We are grateful to the editor's thoughtful and illuminating comments and have now amended the manuscript according to the points. A detailed response to each of the reviewer's points is provided below and we have carefully revised the manuscript (all revisions are highlighted in the text). And we also improved the manuscript with the help of Dr. Dave Chandler (www.GeoEditing.co.uk).

Editor comments:

In addition to the comments made by referees, please address the following comments and suggestions:

1. The review of prior studies of PAH in the Himalayas on page 4 line 15-16 is very terse and has overlooked several relevant publications that provide insight to PAH sources and seasonal variation in the Himalaya:

Chen, P. F., S. C. Kang, C. L. Li, M. Rupakheti, F. P. Yan, Q. L. Li, Z. M. Ji, Q. G. Zhang, W. Luo and M. Sillanpaa, 2015. Characteristics and sources of polycyclic aromatic hydrocarbons in atmospheric aerosols in the Kathmandu Valley, Nepal. Science of the Total Environment 538, 86-92.

Kim, B. M., J. S. Park, S. W. Kim, H. Kim, H. Jeon, C. Cho, J. H. Kim, S. Hong, M. Rupakheti, A. K. Panday, R. J. Park, J. Hong and S. C. Yoon, 2015. Source apportionment of PM10 mass and particulate carbon in the Kathmandu Valley, Nepal. Atmospheric Environment 123, 190-199.

Stone, E. A., J. J. Schauer, B. B. Pradhan, P. M. Dangol, G. Habib, C. Venkataraman and V. Ramanathan, 2010. Characterization of emissions from South Asian biofuels and application to source apportionment of carbonaceous aerosol in the Himalayas. Journal of Geophysical Research Atmospheres 115.

Answer: Have discussed these references in detail. "Some studies of elements and carbonaceous particle concentrations have already been carried out in this region. For example, Bonasoni et al. (2010) provided a detailed description of the atmospheric conditions in the high Himalayas, revealing that brown cloud hot spots mainly influence the South Himalayas during the pre-monsoon season when BC and PM_1 values are higher in comparison to other seasons; Kim et al. (2015) identified and quantified

the contributions of different sources (including brick kilns, motor vehicles, fugitive soil dust and biomass/refuse burning) to particulate carbon in the Kathmandu Valley, using the Solver for Mixture Problem receptor model; Stone et al.(2010) characterized the emission profiles of various biofuels from South Asia and applied them to Godavari, located on the southern edge of the Kathmandu Valley, to improve source apportionment of carbonaceous aerosol in South Asia."

2. In light of the abovementioned source apportionment studies – there is evidence for multiple sources of PAH (e.g. coal, biomass, and fossil fuel use) in the Himalayas. The limitations of using PAH isomer ratios for source identification in the presence of multiple sources should be discussed.

Answer: Have discussed the limitation of using PAH isomer ratios for source identification in section 3.4. "It should be noted that while ratios can be somewhat helpful in distinguishing petrogenic from combustion-derived sources, the diversity of fuels and combustion conditions is likely to produce variations in ratios from a single source, hindering the identification of biomass versus fossil fuel combustion inputs. Additionally, PAHs can be transformed by atmospehric processes so that diagnostic ratios measured in atmospehric samples can differ greatly from those reported for the original sources. As a result, source diagnostic ratios should be used with care and in the context of the studied area."

3. Isomer ratios of PAH have also been utilized as a measure of atmospheric aging, particular photochemical degradation (see Bi et al. 2003). Use of the appropriate isomer ratios to track aging may be useful in establishing quantitative support to evaluate local versus long range transport of PAH.

Bi, X. H., G. Y. Sheng, P. Peng, Y. J. Chen, Z. Q. Zhang and J. M. Fu, 2003. Distribution of particulate- and vapor-phase n-alkanes and polycyclic aromatic hydrocarbons in urban atmosphere of Guangzhou, China. Atmospheric Environment 37 (2), 289-298.

Answer: This paper used BeP/BeP+BaP ratio to evaluate local *versus* long range transport of PAHs. Unfortunately, we didn't detect BeP in this study thus could not use this ratio for the particle aging.

4. I concur with referees concerns with the validity of dry deposition flux estimation. A "rough estimation" using an assumed, untested, and unexplained deposition velocity is not valid. The only conclusion drawn is that flux trends follow concentration, which is obvious from Equation 1 when assuming a fixed deposition velocity. The resulting data are not used "assess the atmospheric environment and its impacts on the Himalayan ecosystem" as noted on page 14 lines 14-15. Consequently, section 3.4 should be removed from the manuscript.

Answer: We agree with the suggestion. In the revision, we deleted this section.

5. In the abstract, a number of improvements are needed: a) clarify the importance of studying PAH in the Himalayas ("understanding: : : remains limited" is too vague);

Answer: Have added this sentence in the abstract "The Himalayas is in the risks of long-range transported atmospheric pollutants (e.g. polycyclic aromatic hydrocarbons (PAHs)). However, knowledge of PAH concentrations, sources and transport pathways remains limited in this region."

b) the names of the sites (with their altitudes) should be listed following "Himalayas:" at line 5;

Answer: The name and their altitudes have been listed (Lumbini: 100 m a.s.l; Pokhara: 813 m a.s.l; Jomsom: 3048 m a.s.l; Zhongba: 4704 m a.s.l; Dhunche: 2051 m a.s.l; Nyalam: 4166 m a.s.l).

c) define x and y in the equations at lines 12-13.

Answer: From our manuscript, both PAH and TSP concentrations have a decreasing pattern with increasing elevation. However, The low-elevation sites had higher PAH concentrations, as they are

more strongly affected by the local pollutant sources. While at the high-elevation sites (e.g. Jomsom and Zhongba), dramatic decrease in TSP and PAH concentrations may be due to pollutants depletion during the long-range transport. We assume this relation also exist in other particle related proxies. But we are not sure it due to lack of measured data. Thus we deleted this sentence from the abstract.

6. In the introduction (page 3 line 15) clarify what "atmospheric mechanisms" specifically need to be understood and why.

Answer: The atmospheric mechanisms mean PAH transport and deposit processes. However, the dry deposition fluxes estimation is not valid according to the suggestion. Thus, we removed 3.4 section and this sentence.

7. The motivation to study PAH should be justified and clarified in the introduction. PAH generally have low acute toxicity to humans, and their most significant endpoint is cancer.

Answer: Have clarified the motivation to study PAH in the introduction. "PAHs have received considerable attention owing to their persistence and toxicity, especially their carcinogenic and/or mutagenic properties (Bhargava et al., 2004). The World Health Organisation (WHO) recommends guidelines in terms of a carcinogenic slope factor, and the European Union indicative limit value is set at 1 ng/m^3 of benzo[a]pyrene (WHO, 2006; European Union, 2005). The United Kingdom has set an air quality standard of 0.25 ng/m^3 benzo[a]pyrene (EPAQS, 1999)".

8. The "sum of PAH" noted on page 3 line 22 is not operationally defined by the method of analysis; indicate the number of PAH and number of rings considered in this summation to provide context for these numbers.

Answer: Have changed "the sum of PAHs" to "16 PAHs (gas + particulate phase)".

9. Likewise, the phrase “total PAH” must not be used in describing the measurements from this study, as not all PAH isomers were quantified. Instead “measured PAH” should be used throughout, e.g. in the caption for Table SI-1.

Answer: Have changed to “measured PAH” throughout this manuscript and the table captions.

10. The following clarifications to the methods are needed:

a) why is hexamethylbenzene used as an internal standard? A number of PAH internal standards are reported in the SI; what is the relationship to this compound?

Answer: We made a mistake here. In our study, 20 ng of anthracene-D10 and benzo(ghi)perylene were added to the eluent solvent as internal standards. We have changed this information in the manuscript on page 18 lines 21-22.

b) Do ambient measurements correspond to local / ambient temperature and pressure, or standard conditions?;

Answer: Yes. The air volume was converted to standard conditions using atmospheric pressure and ambient temperature monitored at each site. We have described this on page 7 lines 16-18.

c) GC film thickness is needed on page 7 line 12;

Answer: The film thickness is 0.25 μm. Have added this information.

d) number of field and laboratory blanks analyzed;

Answer: Have added this information in the manuscript: “Tweleve laboratory and 24 field blanks were extracted and analyzed in the same way as the samples”.

e) detection of analytes (other than naphthalene) in field blanks;

Answer: Have clarified the detection of analytes in field blanks: "In the field blank samples, the average Aecl, Ace, Phe, and Ant concentrations were detected to be 0.59, 0.45, 0.68, and 0.36 pg/m^3, respectively. Other species were below the method detection limit."

f) treatment of field blanks, e.g. field blank subtraction;

Answer: As mentioned above, Tweleve laboratory and 24 field blanks were extracted and analyzed in the same way as the samples. The PAH concentrations were blank corrected but not corrected for the recoveries.

g) number of spike samples;

Answer: Each site, we chose 2 filters as spike samples.

h) preparation of spike samples;

Answer: Each filter was cut to two pieces ($Sample_1$ and $Sample_2$). $Sample_1$ was pretreated in the same way as we described in the manuscript. Add a known quantity (close to the PAH concentrations measured in $Sample_1$) of mixture solution of 16 measuring PAHs to $Sample_2$, then treated that in the same way as $sample_1$. Finally calculate the recovery by using the PAH concentrations of $Sample_1$, $Sample_2$ and added concentration. The recoveries were in the range of 61%-95%. Thus we considered that the pretreat method we used is applicable. We have added this information in the text (Page 8 lines 18-23).

i) a section describing statistical analysis software and methods (e.g. ANOVA).

Answer: Have added the section according to the suggestion. “All of the statistical analysis was conducted using SPSS software (Statistical Package for the Social Sciences, Ver.16.0) and included a regression analysis. Comparisons and correlations were considered statistically significant when $p<0.05$. The data are summarized as the mean ± standard deviation”.

11. The statement about “similar altitudinal distributions” on page 11, line 4 needs clarification. Does this refer to similar logarithmic distributions in prior studies? References are needed.

Answer: Have clarified this sentence and added the reference. “Previous studies have reported a clear decreasing pattern with increasing elevation of persistent organic pollutants in remote mountain areas, either in air, soil, or plants (Gong et al., 2014)”. The reference just showed a decreasing trend of pollutants.

Gong, P., Wang, X., Li, S., Yu, W., Li, J., Kattel, D., Wang, W., Devkota, L., Yao, T., Joswiak, D.: Atmospheric transport and accumulation of organochlorine compounds on the southern slopes of the Himalayas, Nepal, Environ. Pollut., 192, 44-51, doi: 10.1016/j.envpol.2014.05.015, 2014.

12. A value and corresponding reference is needed on page 16 line 1 for the value for wheat burning.

Answer: Have added the value (0.43) and reference (Rajput et al., 2011).

Rajput, P., Sarin, M., Rengarajan, R., Singh, D.: Atmospheric polycyclic aromatic hydrocarbons (PAHs) from post-harvest biomass burning emissions in the Indo-Gangetic Plain: Isomer ratios and temporal trends, Atmos. Environ., 45, 6732-6740, doi: 10.1016/j.atmosenv.2011.08.018, 2011.

13. In SI-1, revise to read “70 eV” and “-20 C until injection.”

Answer: Have revised.