We are grateful to the reviewers' thoughtful comments and have now amended the manuscript according to their points. Some of these comments have been extremely helpful. We have acknowledged the valuable contribution made by the reviewers in this manuscript. A detailed response to each of the reviewers' points is provided below and we have carefully revised the manuscript as a result (all revisions are highlighted in the text). We hope the manuscript is now acceptable for publication in ACP.

1. **One of the most significant developments in air sampling technology in recent years is the evolution of passive samplers. So far, there are several type of PASs have been developed for POPs study, such as PUF disk, PUF/XAD-4 combination, and XAD-2 cartridge. Have the authors already done any comparison, for example XAD-2 vs. PUF disk, before you chose XAD-2 for a five years program?**

   Response: So far, different passive air samplers (PAS) have been designed, which allowed samplers to integrate ambient concentrations over time scales as short as hours/days or as long as weeks/months/years. Based on the field calibration of polyurethane foam (PUF)-PAS and XAD-resin PAS, Gouin et al., (*Environ. Sci. Technol.*, 2008**, 42 (17), 6625–6630) recommended that PUF-PAS and XAD-PAS are suitable for obtaining atmospheric concentrations of POPs on the time scale of seasons and years, respectively. By coating XAD-4 onto PUF disk, the sorbent impregnated PUF disk (SIP) PAS is a modified version of the PUF-PAS. Months and seasons (3 months) of deployment are usually deployed for SIP-PAS.**

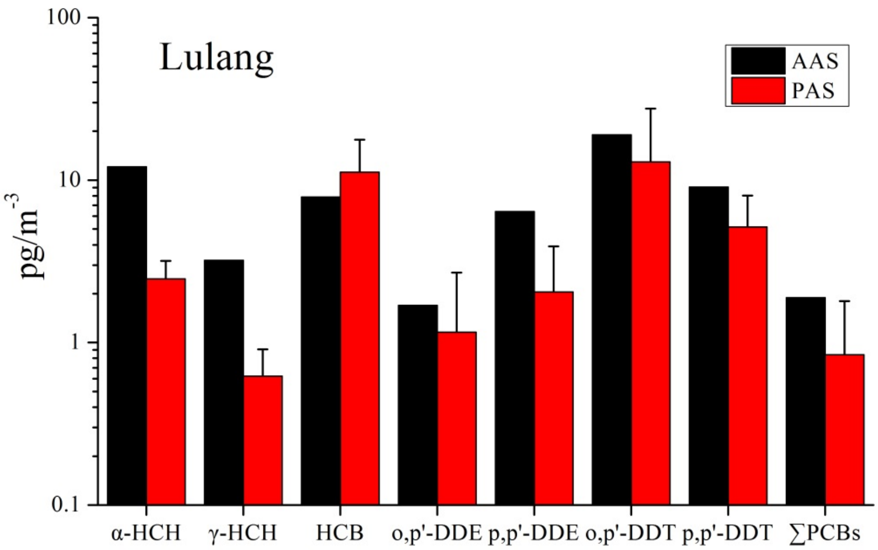
   Measurements in remote areas like the Tibetan Plateau are especially challenging due to the lack of electricity and high sampling costs. Due to these reasons, we finally chose XAD-PAS as the target sampler. If we use PUF/SIP-PAS, the advantage is we can get the seasonal variations of POPs, but the disadvantage is we need to travel across the Tibetan Plateau many times to collect samplers, which increased the sampling cost very much. Considering that we already know that XAD-PAS can give reliable result for year-round sampling, using XAD-PAS will provide benefits for both reliable POPs data and economic sampling cost. Before we conduct our field sampling we did not compare the above-mentioned 3 types of PAS, because we already know the proper deployment time for each of them.

   Reference: Gouin et al., Field Testing Passive Air Samplers for Current Use Pesticides in a Tropical Environment. Environ. Sci. Technol., 2008, 42 (17), pp 6625–6630

2. **XAD-2 based PAS has been deployed at 16 background sites across TP for 5**

years or even long. Although PAS has several advantages for spatial and long-term trends monitoring, the data quality have been always a concern for long-term deployment. The authors should be able to control or compare the PAS data with active sampler at one of these 16 stations. If there any data from an high-volume air sampler available, you may show them in the manuscript in order to prove the data quality generated from XAD-2 based PAS.

Response: Actually, in this study, both XAD-PAS and high-volume air sampler (AAS) were deployed in Lulang and details about the data comparison were provided in Text S3.



The DDTs derived from PAS were not very different from the corresponding AAS concentrations; whereas the low molecular weight OCPs, such as α-HCH and γ-HCH, showed some larger discrepancy (Figure above). This may be caused by different air masses being sampled and different adsorption characteristics for two kinds of samplers. Under these restrictions, the concentration variability within a factor of 2 -3 is deemed to be acceptable (Gouin et al., 2005). Therefore, the differences in the present PAS/AAS comparison (Figure above) were acceptable, which demonstrate that good agreement was found between the derived PAS concentrations and AAS results.

3. “The following compounds were measured and quantified: PCB 28, 52, 101, 138, 153 and 180; HCB; o,p’-DDE, p,p’-DDE, o,p’-DDT and p,p’-DDT; _-HCH, _-HCH, -HCH and _-HCH” in this work, I am wondering how you selected these OCP species and PCB congeners. Are these compounds mostly partitioning in vapor phase or particle phase?

Response: HCH isomers and DDT class chemicals are the most predominant

POPs species in Asia environment. HCH and DDT had been extensively used in China and India. Many studies demonstrated that fresh use of these two class chemicals are still occurring in some south Asia counties, such as India, Pakistan, Nepal and Thailand etc. The Tibetan Plateau is surrounded by China, India, Nepal and Pakistan, thus, its environment is most likely influenced by HCHs and DDTs that emitted from these surrounding countries. This is the reason why we mainly concerned about these chemicals. Regarding PCBs, PCBs (CB28, 52, 101, 118, 153, 138, and 180) were recommended for monitoring by the European Union Community Bureau of Reference and are also six ICES (International Council for the Exploration of the Sea) PCBs. These PCB congeners were selected as indicators of wider PCB contamination due to their relatively high concentrations in technical mixtures and their wide chlorination range (3-7 chlorine atoms per molecule). As the legacy POPs, HCHs (α-HCH, β-HCH, γ-HCH and δ-HCH), DDTs (o,p'-DDE, p,p'-DDE, o,p'-DDT and p,p'-DDT) and PCBs(CB28, 52, 101, 118, 153, 138, and 180) had been widely measured in other studies, which provided the opportunities for data comparison. All these are reasons why we chose these chemicals as target compounds.

High chlorinated PCBs have less volatility and mainly associate with particle phase, while low chlorinated PCBs are relatively volatile and present in gas phase. XAD-PAS are designed for mainly collecting POPs that normally dominate in the atmospheric gas phase. This is why high chlorinated PCBs were less detected in our study (PCB 28 and PCB 52) were regularly detected in samples, while highly chlorinated PCBs such as PCB 180 were never detected in samples).

4. **In the section of QA/QC, you reported that "Duplicate PAS were deployed to check the repeatability and the results showed the average relative deviation of concentrations between duplicates is generally low, which ranged from 9% to 18% for different compounds (Table S4). Uncertainties for chemical analysis were in the range of 25–35% and reasons for the uncertainties were discussed in Text S3." I am wondering how you can achieve a 9% to 18% relative deviation for duplicate PAS when the uncertainties of analytical method ranging from 25-35%. Frankly, I may trust the data when you report relative deviation even higher than 50%, but it is a surprise that you achieved such good results for duplicate PAS.**

   Response:In this study, duplicate samplers were deployed in 3~6 sites for consecutive 5-year monitoring. For every sampling year, the RSD of duplicates (sampler a and b) were calculated and then these RSDs were further averaged for the corresponding compounds. Finally, all RSDs for this 5-year monitoring study were average and RSDs between duplicates ranged from 17% to 24% for different compounds (Please see Table S3 in supporting information). First, our previous statement (RSD ranged from 9% to 18%) got wrong. We feel very sorry for this. Second, for some sampling

year, RSD of individual compounds can reach up to 48% for ∑PCBs and 30-40% for DDT class chemicals. However, when take all 5-year into account, the averaged RSDs (from 17% to 24%) are still low. Basically, the analytical uncertainty is the major driver of difference between co-deployed PASs, highlighting the robust nature and simplicity of PASs.

5. **In the section “3.1 Concentrations of POPs in the Atmosphere of TP” the authors used ng/sample for POPs concentration, while the unit was change to pg/m3 in the section “3.2 Spatial Distribution of POPs across the TP”. I will suggest using the same unit to avoid any confusing understanding.**

Response: Similar comments were also raised by reviewer 1#. Here, we deleted the comparison with unit of ng/sampler.

6. **In the section, “3.3 Dose the soil-air exchange (secondary source) affect the spatial pattern?” The authors calculated air-soil fugacity for selected substance to evaluate that regional re-evaporation from soil can be an important vector for atmospheric POPs. Generally, atmospheric POPs data generated from PAS might be with very high uncertainties more than what we can expect. Fugacity calculated based on such data set is very hard to guaranty the model results. Moreover, as it is a five-year program, it will be very helpful if the authors have collected the soil samples from 16 monitoring sites, and then they use the data from paired air/soil samples for fugacity calculation.**

   Response: We agree with the reviewer’s concern. As we discussed in response to reviewer 1, using a direct soil fugacity would have been preferable. Yet soil concentrations are not expected to vary quickly over time, so the current approach remains a good first estimation of air-soil gradients.

7. **Both Figure 1 and 2 should be improved for publication.**

   Response: We re-organized these figures, please see page 27 and 30 .