

The authors present a rich data set examining the concentrations of PAH's in atmospheric particulate matter in Beijing. PAHs are known to increase the toxicity of aerosols and this work advances our understanding of their dynamics in the atmosphere. The analytical methodology applied in this work is impressive and carefully reported. Where this manuscript needs improvement is the discussion and presentation of the data. I recommend the authors work to increase the clarity of the results, and their interpretations, to ensure their work is as impactful as possible.

General Comments:

Comment 1: The authors have a very data rich paper, but often don't report a measure of variability (e.g., standard deviation). This is particularly potent given the title of the manuscript. In the text and in the tables a measure of variability should be included, and discussed, when reporting any data. Specifically, standard deviations should be included in table 2.

Comment 2: The authors present this work as a higher resolution examination of PAH dynamics because they use relatively high resolution (3h) samples. Despite this, there is almost no discussion of temporal dynamics other than day vs. night or 3h vs. 9h vs. 15 h. I would like to see the authors look into the temporal dynamics on given days and investigate drivers of PAH variability beyond correlation.

Comment 3: Was there meteorological data available at your study site? PAH lifetime, emission and phase are likely influenced by temperature or solar flux. Additionally, analysis of wind direction trends during the specific emission periods would provide additional insight into the importance of different sources. If this data exists, including it in the analysis could increase the impact of the results

Comment 4: Organization. This manuscript, although data rich, is poorly organized with many acronyms, lots of in text data, and unclear conclusions. I recommend the authors make an attempt to trim the text. Specifically, many data references in the discussion could be replaced with reference to the appropriate figure or table. This will allow the readers to better understand the author's conclusions by cleaning up the text.

Comment 5: The authors could improve their argument for the specific significance of their work. The authors do a good job of detailing PAHs in aerosol but don't discuss their site in significant detail. Please elaborate on why there is a pressing need to study PAHs in Beijing within your introduction

Comment 6: For a field study, site selection and significance is very important. Please add some discussion of how representative your site is compared to Beijing in general, what the strengths of the site are, and what are some possible weaknesses.

Specific Comments

Line 20: Restructure the list of compounds. As written, it is easy to miss the period after the major PAHs and not realize a new list has started for the major OPAHs. Use the same sentence structure for each type to avoid confusing the reader.

Line 38: comma after “organic carbon”

Line 58: Clarify “geographic peripheral expansion”

Line 60: change “PAH on 24 PM_{2.5} sampling” to “PAH concentrations in 24 h averaged samples”.

Line 62: Remove “sampling and” as written it is redundant.

Line 65 – 67: Your reference to PMF here seems out of place as you do not perform PMF on your results or discuss it as a future method of analysis.

Line 76: Can you really show a sampling campaign? Sampling scheme or setup would be more appropriate.

Line 82: Please clarify your sampling technique here. You say the filter was changed every three hours, but in line 80 you say also performed nine hour sampling during the day.

Line 83: Rephrase the nighttime sampling period time from. A clearer phrasing could be “Night-time sampling began at ~17:30 and ended at 08:30 the following day.”

Line 88: Was a method blank also performed? If so please explicitly state, if not, please explain how sources of contamination were investigated.

Line 91: What is the purity of these standard compounds? Could there be non-deuterated forms of them within the standard which could contaminate the samples?

Line 102: I’m assuming these were calculated from your deuterated standard compounds. Add a statement saying so. Also, it would be useful to have an idea of the variation of the extraction efficiency as well as the range. Add the mean and standard deviation here if possible. Additionally, if samples were adjusted to account for extraction efficiency that should be stated.

Line 138: Does the simultaneous measurement mean each filter extract was spiked with deuterated compounds, the 16 PAH standard mixture, or the standard curve?

Line 148: This is a very odd way to discuss precision. Why don’t you simply state the mean RSD $\pm \sigma$ from all sample replicates of each type? Since your intraday and interday are similar, there may not be any need to state both numbers.

Line 154: Please restructure this. You have not yet introduced systematic error, but this reads as if you’ve already discussed it.

Line 157: Systematic error is not uncertainty so you cannot combine it with your precision estimates. You could propagate the uncertainty associated with the systematic error to better constrain your overall uncertainty.

Line 158: This, combined with the calculation of extraction efficiencies, is a way to constrain systematic errors. I recommend combining this section with lines 154-157 and your discussion of extraction efficiencies in 102.

.Line 164: Add “The number of...has”

Line 166: Rephrase for clarity. “In China, the official PM_{2.5} annual and 24hr average standards are 35 µg m⁻³ and 75 µg m⁻³, respectively,”

Line 168-169: Either this statement is incorrect, or your methods section is incorrect. In your methods section, you state that you’ve collected 3, 9, and 15 hour averaged samples. Please fix or clarify.

Line 169: A more appropriate way of presenting this would be the mean and standard deviation. “During sampling, the average 24h averaged PM_{2.5} was $X \pm Y$...”

Line 171: Same as above, the mean and standard deviation would be very meaningful here.

Line 174: Again, the mean and standard deviation would be very meaningful here.

Line 171 – 178: All of this information would be presented better in a table. Consider adding the summary data in this paragraph to the top of each section in table 2.

Line 179 - 193: Is there any reason why these specific PAHs are important or interesting besides them being found in the highest concentrations? Some discussion of the particularly interesting compounds would improve the quality of this manuscript.

Line 193: Can you mention some specific controls the local government has put in place or cite some of the policies? If they are effective, as you are implying, perhaps they could be applied in other locales, and it would be useful for the reader to know more about them.

Lines 203-213: Is there a specific reason by people report these specific ratios? Do they mean anything other than the relative amount of each type? As written, they seem like a random descriptor that isn’t adding much meaning to your observations.

207: Wouldn’t the average of the ratio within all the samples be more meaningful. Or maybe a total ratio of total measured PAH:NPAH?

214: Does daily mean 24 h averaged? As written the discussion of BaP seems out of place. Are there specific air quality standards for the other compounds studied? If so, please discuss them as well.

225: As written, it’s unclear which ratio is being explained after the semicolon.

254: I believe you are trying to say you used an isomeric standard to estimate the sensitivity to a compound where a standard is not available. Please add some citation explaining why this is

okay with this class of compounds and a statement explaining why you expect the sensitivities to be similar within your analytical setup. Also, if you have measured any other isomer pairs, it might strengthen your case if you show the reader that they have similar sensitivities.

266 – 316: Correlation can be problematic especially when dealing with interlinked species and non-random sampling. Due to your long averaging times for your filters, and (I'm assuming) relatively quick gas phase measurements it may be difficult to make any concrete conclusions.

278: High CO correlation is not necessarily an indicator of regional sources as you would expect correlations to be high near the sources as well.

272-273: Please explain what values/units you used in your correlation analysis. Right now it is unclear if you used the average concentration over the sampling period, the integrated concentration, or something else (i.e., the median).

306-311: Could this be reversed? Could HONO levels be essential to forming secondary 9-Nitroanthracene?

312: "1/3, 1/9" if you are going to use this symbology to refer to the sampling types, be consistent throughout the paper.

340: Again, some description of the variation of the data is needed.

662: This data would be better presented in box and whisker plots.

672: Figure 2 borders on illegible. This may have occurred during formatting, but this figure needs to be fixed before publication.

707: Wouldn't box and whisker plots again be better here as well?

734: The bars on the plot are very difficult to see. One way to make this clearer could be to make the width of the bars proportional to the sampling time.