Response to the reviewer's comments:
We are grateful to Dr. Kallenborn for his suggestions and comments on the manuscript. Here, we describe the changes introduced into the revised version following his comments and our replies.

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 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.

We have updated the information on PBDE trends in the introduction. However, the recent papers do not show defined increasing or decreasing concentration trends. The review paper by de Wit et al (2010) shows that PBDE concentrations are increasing in air, biota and sediments. The paper by Tanabe and Ramu (2012) shows increasing PBDE concentrations in the marine mammals from the Japanese coastal waters. Among these, only one species showed a decrease in PBDE concentrations but during the 1990s. This paper concludes that there is still an increase of PBDE concentrations but slower than in the 80s and 90s. Recent specific studies have shown increasing PBDE concentrations in sediments (Bogdal et al, ES&T 2008, 42, 6817-6822), air over the Great Lakes (Ma et al., ES&T 2013, 47, 11457-11464), or air in the Canadian High Arctic (Su et al., Atmos. Environ, 2009, 43, 6230-6237)

In a recent study (Kirchgeorg et al, Environ Pollution 2013, 178, 367-374) on PBDE levels in a snow core from an Alpine site covering a time span before and after PBDE regulation, no declines after regulation were observed. In this paper several references on PBDE temporal trends in different matrices are quoted and no temporal trend could be concluded because some papers report increases (the above cited Bogdal et al, 2008; Hermanson et al., 2010, ES&T 44, 7405-7410; Kohler et al 2008, ES&T, 42,6378-6384; Meyer et al, 2012) and others decreases (Schuster et al., 2010, ES&T 44, 6760-6766 and 2011, ES&T 45, 7291; Gevao et al., 2014, Sci. Total Environ. In press).

Furthermore, in a recent review on PBDEs in the environment by Law et al Environ Int. 2014, it was remarked that "more data on temporal trends of PBDE concentrations in a variety of matrices and locations are needed before the current status of these compounds can be fully assessed"

Now, we have changed the introduction and a more accurate description of the state-of-the-art of this topic is now described. In the revised version, it is stated that:

"In the last decades their environmental levels have increased significantly (Renner, 2000; Ikonomou et al., 2002; Usenko et al., 2007; Ma et al., 2013), although recent studies indicate no clear temporal trends in their concentrations in the environment during the last years after their regulation (Birgul et al., 2012; Kirchgeorg et al., 2013; de Wit et al., 2010; Law et al., 2012)."

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.

We have updated this paragraph by adding:

"The estimated global emission of these compounds due to the e-waste production was 280 tons at the end of the last decade (Robinson, 2009)."

We have also updated the second paragraph by changing the oldest references (Boon et al., 2002 and Hites, 2004) by more recent studies.

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.,Sofuogu 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.

This statement was referring to atmospheric deposition data on PBDEs in remote sites. In this paragraph we were commenting different studies on atmospheric deposition of PCBs and PAHs to remote background regions.

We were aware of most of the studies cited by the reviewer. In fact several of them were already included in Table 4 (e.g. Tian et al., 2011). We have now added the paper of Li et al., 2010 to this Table. In the case of the paper by Sofuogu et al., 2013, it describes the PBDEs levels in the atmosphere, not the atmospheric deposition fluxes. A comprehensive search of recent publications on PBDE atmospheric deposition fluxes to remote sites did not provide any further new reference, except the one by Newton et al., 2014 indicated by the reviewer and that by Arinaitwe el at (ES&T, 48, 1458-1466, 2014) which were not available at the time of submission of the manuscript. Now, they have been added to the text. Thus, we can still state that the information of PBDE deposition in remote sites is scarce.

Anyhow, we have rewritten this paragraph for clarification:

"This pathway has been shown to be relevant for the transfer of POPs, such as polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs), to remote background regions (Carrera et al., 2002; Fernandez et al., 2003; Usenko et al., 2010; Sun et al., 2006) but data on atmospheric deposition of PBDEs to remote sites is scarce (Meyer et al., 2012; Newton et al., 2014)."

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.

Vacuum is adjusted in our rotatory evaporation system by a vacuum regulator (membrane pump CVC2, Vacuubrand GMBH, Wertheim, Germany). Depending on the solvent, we work at pressures between 100-300 mbars. In addition, we never heat the sample flasks, which are immersed in water baths at 25 ^{o}C. This information is now included in the revised version of the manuscript.

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 ampling 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.

Our study is focused on bulk atmospheric PBDE deposition to remote sites. Our main interest was to obtain data on the magnitude of this process for assessment of the inputs of these compounds into the European remote background regions and the spatial and seasonal differences in PBDE deposition in Europe. The samples collected and analyzed in the present study allow fulfilling these objectives. The type of samples that we have collected and measured do not allow to determining phase distribution processes because they were collected as bulk atmospheric deposition and they remained between 15 and 28 days in the sample collector. This is the reason why, despite we separated the particle and dissolved phase compounds during sampling and each phase was extracted with specific procedures, the two phases were finally analyzed jointly. However, it is known that the physical-chemical properties of PBDEs involve that they are mostly associated to the particulate phase.

The air samples indicated by the reviewer were collected in mid-1990, almost one decade before the present atmospheric deposition sampling. This time period is too long for a reliable comparison taking into account the regulations on PBDE usage which have occurred during this interval. In any case, these samples were not analyzed for PBDEs nor stored for their later analysis.

The pearsons distribution usually assumes a normal distribution (Gaussian) of the data to be compared. Has the statistical distribution 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.

SPSS software package 21 was used for statistical analysis. All data were tested for normality using Kolmogorov-Smirnov test. In general, the data series did not follow a normal distribution and, as indicated in the manuscript, they were transformed into their log values for comparison with parametric tests. In some instances, non-parametric tests were performed and the results were in agreement with those obtained with the parametric tests. This information has been added in section 2.5. Data Analysis (in Material and Methods)

Detailed Comments

Page 22852/line 25: “Whatmann glass fiber filters (GF/B, 45mm diameter, 1 m retention size”:

1.) Typo - change name to Whatman.

Done

2.)Please add information about city and country of origin for the providing company (as done below for Ehrensdorfer).

This information was already indicated (page 22853, line 9, section 2.2. Chemicals).

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".

This term has been changed and moved to section 2.3.

P22853/ L4: "Trace analysis solvents, isooctane, n-hexane, dichloromethane, cyclohexane, methanol, and acetone, were from Merck" Please provide information about the quality of the used solvents.

All solvents used were for trace analysis. They were included in the Merck categories: SupraSolv® (for gas chromatography analysis) and UniSolv® (for trace organic analysis). These trade names have now been included in the manuscript to provide a more accurate information on the solvents used.

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

We have changed Aluminium oxide by Alumina as indicated by the reviewer.

2.) Aluminum oxide is usually referred to as Alumina.

This change has been introduced in the revised version

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.

Total activated alumina is obtained by heating at 350°C or higher temperature. In our procedure, alumina was heated at 120 °C and therefore it was partially activated. This method has shown to be reproducible and has been used extensively for the analysis of persistent organic pollutants. Its suitability for the analysis of PBDEs has also been confirmed (see reference Quiroz et al., 2008, included in the revised manuscript)

P22853/L10: "wrapped into aluminium foil"

1.) Typo – change to Aluminum

Done

2.) usually the aluminum foil is pre.cleaned before usage. Please provide information on this procedure.

Prior to use, the aluminum foil was rinsed with MilliQ water, acetone and n-hexane. This information has now been included in the revised manuscript.

P22853/L13. ": The standard solution contains 14 PBDEs congeners being two tribromo BDEs" It is recommended t summarise the target PBDEs in table form. Please provide structural information, IUPAC name and CAS numbers for the interested reader.

All this information has been included in Table S1 and Figure S1 (supporting information)

P7 P22853/L21: “from the freeze-dried filters” Provide information on the freeze-drying method including instrumentation.

The following information has been added to the manuscript:

“Frozen glass fiber filters were freeze-dried using an Edwards K4 Modulyo Freeze Dryer (Edwards High Vacuum International Ltd., West Sussex, England) during approx. 6 hours before weighing to obtain the total mass of the collected particles.”

P22854/L3: “aluminium oxide” Please replace aluminum oxide with alumina hroughout the text.

Done

P22855/L7: “The three-day back trajectories were calculated by the Hybrid Single-Particle Lagrangian Integrated Trajector” Please provide information about the HSPLIT version used for this study.

We used the on-line version of HYSPLIT and referenced it as they recommend. For elaboration of the cluster mean graphic, we used HYSPLIT 4 for PC as indicated in pg 22856, L2.

P22857/L20: “The mean deposition PBDE fluxes were similar at all studied sites with differences of two-fold at the most. The most abundant PBDE congener was BDE209, with fluxes, ranging from 71ngm2 mo-1” 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.

The monthly PBDE deposition fluxes were calculated by division of the total PBDE amounts measured in each sample (ng) by the surface area of the collection sampler (m^2) and the sampling period (days). Finally, the data were reported after normalization to one calendar month.

*This is now indicated in a new section: **2.5. Data Analysis,** included in Materials and Methods.*

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”

The following paragraph has been added to the QC section:

“Likewise, method quantification limits (MQL) were determined as the average blank values plus 5 times the standard deviation of the signal corresponding to each congener in the blank samples. They ranged between 0.85 and 42 ng m^{-2} (Table S2 and S3)”.

P22860/L27: “BDE153/BDE154 ratios which should be lower than 1 to indicate significant contributions.” Please provide evidence, that this ratio I such an important threshold value. Refer also to a previous paper to strengthen your argumentation.

We have modified this paragraph:

“The relative contributions of octaBDE can be assessed from the relative proportion of BDE183 and the BDE153/BDE154 ratio which should be higher than 1 (LaGuardia et al., 2006). In the samples considered for study the average values of this ratio were higher than 1 in all cases (Table 6) but the relative proportion of BDE183 was low (Table 3), indicating a low contribution of octaBDE technical mixtures in the European atmospheric deposition.”

Figure 8: R = 0.476 and 0.453 (r2 =0.2265 and r2 = 0.205) are not considered as a significant ratio based on this regression calculation. Be careful with interpretation of this relationship!

The correlation between BDE47/BDE99 ratio and the % of air masses flowing from central/east Europe is at the limit of statistical significance in Gossenköllesee and significant within 95% confidence interval in Redon. However, it is remarkable that both sites show the same trend and that this trend is coherent with the lowest BDE47/BDE99 ratio found in Skalnate, the easternmost site.

We have rewritten this paragraph to be more accurate with the statistical significance of these correlations:

“On the other hand, a negative correlation between BDE47/BDE99 ratios and percent of air mass trajectories flowing from central/east Europe has been observed at Redon (r = -0.453, p<0.05) and Gossenköllesee (r = -0.476, p = 0.05) (Figure 8). These coefficients are only statistically significant in the former and at the limit of statistical significance in the latter. The coincident trends observed in these two sites and the agreement of these low values with the characteristic ratio of the technical pentaBDE mixtures that accumulate in Skalnate, the easternmost site, suggest that this region may be a major source of these compounds.”

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 week 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.

Analysis of pollutants in atmospheric samples and correlation of the results with the calculated backwards air mass trajectories for the dates of the sampling period provides a direct link between the origin of the air masses and the air composition which can be used to identify long-range transport processes in remote areas.

On the other hand, sampling of atmospheric deposition in remote sites and calculation of the backwards air mass trajectories provides integrated pictures of the inputs of long-range transported pollutants into pristine ecosystems. Both approaches are complementary.

Similarly, in water studies anthropogenic impacts can be studied by direct analysis of pollutants in water and by the analysis of integrated pollutant exposure using sentinel organisms, e.g. the well-known “mussel watch” approach.

In the present study, we have followed this second approach. In our case, the results are solid because the long time of collection of atmospheric deposition (two years and four months), the intensity of sampling (monthly in three sites and bi-weekly in one) and the parallel sampling in four remote European sites provide a unique database for description of the long-range transport of organic pollutants to remote areas in a European perspective.

In addition, there is little ambiguity concerning the phase distribution of these compounds. They are known to be associated to the atmospheric particle-phase as consequence of their physical-chemical properties and this is particularly the case for BDE-209, but the presence of these compounds in particles of small sizes, e.g. <0.57 µm (Mandalakis et al Env. Poll. 157, 1227-1233, 2009), <2.5 µm (Deng et al., Env. Int 33, 1063-1069, 2007), allows them to travelling over long distances.

In our study, the correlations observed between backwards air mass origins and atmospheric deposition of these compounds are strong since they are based on the results from the analysis of a large number of samples covering two years and four months. Furthermore, for what concerns the inputs from the western air masses we have found the same correlations in two remote sites separated by 1600 km. In one of those, sampling was performed monthly and in the other bi-weekly. Among the four sites considered for study these are the two situated further to the west which again is consistent with the observed trend.

Last but not least, the experimental approach used in the present study has also been undertaken in previous cases. Thus, ter Schure et al (Environ Sci Technol 38, 1282 (2004)) in a study on the concentrations and distributions of these compounds in air and atmospheric deposition in a remote Baltic Sea area found that the PBDE congener pattern in rain was similar to that in air, concluding that rain deposition was related to atmospheric PBDE concentrations. Meyer et al., Environ. Sci. Technol. 46, 826-833 (2012) assessed the influence of PBDE emissions from densely populated areas of North America into the Canadian Arctic from the sampling and analysis of atmospheric precipitation in these remote sites. Li et al. J Environ Monitor,12, 1730-1736 (2010) determined atmospheric deposition fluxes in the Pearl River region (South China) and investigated their potential source regions through back trajectory analysis of air masses. Newton et al. Environ. Sci.: Processes Impacts 16, 298-305 (2014) collected bimonthly bulk atmospheric deposition in Arctic and subarctic sites of northern Sweden which were used for the analysis of PBDEs, PCBs, dechlorane plus and other pesticides and backwards air mass trajectories were used for assessing the origin of these compounds. There are other examples in the scientific literature documenting this approach.