Interactive comment on “Nitro-polycyclic aromatic hydrocarbons – gas-particle partitioning, mass size distribution, and formation along transport in marine and continental background air” by Gerhard Lammel et al.

Anonymous Referee #2

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The Lammel et al. manuscript reports on measurements, partitioning calculations, and back-trajectory modeling of PAHs and nitro-PAHs at two European sites. The sites were chosen to study the transport of PAHs and nitro-PAHs from urban locations. Samples were collected using high-volume samplers, and particles were collected onto quartz fiber filters and gases using polyurethane foam plugs. Samples were extracted using dichloromethane and extracts were analyzed by gas chromatography-mass spectrometry. Partitioning calculations were performed using a recent poly-parameter linear free energy relationship. Particle trajectories were modeling using HYSPLIT and FLEXPART. There are a lot of high quality data and analyses presented in the manuscript,
and the work should prove to be a valuable contribution to the field. There are no concerns with the methodology employed. The primary concern is with the quality of the representation—the technical writing and organization. There are a lot of grammatical errors and undefined symbols and acronyms. It is hard to follow the results as they are presented, particularly with regard to the discussions and conclusions being presented. I recommend significant attention to the quality and clarity of the presentation, and perhaps another round of reviews following revision.

Several specific suggestions and recommendations are provided below regarding grammar/symbols/abbreviations through the abstract and introduction. The intent here is to provide examples of the types of changes that need to be made throughout the manuscript; all such needed changes are not noted beyond the introduction.

Abstract: Why are the fractional doses not reported for the background site? Or is that being used to characterize the fraction at the marine site? This is not clear in the abstract as written. On line 25, it is suggested to replace “were received” with “were calculated”.

Line 22-23: For easier reading, recommend to move “with 2-nitrofluranthene . . . most abundant” out of the parenthesis and to the end of the sentence, and then start a new sentence with “While the concentration . . .”

Line 31: NPYR is not defined or otherwise discussed

Line 36-37: Seems out of place. Recommend to remove or better integrate with rest of paragraph.

Line 49-50: Add comma after “more than one third”. On line 50, ambient what? Aerosols?

Line 55: Insert “a” between “only” and “few”

Line 60: Suggest to change “indicative for” to “indicative of” or “indicators for”
Line 63: SVOCs is not defined

Line 69: Suggest removing “However . . . studied”. It has already been established (lines 66-67) that most NPAH observations are in urban areas. Could then use something like . . .”Though there are a few studies in unpolluted environments: studies were conducted by . . .”. Also suggest to change all the “were” to “have been” (lines 70-75) as measurements may be ongoing or planned.

Line 66-67: Suggest to change “suspect to” to “expect to”

Line 79-80: Suggest to change “lack of data . . . obvious” to “However, there is limited NPAH data from remote atmospheric environments . . .”

Line 80: Revise “aim of study to study”

Line 112-113: What was the collection period of for the QFF samples at K-puszta?

Line 116: Ni is undefined

The comments/suggestions below address the clarity of the manuscript and analyses/conclusions presented. There are some technical questions here, but I think that more of the questions/comments arise from organizational structure and imprecise wording.

Conceptually, the urban fractional dose is clear; but how exactly were they calculated? I think much more time needs to be spend on the discussion of the calculated doses, sensitivities, and uncertainties to support the first paragraph of the conclusions section.

Line 209-213: The discussion of the two OM phases is confusing as written. Is one phase low molecular mass/water soluble OM and the other phase high molecular mass/organic soluble OM?

Line 229: What is meant by “the substance patterns are similar”? Diurnal patterns? Monomer ratios? Monomer fractions? (also Line 254)
Line 241: Assume “this” data set refers to the marine data set? Language throughout could be more specific and precise (see also comment above).

Line 256: What is the evidence for the secondary emissions of 4rPAHs? Are the cited studies part of the same campaign? Or is this an inference based on a prior study? Or using measured monomers or ratios using a method in the cited study?

Line 262-267: Is the day/night ratio of PAHs to n-PAHs sufficiently explained by photochemistry? Or may there be measurement-based explanations as well? For example, do the citations referenced support that the photolysis rates of n-PAHs are faster than those of PAHs? If so, this should be explicitly stated. Is the much higher day/night ratio of PAHs surprising given reported oxidation timescales of hours? What exactly is meant by “the same could be reflected” in line 267? Emission rates are governing ratios (based on line 271)? Or that n-PAHs are photochemically degraded faster than precursors (based on line 275)?

Are the lowest reported remote concentrations due to changing urban emissions? Changing photochemistry? Or improvements of in analytical techniques? Some combination? Further discussion on this point is warranted.

Line 289-290: Particle phase distributions? What does the symbol theta represent?

Was the pp-LFER also used for the PAHs? Discussion focused on n-PAHs. How sensitive are the partitioning results to the assumptions in the pp-LFER? What are the uncertainty bounds on the phase predictions? Are they significant enough to influence any conclusions presented regarding phase partitioning? More discussion on the shallow slopes observed for the marine n-PAHs would be valuable. The model does not seem to be capturing any of the observed variance.

Is there a distinction between observed yield and potential yield? Or are these terms being used interchangeably?

Line 436-437: What is meant by “hardly limited by”? 
Line 446-447: “Levels” of what?

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