

Interactive comment on “Source apportionment vs. emission inventories of non-methane hydrocarbons (NMHC) in an urban area of the Middle East: local and global perspectives” by T. Salameh et al.

Dear referee,

We would like to thank you for your comments, which significantly improved the quality of this work. We have revised the manuscript entitled “Source apportionment vs. emission inventories of non-methane hydrocarbons (NMHC) in an urban area of the Middle East: local and global perspectives” according to your comments. Kindly find below our response to the comments.

Sincerely yours,

Thérèse Salameh

Anonymous Referee #1

The authors use two datasets comprised of 56 non-methane hydrocarbons, CO, and NO_x measured in winter 2011 and summer 2012 at a suburban site near Lebanon in order to investigate the relative contributions of various NMHC sources in the region using positive matrix factorization. The output of the source-receptor model is used to identify the major NMHC sources by comparison to measured sources and literature values, quantify the relative contributions of each modeled source, compare seasonal differences in sources and source strengths, and compare the source factors to local and global emission inventories. Gasoline and traffic-related sources are the largest contributors to observed NMHC levels in both seasons. The observed NMHC to CO ratios are used to compare the PMF-derived source profiles associated with on-road transportation to emission inventories. The authors have determined that the relative contribution of the on-road transportation sector to total NMHC emissions agree with the inventory; however, the overall magnitude of NMHC emissions in the inventory may be underestimated by a factor of 1.3-1.6 in the Lebanon NEI and a factor of 2.7-3.4 in global emission inventories for the road transport sector.

I recommend publication with major revisions as detailed below:

Major comments:

1. The NMHC measurements are not presented in the manuscript, only the PMF outputs are presented. In the introduction, the authors state that data in this region of the world is particularly scarce and is only limited to a few pollutants. I'm curious why the authors would not publish their measurements here, even if it appears in the supplemental material. If the NMHC measurements are published elsewhere (or will be), then it should be made more clear within the manuscript. It looks to be a very valuable dataset that would be very useful to the larger scientific community.

Thank you for your comment. The data have been already published and described in details in two papers Salameh et al. 2014 and Salameh et al. 2015 that we cited within the manuscript. For more details, please find below are the two references.

Salameh, T., Afif, C., Sauvage, S., Borbon, A., and Locoge, N.: *Speciation of Non-Methane Hydrocarbons (NMHC) from anthropogenic sources in Beirut, Lebanon, Environ. Sci. Pollut. R., 21, 10867–10877, doi:10.1007/s11356-014-2978-5, 2014.*

Salameh, T., Sauvage, S., Afif, C., Borbon, A., Leonardis, T., Brioude, J., Waked, A., and Locoge, N.: *Exploring the seasonal NMHC distribution in an urban area of the Middle East during ECOCEM campaigns: very high loadings dominated by local emissions and dynamics, Environ. Chem., 12, 316–328, doi:10.1071/EN14154, 2015.*

2. The author's state that photochemistry was negligible, but do not provide any basis for this conclusion. This is particularly important topic since the PMF results show 2 different traffic related sources in which the author's identify by day and nighttime traffic patterns. Please provide a brief description of how the author's came to this conclusion as is very important for the interpretation of the PMF results.

The impact of photochemistry was largely described in Salameh et al. 2015 as follows: It was assessed through the comparison of night-time and daytime scatterplots during summer and during winter. We assume that there is no photochemistry during night-time and the composition of emissions does not change. The advantage of using the mixing ratios of pairs of ambient NMHC species is that they are not sensitive to dilution and air-mass mixing compared with absolute concentrations themselves. Examining the ratios is useful in exploring the influence of photochemical depletion for compounds with different atmospheric lifetimes. The results show that the most reactive compounds are slightly affected by photochemistry (ethylene, m,p-xylenes) in summer and not affected at all in winter. As for less-reactive NMHC, there is no effect of photochemistry during both seasons. Therefore, the contribution of the most reactive species within the PMF factors is independent of their reactivity. Please refer to Salameh et al. 2015 for more detailed information.

3. The uncertainties (i.e., the actual numbers) of each NMHC should be clearly stated in Section 2. Why did the author's not include CO and NO_x (especially as NO or NO₂) in the PMF analysis? It's not clear which species were excluded from the PMF analysis. For example, P26802 L10 says that species below detection limit were excluded, but P26804 L7 says they were replaced by 1/2 the detection limit, while P26813 L17 states that terpenes and OVOCs were not included in the PMF analysis. Does this mean they were measured, but excluded? If so, why? P26804 L5: What constitutes "missing data"? Were whole samples replaced by median values or only a few individual points within a sample. What percentage of the data was missing or replaced?

The uncertainties are added in section 3.2 after the description of the estimation of the uncertainty: "The relative expanded uncertainty ranged between 4 and 27% in summer for all the compounds excepting for octane which was 38%. In winter, the relative expanded uncertainty was between 5 and 35% for most of the compounds excepting for trimethylbenzenes, isoprene, octane and heptane which had an uncertainty between 40 and 60 %."

We did not include the CO and NO_x in the PMF analysis because we preferred to use them as ancillary data to help in the identification of the PMF factors. Some OVOCs were measured and will be the subject of an upcoming study.

We changed the sentence P26802 L10 : “NMHCs which are not identified during both seasons (like tetrachloroethylene) or their concentrations were frequently below the detection limits, were not considered in the PMF analysis.”

In Salameh et al. 2015, table 1 shows the mean, median, percentage of values below the detection limit (DL), maximum, and standard deviation of each species. The concentrations below detection limit of the species used in the PMF were replaced by 1/2 the detection limit.

The percentage of missing data ranged from 0 to 6%. Therefore, only a few individual points were replaced by median values.

4. There is very little discussion on solvent use as a potential NMHC source as the PMF analysis did not resolve a factor for this potential source. Does this mean that solvent use is a negligible source in the region, at least in comparison to traffic related sources? If so, how does this compare to the emission inventory?

Effectively, according to the national emission inventory, the anthropogenic sources including NMHC product source (like solvent use) is a negligible source of NMHC with a relative contribution of 6 % of total emissions.

5. Figures 2 and 7 are extremely hard to decipher and yet they are the subject of the majority of the discussion section. In the current format, the species names are unreadable and it is very difficult to compare the contributions of the various sources to a particular compound (e.g., propane). Suggested changes include stacking the various Factor outputs (panels) vertically so that the width of the graphs can be expanded and the names of the individual species can more easily read on the bottom of the figure. Also, it would make more sense to arrange the species by chemical family (alkanes, alkenes, aromatics, etc.) rather than by retention time which has nothing to do with the source profiles or interpretation of the PMF results.

We changed figures 2 and 7 following your suggestion, and we arranged the species by chemical family. We changed also the numbering of the factors according to their names in both seasons.

Minor comments and technical corrections in order of appearance:

1-P26797 L13-16: I'm not sure what the following means: “The national road inventory shows lowest (lower?) emissions than the ones from PMF but with reasonable difference (?) lower than 50%. Global inventories show higher discrepancies with lower emissions up to a factor of 10 for the transportation sector.” Please rephrase.

The text is changed to: “The PMF analysis finds reasonable differences on emission rates, of 20 – 39 % higher than the national road transport inventory. However, global inventories (ACCMIP, EDGAR, MACCity) underestimate the emissions up to a factor of 10 for the transportation sector.”

2-P26798 L5: urbanization → urban

We kept “urbanization” because it describes better what we want to say.

3-P26798 L11: deteriorated → deteriorating?

We mean “the air quality has been deteriorated” and not deteriorating.

4-P26798 L24: Non-Methane Hydrocarbons does not need to be capitalized.

We changed it.

5-P26798 L24-P26800 L5: This is one massive paragraph that can easily be split into multiple smaller paragraphs that will be easier to read and digest.

We separated the paragraph into 5 smaller paragraphs.

6-P26799 L4: exhausts → exhaust

Changed.

7-P26800 L14: “and” in China

Added.

8-P26804 L1-4: Why can the integration error be neglected? What is a “coverage factor”?

The systematic integration error can be neglected because the chromatograms of the air samples and the calibration gas for the considered species were quite similar, without any integration issues. This explanation is added to the manuscript.

The coverage factor “k” is chosen on the basis of the desired level of confidence to be associated with the interval defined by $U = k \cdot u_c$ (where “U” is the expanded uncertainty, “ u_c ” is the total calculated uncertainty). $k = 2$ defines an interval having a level of confidence of approximately 95 %, when the normal distribution applies.

9-Move Section 4.1 to Section 2

We find that it is more convenient to keep it within the results section since it describes the meteorological conditions during these measurements campaigns.

10-P26804 L23-25: Is domestic heating source included in Factor 1 (regional traffic)? This is confusing.

Factor 1 represents a combustion source mainly related to regional traffic. Nevertheless, the small peaks observed in the evening at 20:00 - 21:00 can be also due to domestic heating because the measurement site is surrounded by a residential area. We could not separate the combustion related to domestic heating because we do not have specific tracers.

11-P26807 L14: Please describe what “hot soak” refers to.

Hot soak is a type of evaporative emissions which occurs when a warmed-up vehicle is stationary and the engine is stopped.

12-P26807 L18: Remove “In fact,” and “the” before light-duty

Removed.

13-P26808 L1” “The fingerprint is consistent with the one of other gasoline evaporation profiles.” Please rephrase for clarity.

The sentence is changed to: “The fingerprint of the chemical composition is consistent with the ones of typical gasoline evaporation profiles established by near field measurements.”

14-P26810 L7: North-East → northeast; South-West → southwest

Changed in the text.

15-P26810 L11: It says that the emission increase, but does the composition change? The composition is most critical to the PMF analysis.

According to the French study of Fontaine (2000), less significant changes impact the composition when the vehicles speed is low in the case of vehicles running on gasoline without a functioning catalytic converter. Even though, the contribution of alkynes, alkenes and alkanes is more pronounced at low vehicles speed.

Vehicle engine, vehicle age and maintenance, environmental conditions especially ambient temperature, as well as emission controls, and fuel composition are important factors to consider because they may change from a place to another.

16-P26810 L24: change “suggesting the gas leakage” to “suggesting natural gas (or liquefied petroleum) gas leakage”

Changed.

17-P26812 L30: “. . .but always maintained as the main contributor to NMHC emissions.” This needs rephrasing or removal.

Removed.

18-P26813 L8: “. . .and is associated with levels of uncertainties. . .” I’m not sure what this means.

Removed.

19-P26813 L27: somehow → somewhat

Changed.

20-P26815 L1: “reasonably underestimates” Remove “reasonably” or rephrase.

We removed “reasonably”, the sentence is changed to: “These results suggest that the inventory underestimates the road transport emissions in a reasonable way.”

21-P26815 L3: “estimated to be 29 Gg”

We added “be”.

22-P26815 L22: “estimate to be 8 Gg, which is a factor of 6 to 10 lower than the annual emissions. . .”

The sentence is changed to: “The total annual NMVOCs transportation emissions from the ACCMIP are estimated to be 8 Gg, which is a factor of 6 to 10 lower than the annual emissions...”

23-P26815 L25: “factor of 2 to 3” . . .what?

We changed the sentence to: “...the annual transportation emissions are estimated to be 23 Gg, which is a factor of 2 to 3 lower than the road transport national inventory emissions and the annual emissions obtained via the PMF results.

24-P26815 L28-P26816 L3: This is an incomplete sentence and the meaning is unclear.

The sentence is changed to: “This comparison reveals that global inventories are not consistent between each other. Additionally, The global inventories do not describe the methodologies applied neither the data used (emission factors, NMVOC species considered...)”