Dear editor,

We very appreciate the reviewers' comments, which surely improve our manuscript. According to reviewer's comments, the point-by-point responses to each of the comments are provided below. A 'track change' version of the manuscript is also attached.

Response to SC1

Response: Thank you for your comment on the manuscript regarding missing reference for the Figure 1. We have now provided the proper citation based on your suggestion. Please see the caption for Figure 1, p. 18, line 461, and p. 24, lines 544-545.

The cited reference is,
Karki, R., Talchabhadel, R., Aalto, J. and Baidya, S. K.: New climatic classification of Nepal, Theor. Appl. Climatol., 125(3–4), 799–808, doi:10.1007/s00704-015-1549-0, 2016.

Response to SC2

The authors thank SC2 (Dr. I.C. Yadav) for his inspiring comments. All the comments are addressed point-by-point below.

Comment 1: In my opinion, Hetauda is not an agricultural city; rather it is an industrial estate with a number of medium and small-scale industries particularly cement industries. on what basis author called it as agricultural cities. If you have a reference, plz cite them.

Response: The primary objective of our work was to assess the atmospheric level of various POPs in different land types, and to find the probable source area and to identify seasonalities of POPs. We did not aim to classify the city. We agree that Hetauda could be considered as an industrial city before 1990, since all the industries were full-functional. But after 1990, most of the industries are non-functional (e.g. Hetauda textile industry), either due to national policies or due to heavy power cut-off (up to 17h per day). At present, the industries in Hetauda are run for limited production e.g. (Hetauda cement factory ~60%). Rather, Hetauda is mostly surrounded by agriculture land and forest where use of OCPs could be the possible emission source. Hetauda is

the headquarter of Makawanpur district, the vegetable products of this district are sold in the surrounding markets, including even the capital city. Additionally, according to "statistical information on Nepalese agriculture 2070-2073" , district of agro-production capacity (crops and vegetables) has been increased gradually. Given the industrial activities became shrinking and agricultural activities are growing, we prefer to regard Hetauda as an "agriculture city".
Source: Government of Nepal Ministry of Agricultural Development (2014). Retrieved from: http://www.moad.gov.np/en/publication

Comment 2: What recovery standard and in what amount was used in this study?

Response: We used 20µl 1,2,3,5-tetrachloro-4,6-dimethylbenzene with the concentration of 100 pg μl^{-1} as recovery standard. Please see manuscript p. 5, lines 134-135.

Comment 3: The climate data such as precipitation and temperature need a reference.

Response: Thank you for your concern on the figure for the monthly climate variations. For this figure, we obtained the data from Department of Hydrology and Meteorology (DHM), Nepal. The figures are now provided with proper reference. Please refer p. 6 lines 59-60 in supporting information. This has been acknowledged in the manuscript. Please see p. 16, lines 456-457.

Comment 4: L75-76 in the supplement, is not clear as what amount of international standard was used. Also, PCB-209 may not be appropriate standard because PCBÂ 209 can be detected in urban environmental samples, especially due to the emissions from pigments and dyes where it is present as a byÂ product (Anezaki and Nakano, Environ Sci Pollut Res (2014) 21:998Â 1009). Hence, 13C-PCB-141 isotope labelled internal standard is most appropriate standard that compensates for errors and losses throughout the procedures.

Response: We don't know what does the "**international standard**" mean, do you mean "internal standard"? If you mean "internal standard", we used pentachloronitrobenzene (10 ng) and PCB-209 (2 ng) as the internal standard. There

are several studies which used PCB-209 as recovery standard (e.g. Chakraborty et al., 2015; Wang et al., 2016; Yadav et al., 2017). If a chemical can be used as recovery standard, it can generally be used as internal standard. We double checked the peak area of PCB-209 in the samples and QC solution (mixture of recovery and internal standard, but with the same concentration as real sample), we found peak area of PCB 209 in samples is very close to that found in QC, which implies our GC-MS is stable and no additional PCB-209 come from atmosphere to sample.

We appreciate your suggestion but internal standard can only indicate the fluctuations of instrument only recovery standard can be used to evaluate the errors and losses throughout the clean-up procedures!

References

Chakraborty, P., Zhang, G., Li, J., Sivakumar, A. and Jones, K. C.: Occurrence and sources of selected organochlorine pesticides in the soil of seven major Indian cities: Assessment of air-soil exchange., Environ. Pollut., 204, 74–80, doi:10.1016/j.envpol.2015.04.006, 2015.

Wang, W., Wang, Y., Zhang, R., Wang, S., Wei, C., Chaemfa, C., Li, J., Zhang, G. and Yu, K.: Seasonal characteristics and current sources of OCPs and PCBs and enantiomeric signatures of chiral OCPs in the atmosphere of Vietnam, Sci. Total Environ., 542, 777–786, doi:10.1016/j.scitotenv.2015.10.129, 2016.

Yadav, I. C., Devi, N. L., Li, J., Zhang, G. and Breivik, K.: Possible emissions of POPs in plain and hilly areas of Nepal: Implications for source apportionment and health risk assessment, Environ. Pollut., 220, 1289–1300, doi:10.1016/j.envpol.2016.10.102, 2017.

Comment 5: Why DDT is low in Hetauda despite being warmer and agricultural city? Any idea?

Response: In fact, the result was opposite to our expectation. Diminished use of technical DDT can be the likely reason behind the quantity observed in Hetauda. However, we have no accurate reason to explain the lower concentration of DDT in Hetauda. This deserves a future research.

Comment 6: In Fig 3, How do you interpret the concentration of HCB at the industrial site (K3), why it is so high?

Response: The fuel combustion in thermal power plants for industrial purpose during the dry season when there was a severe crisis of electricity from national grid might be the possible cause of high atmospheric HCB in an industrial area (K3). Besides

pesticides and byproduct of industrial reactions, fuel combustion is another important cause of HCB emission into the atmosphere (Wang et al., 2010). Bailey (2001) has estimated that ~ 30% of total global emission of HCB comes from combustion. Please see p. 12, lines 321-322.

References

Bailey, R. E.: Global hexachlorobenzene emissions, Chemosphere, 43(2), 167–182, doi:10.1016/S0045-6535(00)00186-7, 2001.

Wang, G., Lu, Y., Han, J., Luo, W., Shi, Y., Wang, T. and Sun, Y.: Hexachlorobenzene sources, levels and human exposure in the environment of China., Environ. Int., 36(1), 122–30, doi:10.1016/j.envint.2009.08.005, 2010.

Comment 7: Why authors only compare and discuss their finding with past studies from India and abroad but not with the past studies of the same region. Although not many studies done in this region, but few studies are well reported (eg. Yadav et al., 2016, Guzzella et al., 2016; Guzzella et al., 2011; Gong et al., 2014, Cizmas et al., 2015 and so on) that should be consulted to discuss the finding of present data.

Response: The main objective of the paper was to assess the source area and strength, however, we compare the present study outcomes with India and abroad because these areas have similar climate as the present study area. Regarding the data reported for Nepal cities, they were compared in supporting information (Table SI-7, p 21).

Comment 8: Another important concern for me is that author did monitoring of PAS for 1 year, however they compared and discussed their data with previous international studies that were based on short-term monitoring of PAS.

Response: We got the concentrations of POPs from one-year monitoring. We tried our best to find the studies which were similar to ours (with 1 year sampling period), but we did not obtain. So, we have to use our values to compare with the concentration data which were from the region of similar climate though they were the results of short-term study.

Comment 9: Table 1 should also include past studies of the same region to better discuss your finding.

Response: Thank you for your suggestion. We are sorry to say, due the size limitation of the table 1 we are unable to include values from the previous works, instead they are presented in the Table SI-7. Please see p. 21, supplementary materials.

For RC1

Comment 1: Although the authors do a good job at explaining how site specific sampling rates are determined using Depuration Compounds, there is no information on how the effective air sample volumes are calculated. I am specifically wondering about the more volatile compounds like HCB and a-HCH (and perhaps also g-HCH) that will equilibrate or approach equilibrium during the 2-month deployment periods. This approach to equilibrium should result in reduced effective air sample volumes for these compounds relative to the other POPs. This effect may also have a role to play in observed seasonality since the sorptive capacity of the PUF disk is temperature dependent.

Response:

Effective air volume for all the chemicals were calculated using the formula given by Harner et al., (2013),

$$V_{Air} = (K'_{PSM-A}) + (V_{PSM}) \times \left\{1 - \exp\left[\frac{k_A}{K'_{PSM-A}} \times \frac{1}{D_{film}}\right] t\right\} \quad \text{..... (1)}$$

Where,

V_{Air} = effective air sample volume (m^3)

$K'_{PSM-A} = K_{PSM-A} \times \delta_{PSM}$

Shoeib and Harner, 2002 have used a relationship between K_{PSM-A} and K_{OA} as

$logK_{PSM-A} = 0.6366 logK_{OA} - 3.1774$ ..... (2)

δ_{PSM} = density of PUF (g/m^3)

V_{PSM} = Vol. of the passive air sampler (m^3)

k_A = air side mass transfer coefficient ($m\ d^{-1}$)

D_{film} = effective film thickness (m)

t = time (day)

k_A = the sampling rate R ($m^3\ d^{-1}$) divided by the surface area of the PUF disk ($341 cm^2$)

The air concentrations of compounds were obtained by dividing the concentration (pg/sample) by effective air sample volume (V_{Air}).

In general, average concentrations, of HCB, α-HCH, γ-HCH, PCB-28, PCB-52, α-endo and o,p′-DDE increased by 46%, 40%, 26%, 25%, 18%, 13% and 11% respectively. There was no significant variation in the concentrations of less volatile chemicals. Though the concentrations of some of more volatile analytes changed, in overall, there were no changes in "distribution patterns" and the "seasonality" from previous analysis. All the numbers, figures and tables are reformed based on the new calculation (please see Figures 2-4 in the manuscript; Figures SI-3-12 in supplementary materials; Table 1 in the manuscript and Tables SI-5-11 in supplementary materials)

For details about calculation, please see pp. 6-7, lines 159-173 of the manuscript.

References

Harner, T., Su, K., Genualdi, S., Karpowicz, J., Ahrens, L., Mihele, C., Schuster, J., Charland, J. P. and Narayan, J.: Calibration and application of PUF disk passive air samplers for tracking polycyclic aromatic compounds (PACs), Atmos. Environ., 75, 123–128, doi:10.1016/j.atmosenv.2013.04.012, 2013.

Shoeib, M. and Harner, T.: Characterization and Comparison of Three Passive Air Samplers for Persistent Organic Pollutants, Environ. Sci. Technol., 36(19), 4142–4151, doi:10.1021/es020635t, 2002.

Comment 2: There are several places in the paper (e.g. line 164) where results are reported to several significant figures. This should be reduced to 2 or at most 3 significant figures.

Response:

Thank you for your concern, 2 significant figures were chosen for our data. Please see line 34-36, line 205-206, line 220, line 240, line 252-253, line 332, and Table 1 (page 19) in revision. However, data from previous studies, e.g. India, Pakistan, and Vietnam were cited from literatures, we just kept their original values in Table 1.

Comment 3: should read ": : :Global Atmospheric Passive Sampling (GAPS) network: : :"

Response:

We read the suggested research paper and modified the lines 96-99, p. 4, in the introduction section.

Reference

Shunthirasingham, C., Oyiliagu, C. E., Cao, X., Gouin, T., Wania, F., Lee, S.-C., Pozo, K., Harner, T. and Muir, D. C. G.: Spatial and temporal pattern of pesticides in the global atmosphere., J. Environ. Monit., 12(9), 1650–1657, doi:10.1039/c0em00134a, 2010.

Comment 4: line 347, Eq. 1 and related text and SI. Shouldn't these rate constant k-values be lower case?

Response: Thank you for the suggestion. We have changed all the 'K' in the equation and related text into lower case 'k' as per the reference used. Please see p. 16, lines 405-410 in the manuscript.

Comment 5: Figure 1 – what is the source (reference?) of the climate classification map?

Response: We now have provided the proper reference to the climate classification map of Nepal. Please see the caption for Figure 1, p. 18, line 461, and p. 24, lines 544-545.

The cited reference is:

Karki, R., Talchabhadel, R., Aalto, J. and Baidya, S. K.: New climatic classification of Nepal, Theor. Appl. Climatol., 125(3–4), 799–808, doi:10.1007/s00704-015-1549-0, 2016.

Comment 6 : Figure 2 - please double check y-axis label. Also why does it end at 2640? The spacing seems strange 2600 to 2640 takes up almost a third of the figure (y-axis scale) yet 0 to 40 (the same 40 units) takes up much less space. I realize that this is a broken scale but something seems off with the spacing.

Response:

The Y-axes of all the figures in the manuscript are double checked so that the axis label is correctly given. The Figure 2 has been reconstructed based on the recalculated data. The Y-axis has been provided with a break so that a large variation in concentrations of analytes can be understood clearly. The highest concentrations of the dominant chemicals are presented in the figure. Please see p. 19, Figure 2.

Comment 7: Table 1 – use consistent number of significant figures throughout i.e. either 2 or 3 significant figures.

Response: Regarding data of the current study, the quantitative values were kept to 2 digital numbers (Please see p. 20, Table 1). While, data from India, Pakistan and Vietnam were obtained from other references, we kept their original values in Table 1.