Response to Reviewer 1’s comments

1) The text of this manuscript has very little, if any, relation to its title which says that it’s a guideline for using MBR to estimate POP fluxes. In fact, POPs is only mentioned once on each of the last two pages of text. ... As for the appropriateness of publishing this manuscript in Atmos. Chem. Phys., if the authors change the title to reflect what is actually in the text, then yes, it is appropriate.

We agree with the reviewer that the proposed title was not a good reflection of the actual contents of the paper. In response we have changed our title to: “Comparison of eddy covariance and modified Bowen ratio methods for measuring gas fluxes and implications for measuring fluxes of persistent organic pollutants”, which we think is a better summary of the findings brought forward in this study.

2) ... All these K estimations were made for fluxes measured over a mixed deciduous forest between about 30 and 45 m. This would be fine if POP fluxes were to be estimated there, but I suspect that deciduous forests are not POP “hot spots.” Nowhere in the text is there any mention of where POP volatilization fluxes are likely to be measured.

This comment is very similar to that received from Reviewer 2: “The data and example illustrated in this study is based on FLUXNET data on a tower above a tree canopy. Gradients in H2O and CO2 were apparent and allow comparison between EC and MBR derived fluxes. Importantly, MBR, based on the concept of turbulent diffusion, requires the measurement of a clear gradient. For the canopy scenario given here, would this be apparent (i.e. measurable) for POPs, given the heights where CO2/H2O data were collected (~30 and ~40 m) would likely result in concentration differences that could be non-existent for POP chemicals? While this study is not necessarily advocating the use of the FLUXNET/boreal towers for applying MBR to estimate POP fluxes, there is an implicit assumption that this will be the basis of follow up studies- is this the case?” And we feel that the same reply is applicable here as well.

The motivation for us to use a dataset from FLUXNET was its previous usage in the study of Choi et al. where the authors studied the fluxes of PAHs above a forest canopy. In that study, the authors reported a clear gradient in the concentrations of several PAHs in air at 30m and 40m height. The observed gradient was attributed to the presence of leaves in the canopy which were fully developed and provided a significant sink for the PAHs.

The forest filter effect describes the phenomenon in which the deposition of semi-volatile compounds to forested soils is larger than to non-forested soils due to the uptake in leaves and subsequent transport to the soil by shedding of leaves and waxes. The forest filter effect is thought to be most pronounced for chemicals with a log Koa between 7 and 11 and a log Kaw larger than -6. The compounds for which a significant downward flux to the canopy was observed in the study by Choi et al. included phenanthrene, anthracene and pyrene which have log Koa values of 7.5, 7.6 and 8.8 and log Kaw values of -2.8, -2.6 and -3.3 respectively which fit well within the range for the forest filter effect to be substantial. It can be expected that for other compounds with similar properties, a similar gradient could be measured above a developed forest canopy.

Currently we have no plans to do any flux measurements of POPs using FLUXNET/Boreal towers ourselves. However, we see a wide range of possible applications of the MBR method to measure fluxes of POPs and POP-like chemicals. We will include the following paragraph in the discussion section of the revised text to clarify and point out the broad range of potential applications:

“There is a wide scope for applying the MBR method to measure fluxes of POPs and POP-like chemicals in the atmosphere. A key data gap for many POPs is a lack of measurements of the
fluxes of POPs from dispersed sources to the atmosphere (McKone and MacLeod, 2003), and the studies by Rowe et al. for PCBs from the Hudson river (Rowe and Perlinger, 2012), by Perlinger et al. for HCHs and hexachlorobenzene over Lake Superior (Perlinger et al., 2005) and by Kurt-Karakus et al. with treated soils (Kurt-Karakus et al., 2006) demonstrate that the MBR method could help fill that gap. Our results reported in this paper imply that measurements of fluxes of POPs could be accomplished using the MBR method with passive air samplers instead of the active samplers that have so far been used in these studies, as long as the direction of flux does not change during the sampling period and the concentrations gradients are large enough to be measured.”

3) ... There is no discussion as to the surface and source conditions that need to be met for any of the flux estimation methods to work properly and yield reasonable results and how having “non-ideal” conditions will affect the flux estimations.

We agree with the reviewer that we did not provide a detailed specification of conditions needed to perform flux measurements using the MBR method. We believe that our new title: “Comparison of eddy covariance and modified Bowen ratio methods for measuring gas fluxes and implications for measuring fluxes of persistent organic pollutants” reflects the contents of our study better and that it makes it clear that we’re not offering step by step guidelines for the use of the MBR method.

There is a wide range of literature available which focuses mostly on the conditions needed to make reliable measurement with the eddy covariance approach, which we think is outside the scope of our study. Our study shows however, that changes in the direction of fluxes during the sampling period can affect the flux estimations with the MBR method. This is an example of a non-ideal condition that affects the MBR method but not the EC measurements which fits well within the scope of our study, where we compare the two methods.