

We are grateful to the reviewer's thoughtful and illuminating comments and have now amended the manuscript according to their points. We have acknowledged the valuable contribution made by the reviewers in this manuscript. 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).

5 Reviewers comments 2:

This manuscript fail to provide any new information or concept on the sources, occurrence and atmospheric transport of PAHs. This is largely due to very poor quality of discussion throughout the manuscript. There are several hand-waving and qualitative statements in addition to poor concepts. Overall, the manuscript is extremely poor in English (with efforts of 11 authors), making it difficult at several places to understand what is being conveyed. Another irrelevant section in the manuscript pertains to estimation of dry-deposition fluxes of PAHs. It is not clear why it is relevant and important to estimate PAHs fluxes at the six sampling sites. This manuscript cannot be recommended for publication in ACP even after major revision. Authors have presented data for only two parameters, TSP and PAHs, which is not adequate to merit publication in in ACP. I would not like to go through the manuscript again.

15 General comments:

1) Overall, the manuscript is poor in English, making it difficult at several places to understand what is being conveyed.

20 Answer: The manuscript has been edited by one professional editor (Dr. Dave Chandler; www.GeoEditing.co.uk) who is native English speaker. All changes according to reviewers comments are marked in blue in the text. And some sections also have rewritten to make them logical and clear.

Section on estimation of dry-deposition fluxes of PAHs is irrelevant. It is not clear why it is important to estimate PAHs fluxes at the six sampling sites. The impact on aquatic systems, if any, or human health

has not been addressed at all. Simply reporting the fluxes (rough estimates – as claimed by authors themselves on page 14, lines 14-15) do not make any sense. With large uncertainty in the deposition velocity of any atmospheric constituent, it is conceptually wrong to state the deposition fluxes of PAHs up to 1st or 2nd decimal units (25.9 ug/m²/d at Pokhara, at Nyalam as 6.74 ug/m²/d and so on – Section 3.4, Pages 13-14). The entire concept of using deposition velocity and deposition fluxes is not valid at all for sampling sites located in high altitude regions. The concept may still be applicable for deposition over high altitude lakes.

Answer: Have deleted this section according to the suggestions of the reviewer and editor.

2) Abstract is still very poorly written: Page 2, lines 1-2: 1) Why it is important to understand transport of PAHs across Himalayas? 2) Why only from the Indo-Gangetic Plain?

Answer: As mentioned above, the manuscript has been improved carefully by one professional language editor. PAHs have received considerable attention for their persistence and toxicity, related to carcinogenic and/or mutagenic properties (Bhargava et al., 2004). The Tibetan Plateau is in the immediate vicinity to South Asia, which is one of the most important source region for particle pollutants in the world (Bond et al., 2007; Zhang and Tao, 2009). Pollutants emitted from South Asia can gather in the foothills of the Himalayas and can be lifted to high altitudes through some big valleys (Qiu, 2013) or even travel over the high elevated Himalayas and invade into Tibetan Plateau which are critical to both the South Asian summer monsoon and hydro-glaciological resource variability in the Himalayas (Bonasoni, 2008; Lüthi et al., 2015; Xia et al., 2011). However, the exact knowledge on source regions, transport pathways and time trends of PAHs to the Himalayas remains uncertain due to lack of observed data. Thus, it is important to understand the transport of PAHs across Himalayas. For the studied region, the dominant surface wind direction is from the south during the monsoon season. While in other seasons, northwesterly winds prevail due to the effect of the westerly winds. Most of

these air masses pass through the Indo-Gangetic Plain (IGP). Thus, the transport of pollutants from the IGP is considered to be the most important pathway controlling the levels of PAHs over the Himalayas (Gong et al., 2014; Bonasoni et al., 2010). Thus we focus on in situ PAHs observation across the Himalayas transported from the IGP. We have added the significance of studying PAHs transported from IGP across the Himalayas in the abstract section. “ 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.”

References:

- 10 Bhargava, A., Khanna, R., Bhargava, S., Kumar, S.: Exposure risk to carcinogenic PAHs in indoor-air during biomass combustion whilst cooking in rural India, *Atmos. Environ.*, 38, 4761-4767, doi: 10.1016/j.atmosenv.2004.05.012, 2004.
- Bonasoni, P.L., P; Angelini, F ; Arduini, J ; Bonafe, U ; Calzolari, F ; Cristofanelli, P; Decesari, S ; Facchini, MC ; Fuzzi, S 2008. The ABC-Pyramid Atmospheric Research Observatory in Himalaya for aerosol, ozone and halocarbon measurements. *Science of the total environment* 391, 252-261.
- 15 Bond, T., Bhardwaj, E., Dong, R., R, J., 2007. Historical emissions of black and organic carbon aerosol from energy - related combustion, 1850–2000. *Global Biogeochemical Cycles* 21, GB2018.
- Gong, P., Wang, X., Yao, T.: Ambient distribution of particulate-and gas-phase n-alkanes and polycyclic aromatic hydrocarbons in the Tibetan Plateau, *Environ. Earth. Sci.*, 64, 1703-1711, doi: 10.1007/s12665-011-0974-3, 2011.
- 20 Lüthi, Z.L., Skerlak, B., Kim, S.W., Lauer, A., Mues, A., Rupakheti, M., Kang, S.C., 2015. Atmospheric brown clouds reach the Tibetan Plateau by crossing the Himalayas. *Atmos. Chem. Phys* 15, 6007-6021.
- Qiu, J., 2013. Pollutants Capture the High Ground in the Himalayas. *Science* 340, 1042-1042.
- Xia, X., Zong, X., Cong, Z., Chen, H., Kang, S., Wang, P., 2011. Baseline continental aerosol over the

central Tibetan plateau and a case study of aerosol transport from South Asia. *Atmospheric Environment* 45, 7370-7378.

Zhang, Y., Tao, S.: Global atmospheric emission inventory of polycyclic aromatic hydrocarbons (PAHs) for 2004, *Atmos. Environ.*, 43, 812-819, doi: 10.1016/j.atmosenv.2008.10.050, 2009.

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3) Page 2, lines 20-24: Isomer ratios are expected to help in identifying the specific source signature, whereas, authors have stated that – quote “isomer ratios suggested that atmospheric PAHs from the Nepal sites were mainly associated with emission of biomass, coal burning and petroleum burning”. This is a very qualitative statement.

10 Answer: Although with uncertainty, isomer ratios have been widely used to detect combustion-derived PAHs (Tobiszewski and Namiesnik, 2012; Yunker et al., 2002). They can help to identify the specific sources. However, it cannot quantify the contributions from different sources. Therefore, we changed this sentence to “Both IndP/(IndP+BghiP) and Fla/(Fla+Pyr) ratios suggested that atmospheric PAHs from urban and rural sites were mainly associated with emission from biomass burning, coal burning and petroleum combustion. However, the contribution of biomass burning increased at remote sites.” to
15 make it clear.

Reference:

Tobiszewski, M., Namiesnik, J.: PAH diagnostic ratios for the identification of pollution emission sources, *Environ. Pollut.*, 162, 110-119, doi:10.1016/j.envpol.2011.10.025, 2012.

20 Yunker, M., Macdonald, R., Vingarzan, R., Mitchell, R., Goyette, D., Sylvestre, S.: PAHs in the Fraser River basin: a critical appraisal of PAH ratios as indicators of PAH source and composition, *Org. Geochem.*, 33, 489-515, doi: 10.1016/S0146-6380(02)00002-5, 2002.

4) Conclusion: Page 18, lines 10-15: If inferences are drawn only from AMBTs, then it is not of much

relevance to measure chemical constituents (example PAHs). Their long-range transport cannot be considered “conservative”.

Answer: In our study, we measured PAH concentrations and compositions at six sites. The results showed that three remote sites could be affected by pollutant emitted by the IGP. We got the results from the change of PAH concentration and composition along two south-north transects. Not only from air mass backward trajectory (AMBTs). But the Hysplit model was used to support our conclusion here. Atmospheric residence time with respect to airborne transport is related to the behavior of the carrier particles. Most particulate phase PAHs are adsorbed onto particles in the accumulation mode (0.2-2 μm) (Miguel and Friedlander, 1978). These particles only deposit slowly from the atmosphere and, depending on atmospheric conditions, may be airborne for days or even weeks, being transported over long distances (in excess of 1000 km) (Harrison et al., 1996). Thus, PAHs can undergo long-range atmospheric transport and have been identified in the world’s most remote parts (Ding et al., 2007; Masplet et al., 1988).

References:

Ding, X., Wang, X.M., Xie, Z.Q., Xiang, C.H., Mai, B.X., Sun, L.G., Zheng, M., Sheng, G.Y., Fu, J.M., Poschl, U.: Atmospheric polycyclic aromatic hydrocarbons observed over the North Pacific Ocean and the Arctic area: spatial distribution and source identification. *Atmos. Environ.* 2007, 41, 2061-2072.

Harrison, R.M., Smith, D.J.T., Luhana, L.: Source apportionment of atmospheric polycyclic aromatic hydrocarbons collected from an urban location in Birmingham, UK. *Environ. Sci & Technol.* 30, 825-832.

Masplet, P., Pistikopoulos, P., Beyne, S., Mouvier, G.: Long-range transport and gas particle distribution of polycyclic aromatic hydrocarbons at a remote site in the Mediterranean-Sea. *Atmos. Environ.* 1988, 22, 639-650.

Miguel, A.H., Friedlander, S.K.: Distribution of benzo(a)pyrene and coronene with respect to particle

size in Pasadena aerosols in the sub-micron range. *Atmos. Environ.* 1978, 12, 2407-2413.

5) Page 18, line 8: What authors mean by “higher deposition efficiency”? How this is built in the concept of using deposition velocity for PAHs? How deposition efficiency is assessed from the data presented in the manuscript.

Answer: Concentration of high molecular weight PAH decreased from the south to north along two transects. Besides the emission source effect, the deposition efficiency is another reason. PAHs (3+4 ring) and PAH (5+6 ring) behave differently due to their chemical characteristics and nonvolatile PAHs are more efficiently removed from the atmosphere than semi-volatile PAHs. Thus we considered that high molecular weight PAHs have higher deposit velocity than light molecular weight PAH (Schauer et al., 2003). However, we cannot get this from the deposition velocity. Thus we changed the description of this sentence to “while the increase of 3-ring PAHs at the other three remote sites might reflect the gas/particle phase transition or higher deposition efficiency of high molecular weight PAHs during long-range transport.”

Reference:

Schauer, C., Niessner, R., Poschl, U.: Polycyclic aromatic hydrocarbons in urban air particulate matter: decadal and seasonal trends, chemical degradation, and sampling artifacts. *Environ. Sci. Technol.*, 37, 2861-2868. DOI: 10.1021/es034059s

6) Specific Comments:

a) Abstract, Page 2, line 11: What is the concept of logarithmic decreasing pattern of PAHs with increasing elevation? Is this is an empirical relation only applicable to PAHs?

Answer: The low-elevation sites had higher PAH concentrations, as they are more strongly affected by the local pollutant sources. While the long-range transport of pollutants predominates at the high-

elevation sites, dry and wet deposition during the long-range transport is likely to be the main reason for the decrease in TSP and PAH concentrations. From our manuscript, both PAH and TSP concentrations have a decreasing pattern with increasing elevation. We assume this relation also exist in other particle related proxies. However, we are not sure it due to lack of measured data. We have deleted this sentence from the abstract.

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b) Page 3, lines 14-15: Why study of PAHs in remote sites is needed for the understanding of the atmospheric mechanisms involved in the long-range transport of these pollutants? Which “atmospheric mechanisms” authors are referring to during long-range transport?

10 Answer: PAHs are persistent in the atmosphere thus can be transported over long-distance to even remote areas such as the Himalayas. 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.

15 c) Page 4, line 8: ABC is not “Asian Brown Cloud”. It refers to “Atmospheric Brown Cloud”.
Answer: Have changed.

d) Page 5, lines 5-19: Why these sources are not important for the contribution of PAHs measured at the six sampling sites?

20 Answer: Actually, these sources are very important for the contribution of PAHs at the six sampling sites. We listed the nearby emission sources around the sampling sites here and in section 3.1, we discussed the influence of these sources on the spatial and seasonal distribution of measured PAHs.

e) Page 9, lines 4-8: Based on TSP and PAHs concentrations, it is conceptually incorrect to conclude

impact and transport of pollutants from the IGP in the winter. What about contribution from intermediate/downwind sources.

Answer: Here, we compared our results (Jomsom, TSP: $96.2 \pm 40.8 \mu\text{g}/\text{m}^3$; PAHs: $11.1 \pm 2.97 \text{ ng}/\text{m}^3$) with Barapani (PAHs: $14.1 \text{ ng}/\text{m}^3$) which measured OC, EC, WSOC, and PAHs of $\text{PM}_{2.5}$ in Barapani and concluded that the study region was influenced by the long-range transport of aerosols from the IGP in winter (Rajput et al., 2013). Considering that Barapani is located in the foothill of the Himalayas, which has similar geographical conditions with Jomsom, we inferred that Jomsom might also be affected by emission from the IGP. The intermediate regions can also affect the studied region. However, emissions from these regions are less compared to those of the IGP, thus, we considered that the study is mainly affected by the IGP.

Reference:

Rajput, P., Sarin M., Kundu, S.S.: Atmospheric particulate matter ($\text{PM}_{2.5}$), EC, OC, WSOC and PAHs from NE-Himalaya: abundances and chemical characteristics, *Atmos. Pollut. Res.*, 4, 214-221, doi: 10.5094/APR.2013.022, 2013.

f) Page 9, lines 3-5: Std. deviation on PAHs cannot be stated as $5.65 \text{ ng}/\text{m}^3$, $2.97 \text{ ng}/\text{m}^3$ and so on. Are these significant up to 2nd decimal units.

Answer: We have changed these significant to 1 decimal unit.

g) Page 10, lines 10-14: It is not clear what authors are trying to infer and convey. It is rather poor discussion on spatial and temporal variability of TSP and PAHs along south-north transects.

Answer: The seasonal variation of PAH and TSP concentrations are clear at urban and rural sites while not apparent at remote sites. However, the seasonal averaged concentrations of TSP and PAH in Zhongba and Nyalam (located in the Tibet-Himalayas) showed similar seasonal variation with those in

Nepal (Table SI-1). Have rewritten this sentence to “Despite decreasing markedly from south to north along these two transects, the TSP and PAH concentrations in Zhongba and Nyalam exhibited similar seasonal variations with those in Nepal, suggesting that the northern side of the Himalayas might have similar sources for atmospheric PAHs”.

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h) Page 11, lines 7-8: Concentrations of PAHs in soils and variability with altitude is out of context and irrelevant. PAHs in soils cannot be assumed to be derived from atmospheric deposition.

Answer: Have deleted this sentence.

10 i) Page 11, lines 9-10: Which “nearby contaminant sources” authors are referring to?

Answer: Have changed “nearby pollutant sources” to “local pollutant sources” and clarified that in the section. The local pollutant sources: for example the burning of large amounts of crop straw and wood at Lumbini; eleven cement factories and more than fifty other industries along the nearby Lumbini–Bhairahawa industrial corridor; increasing numbers of vehicles and hotels at Pokhara..

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j) Page 11, line 12: “Thus, less local anthropogenic emissions ——. What is “less”?”

Answer: Those remote sites have few anthropogenic emission sources compared to urban and rural sites. Have changed this sentence to “Local emission of PAHs was very limited and the long-range transport of pollutants from lowland source regions might predominate 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.”

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k) Page 11, lines 12-15: The entire discussion is very qualitative and poorly written.

Answer: As mentioned above, we have rewritten this sentence to “Local emission of PAHs was very

limited and the long-range transport of pollutants from lowland source regions might predominate 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.”

- 5 l) Page 11, line 15: “—thus we just gave a rough estimate of the regression analysis in this study”. What is the relevance of giving “rough estimate”?

Answer: It is not precise and we have deleted this sentence.

- 10 m) Page 12, line 8:“—— indicating that biomass combustion is the main source for particulate PAHs in Lumbini”. How biomass combustion source can be inferred from particulate PAHs? What are the concentrations and ratio of OC and EC?

15 Answer: Lumbini is a typical rural site based on rice and wheat planting. Biomass such as agricultural wastes, animal dung, and wood, are burned almost throughout the year mainly for cooking activities. Burning of large amounts of agro-residue also occurs in the IGP, especially in the pre- and post-
20 monsoon seasons (Ram and Sarin, 2010). While some brick kilns are mainly operated from January to April (Chen et al., 2015). In addition, the profile of PAHs we observed in Lumbini was similar to that of residue combustion aerosols collected at North India (Rajput et al., 2011). Thus we conclude that biomass burning is the main source of PAHs in atmosphere of Lumbini. The average concentrations of OC and EC at Lumbini were found to be $32.06 \pm 23.77 \mu\text{g}/\text{m}^3$ and $6.44 \pm 3.64 \mu\text{g}/\text{m}^3$, respectively. And the average OC/EC ratios in Lumbini were 4.66, 4.31, 5.24, and 6.47 for the pre-monsoon, monsoon, post-monsoon, and winter seasons, respectively (annual average: 4.82 ± 2.27).

Reference:

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.

Ram, K., Sarin, M.: Spatio-temporal variability in atmospheric abundances of EC, OC and WSOC over Northern India, J. Aerosol. Sci., 41, 88-98, doi: 10.1016/j.jaerosci.2009.11.004, 2010.

5 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.

n) Page 15, lines 16-19: There is no new understanding emerging from this qualitative discussion.

Answer: As mentioned in point 3, isomer ratios cannot quantify the contributions from different sources.

10 However, they have been proved helpful to detect combustion-derived PAHs. From the data, we can found that these ratios are different from those in Lumbini and Jomsom, reflecting influence of local emissions at Lumbini. However, the urbanization development of Pokhara is far less than Kathmandu; thus, pollutants from local sources can affect Pokhara itself but not its adjacent region such as Jomsom.

15 o) Page 17, lines 1-2: What is “thermally driven flows through Himalayan valleys and up sides?”

Answer: Have changed this sentence “--thermally driven flows through Himalayan valleys.”

p) Page 17, lines 3-4: “— diurnal valley wind system often occurs that blows up valley—“. What authors mean by “that blows up valley— “?”

20 Answer: Have changed this sentence to “A diurnal valley wind system often occurs in mountainous terrain, with an up-valley wind during the day reversing to a down-valley wind during the night.”

7) There are several confusing & qualitative statements:

Page 2, lines 22-24;

Answer: Have changed this sentence to “Similar compositions were found at three remote sites located on both sides of the Himalayas (Jomsom, Zhongba, and Nyalam), suggesting that the northern side of the Himalayas may be affected by anthropogenic emissions from the IGP via long-range atmospheric transport.”

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Page 3, lines 14-15;

Answer: As mentioned in specific comments of (b), we deleted this sentence.

Page 9, lines 7-10;

10 Answer: Have changed this sentence to “Considering that local emissions in Jomsom are very low, the measured pollutants probably accumulate by the up-valley winds that transport pollutants from the IGP.”

Page 10, lines 10-15;

15 Answer: Have changed this sentence to “Despite decreasing markedly from south to north along these two transects, the TSP and PAH concentrations in Zhongba and Nyalam exhibited similar seasonal variations to those in Nepal, suggesting that the northern side of the Himalayas might have similar sources for atmospheric PAHs.”

20 Page 11, lines 6-7; 10-15;

Answer: Have changed this sentence to “For example, Gong et al. (2014) observed that the concentrations of persistent organic pollutants (POPs) such as hexachlorobenzene (HCB) and polychlorinated biphenyls (PCBs) in the atmosphere decreased by a factor of approximately 3 times from 135 m to 5100 m.” and “Local emission of PAHs was very limited and the long-range transport of

pollutants from lowland source regions might predominate 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”, respectively.

5 Page 13, lines 3-4;

Answer: Have changed this sentence to “In contrast, the PAH profiles in Zhongba and Nyalam were similar to that in Jomsom (Fig. 4), which might also reflecting the long-range transport of pollutants from south to north across the Himalayas”.

10 Page 14, lines 12-15;

Answer: As mentioned in the point 1, we deleted this section according to the reviewer’s and editor’s suggestion.

Page 15, lines 16-19

15 Answer: Have changed the sentence to “a significant level of rural biomass burning occurs around the city. Thus, the PAHs in Pokhara were derived from the combined impacts of local contributions from Pokhara city (both biomass and fossil burning) and polluted air mass transport from upwind IGP which often arrived during the winter and pre-monsoon seasons.”

20 8) English errors:

Abstract, Page 2, line 11: “exhibited”; wrong English;

Answer: Have changed “exhibited” to “showed”.

Page 3, line 1: “long-range transportation”

Answer: Have changed “long-range transportation” to “long-range atmospheric transport”.

Page 6, line 17: “pre-burned”; What is pre-burned? It should be “pre-combusted”!

Answer: Have changed “pre-burned” to “pre-combusted”.

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Page 8, line 2: “All analytic”; What is analytic?

Answer: Have changed “analytic” to “analytical”.

Page 10, lines 14 and 20: “long-range transported pollution”? Wrong English “transported”!

10 Answer: Have changed “long-range transported pollution” to “long-range transport pollution”.

Page 10, line 21: “— concentrations “exhibited”—”.

Answer: Have changed “exhibited” to “showed”.

15 Page 11, line 9: “The low elevation sites displayed—“; displayed is not a correct word to use!

Answer: Have changed “displayed” to “showed”.

Page 12, line 15: “PAHs are present”; not is present.

Answer: Have changed “PAHs is present” to “PAHs are present”.

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Page 12, line 16: “— PAHs scarcely reenter the atmosphere—“ . Very poor English!

Answer: Have changed this sentence to “PAHs were rarely returned to the atmosphere”.

Page 14, line 21: “— indicates non-burned petroleum—“ . Non-burned is incorrect word.

Answer: Have changed “non-burned” to “non-combusted”.