7809 (2005); Breivik et al., Environ Sci Technol, 40, 4612-4618 (2006); Arellano et al., Environ Sci Technol, 45, 9268-9275 (2011)) indicating that a significant fraction of the BDE209 emitted to the environment can persist enough in air to be transported over long distances.

P22862/L10: “Such particle diversity is also a likely cause for the lack of correlation between BDE209 and total particle deposition”. The GFF method used for particle collection in the here presented study is not sufficient to quantitatively collect the complete particle fraction deposited onto the ground. Usually GFF (Whatman) has a limiting “cut-off” (most GFFs used for High-Volume air sampling for POP analysis have a cut-off of 50 m). Thus, the fine fraction is not accounted for at all. In addition, the particle composition is not examined as well (neither with denuder techniques nor with SEM based examination). Therefore this scientific speculation cannot be supported by the here provided data material and should thus be omitted.

We agree with Dr Kallenborn that this statement is speculative as we did not analyzed particle composition. We though these differences in particle diversity with altitude could be reasonable. We will eliminate this paragraph from the final version.

P2873/L31: “sYstem” Typo.

This is not a typo; Y is capitalized here to remark the origin of the acronym READY.