Response to referee 1:
Author's response in Blue

The authors presented measurement results of 17 polycyclic aromatic hydrocarbons (PAHs) in Beijing, China, and Delhi, India in summer. The sampling was conducted with higher time resolutions (~3 hours for daytime samples and ~15 hours for nighttime samples) as compared to traditional 24-h samples. The PAHs were quantified with GC-Q- ToF-MS. Results showed that PAH concentrations were higher in Delhi than those in Beijing, and the summer PAH concentrations were lower than those in winter in Beijing. From the measured PAH profiles, sources of PM-bound PAHs in these two mega-cities in developing countries were inferred. In addition, health risks were calculated from the measured PAH concentrations. The study is well designed and the analysis is rigorous. The manuscript is fairly well written. I recommend Minor Revision before publication, with a few comments as follows.
We thank the reviewer for commenting this paper. Your corrections and recommendations, helped us to better present the data and we believe the paper has been improved.

Major
1. It was stated in the abstract (and in the conclusion) that "in Delhi 25% of the emissions were attributed to long-range atmospheric transport". The only evidence the authors used to support this is on L425-427/P9, showing that 25% of data in Delhi had a Bap/(BaP + BeP) ratio of less than 0.5. This evidence is a little bit thin to support such a statement. I suggest the authors to either elaborate this with more evidence, or tone down such an unsupported statement.
The statement of 25 % was removed from the abstract and conclusions, however, the data from BaP/(BaP + BeP) ratio are still useful to support future studies investigating on local and regional emissions in Delhi. Therefore, we prefer to keep this statement in the text, we reported the results as a possible contribution from regional pollution at the sampling site in Delhi. A new discussion was added to the text and detailed in the next question.

Changes to the text:
L.577-580 removed and replaced by: This ratio suggests a larger contribution from local sources in both cities.

2. The issue of oxidation during sampling to the interpretation of results. First, in the paragraph of L192/P5, the authors noted that this effect could be an additional source of uncertainty (10 – 30%) to conventional analytical uncertainties (25 – 30%). The question is, what is the overall uncertainty if both of these two errors are taken into account? Second, after acknowledging this source of potential negative artifact, the authors used it in Section 3.3 to infer particle aging and then to regional transport of PM. Such inference may be conflicting without quantitative assessment on how such "on-filter" oxidation affect the indicator, i.e., the BaP/(BaP + BeP) ratio. Please clarify.
To determine the bias on the results we have used the ''top-down'' approach where the bias determination can be based on recovery efficiency to correct PAHs concentrations. The analytical uncertainty is due to available information on laboratory test performance. The "on-filter" oxidation is a type of chemical degradation/transformation. Therefore, the values of PAHs concentration has to be considered only as a lower limit due to "on-filter" oxidation. It is, however, different from the analytical uncertainty which estimates the lower and upper limits of the results.
A quantitative assessment of "on-filter" oxidation was not a part of this study, our assessment was based on the concentration of ozone measured at the sampling site and compared to quantitative assessment used in previous studies, as reported in section 2.4 for the error

evaluation. BaP and BeP were among the major compounds quantified in this study, and as shown in Table 1, their mean concentrations are similar in the margin of the analytical uncertainties, this support the statement suggesting local emissions as major contributors of PAHs in both cities.
According to Tsapakis and Stephanou (2003) the relative reactivity of BaP, with respect to degradation on glass fibre filters, was 1.6 times higher than BeP. Taking into account the estimated error on sampling artefact in each city and assuming that "on-filter" oxidation was affecting BaP and BeP, as suggested by Tsapakis and Stephanou (2003), the ratio of BaP/(BaP + BeP) will be affected negatively by an average of 4 % (day) and 1.6 % (night) for Beijing, 7 % (day) and 3 % (night) for Delhi.
This assumption will therefore be affecting Delhi results suggesting more contribution from long range transport.

Changes to the text:
L.427: However, this assumption does not take into account the ''on-filter'' oxidation errors during sampling. Tsapakis and Stephanou (2003) reported a relative reactivity of BaP of 1.6 times higher than BeP, with respect to degradation on glass fibre filters. Using the reactivity factor of 1.6, the ratio of BaP/(BaP + BeP) will be affected negatively by an average of 4 % (day) and 1.6 % (night) for Beijing, 7 % (day) and 3 % (night) for Delhi. This assumption will therefore be affecting Delhi results suggesting more contribution from long range transport. Therefore, the indicator of particle aging should be used with careful in the summer season unless ozone ambient concentrations are below 30 ppb, and consequently the negative artefacts are considered not significant (Tsapakis and Stephanou 2003).

L.446: Tsapakis and Stephanou (2003) reported a relative reactivity for BaP, Pyrene and Fluorene of 0.86, 0.82, and 0.68 respectively. The relative reactivity of BaP and Pyrene are similar and therefore does not affect the indicator Pyrene/BaP values. Pyrene is by 20 % more reactive than Fluorene, the ''on-filter'' oxidation has little effect on the indicator Fluorene/Fluorene+Pyrene values, because of the large difference in the defined threshold values which were 6 and 30 for petrol and diesel cars, respectively.

3. Section 3.5. It is not clear why the authors preferred to use only BaP for the cancer risk calculation. Both Table 2 and L524-527/P12 indicated that other PAHs may contribute another half of the risk. Would the reported LECR per million people values be under-estimated if other PAHs are not taken into account?
It is true that other PAHs have high Toxicity Equivalency Factor (TEF) but these TEF are relative to BaP. In Eq. 2 the use of BaP instead of BaPeq was recommended by U.S.EPA, 2002, and Boström et al., 2002, and that's because the unit risk (UR) already include the toxicity values of other PAHs, it is referred to as the surrogate approach.
The use of BaPeq (taken into account other PAHs) instead of BaP will overestimate LECR by 83% for Beijing, and 92 % for Delhi.

Minor
1. L146/P4: suggest to change "17-PAHs" to "17 PAHs", and change in a number places (e.g., L223/P5) "24 h mean concentration" to "24-h mean concentration".
"17-PAHs" corrected to "17 PAHs" and 24 h corrected to 24-h in the entire manuscript.

2. L156/P4: please change “PAHs concentrations” to “PAH concentrations”.
“PAHs concentrations” corrected to “PAH concentrations” in the entire manuscript.

3. L187/P4: why higher error could be attributed to samples analysed previously in wintertime? Memory effect? If so, why were the Delhi samples not affected? Were the Delhi samples analysed after Beijing summer samples?
In fact, lower %RSD was attributed to samples analysed in wintertime Beijing. The precision of sample replicates in wintertime Beijing and summer Delhi showed better %RSD (<10%) for few compounds as shown in Table S1. However, the maximum %RSD in summer Beijing was 13.7 % which is acceptable. This is a type of random errors where it is difficult to determine the origin of the error. L185 to L187 were removed from the text as this error is not related to lower PAH concentrations in summer Beijing.
Yes, Delhi samples were analysed after Beijing summer samples.

4. L199/P5: Tsapakis and Stephanou 2003: please use proper citation.
Corrected as Tsapakis and Stephanou (2003)

5. L207/P5: please add “that” after “than”.
Corrected

6. L414&L425/P9: please use BaP/(BaP + BeP) consistently.
Corrected

7. L418/P9: please change “[ratio = 0.5]” to “(ratio = 0.5)”.
Corrected

8. L431-434/P10: this seems like two sentences. Please revise.
Corrected as follow: We calculated the ratio value for Pyrene/BaP using the data reported in a previous study (Rogge et al., 1993), where the authors quantified more than 100 organic compounds in exhaust emissions fine particulate matter. The ratio value for Pyrene/BaP was ~ 0.7 for noncatalyst-equipped petrol cars, ~1.3 for catalyst-equipped petrol cars and >16 for heavy duty diesel engines.

9. Figure 3: in addition to the non-preferable “17-PAHs” on the graph and in the caption, I do not see the usefulness of putting “17-PAHs” on the graph. Please remove them on the graph and change to “17 PAHs” in the caption.
In Figure 3, ∑17 PAHs was added to the y axis and removed from the x axis. 17 PAHs corrected in the caption and in Figure 1 and 2.

10. Figure 5: please change the title of the y axis to “Bap/(BaP + BeP)”, as well as that in the caption.
Corrected

11. Table 1: please change “PAHs concentrations” to “PAH concentrations”.
Corrected