The authors made considerable efforts to modify the paper. However, unfortunately, there are still multiple issues which have not been addressed in a satisfactory way. Still, the current state of the paper and the replies made by the authors do not warrant the publication of the paper in its current stage in ACP without additional discussion.

Here I repeat those issues, which I believe need some additional clarification. First I list my initial review point, then I add my updated remark, denoted as "Reviewer Remark":

1) On the background of existing literature I am not sure what the real novelty of this paper in terms of methods and results is. While I agree that C2 and C3 measurements have not yet been done before in Athens, it seems the inclusion of these does not yield more findings than already reported by Kaltsonoudis et al (2016). On the other hand, the Panopoulou et al paper makes same sketchy description of meteorological impacts on NMHCs, but lacks some elaborate analysis similar to those presented in Rappengluck et al (1998) for transport effects and also Kourtidis et al (1999) for temperature effects for Athens. As the authors make an important point on page 2, L27-29, that there have been new conditions during the economic crisis years (i.e. competing traffic vs wood burning emissions) it would be actually meaningful to perform a comparative analysis between the data sets reported 20 years ago and the ones reported by Panopoulou et al. It seems both studies include continuous NMHC measurements and PMF source apportionment analysis would be feasible and would provide interesting insights.

Reviewer Remark:
In their reply the authors did not really demonstrate the novelty of their work apart from the fact their measurements include C2-C3 data. However, including these NMHCs did not yield new knowledge about NMHC sources in Athens beyond the paper by Kaltsonoudis et al (2016). Kaltsonoudis et al (2016) even included detailed source apportionment for traffic and biomass burning for the Athens case, which the authors did not apply and did not address. What do we learn from this paper which has not yet been described earlier. Even worse, now the authors have removed the reference to Kourtidis et al (1999) who described temperature effects on traffic NMHCs emissions for the same city.
The authors argue: "However direct comparison with the work performed 20 years ago is difficult considering differences in sampling period (summer versus winter and thus different photochemistry), location, sampling method and analytical techniques." I cannot completely follow this argument. Emission studies can be done for nighttime periods minimizing photochemical effects, all the locations are within the GAA and sampling methods and analytical methods are similar. Even, if the suite of NMHCs may not be exactly the same, the application of source apportionment methods such as PMF would just require a few

representative NMHC describing source profiles. There is not necessarily a "complete" suite of NMHCs necessary as it is also shown by the Kaltsonoudis et al (2016) paper, who applied a rather limited VOC dataset. Did the authors try at all to make comparisons? Again, in my opinion this would be feasible and would provide interesting insights.

Abstract:

Page 1 L20-21:
What do the authors exactly mean by local meteorology, as this term is quite unusual? Its connotation would mean that it is not representative for a larger fetch.
I disagree that "local" meteorology would control the variability of NMHC levels alone. What about the temporal variability of NMHC emissions?

Reviewer Remark:
In general, the term microscale meteorology is fine. However, still this would rather describe specific condition for mixing processes observed at a given site, which may not necessarily be similar for a different location within the urban area. For instance, the built environment would have a significant impact on these exchange processes.
In addition, what about PBL variations, which are not included in the term microscale meteorology? Also, still I believe, that the temporal variability of NMHC emissions is an important factor. Just one example: a typical morning rush hour peak is not entirely explained by low winds that are often observed during the same time frame. I would believe that low a PBL height and increased traffic emission would be an important factor. I would still doubt that "Microscale meteorological conditions and especially wind speed seems to ***control*** the variability of NMHC levels...".

Page 1 L27-29:
Why does the present data not allow for the quantification of the relative contribution of fossil fuel and wood burning for heating purposes?

Reviewer Remark:
Still, I have some trouble with the revised sentence. On one side the authors state that "For the night peak, the selected tracers and profiles ***clearly*** indicate contribution from both traffic and domestic heating..." and then continue that more specific tracers would be necessary for quantification of these sources. The latter statement contradicts the first statement: either the data set has tracers and profiles that clearly indicate these specific sources or not. Also, the authors emphasize the night peak. This implies that daytime emissions would be different. I would assume that domestic heating sources would become less, while traffic sources would

increase and it seems the authors can differentiate this different behaviour in their data-set. Why would, for instance, a source apportionment method based on these specific tracers and profiles not be able to estimate relative source contributions?

Introduction:

Page 2 L30: It would be fair to mention how many NMHCs were actually measured as it seems that the paper does not report some important NMHCs such as 1,3-butadiene and others, for instance.

Reviewer Remark:
The reply given by the authors is not convincing. Still, it would be fair to mention the number of NMHCs measured in this range and the number of NMHCs in this range used in this study in order to make a precise statement here and avoid any speculation.

Page 2 L34 - page 3 L1: The authors should mention why the analysis is restricted towards traffic and heating impact on NMHC levels.

Reviewer Remark:
There is still an issue: in order to perform an "..investigation of traffic and residential heating impact ***on*** the NMHC levels..." the authors would still need to verify and quantify any other potential NMHC source, regardless of its magnitude. Even, if there are no industrial sources as the authors point out, I still would think there are solvent and evaporative sources. Also, what about the port of Athens?
Otherwise, I am not sure what still needs to be investigated exactly for this task (3).

On line NMHC measurements

Page 4, L5: Why is only toluene used? Why not at least ethylbenzene and the xylenes in addition? Would the exclusion of these NMHCs not introduce a bias into the data analysis, as important tracers for solvent emissions are excluded? What are the uncertainties and the detection limits for this GC?

Reviewer Remark:
This appears cherry-picking to me. The authors state that they focus on C2-C6 NMHCs, then include a C7 compound from a different instrument, but exclude other NMHCs from that different instrument. The authors did not address my question whether the exclusion of these other NMHCs would introduce a bias in the data analysis. As the authors have a complete suite of NMHC measurements, it would be straightforward to include all NMHCs, and make a

source apportionment analysis first, and not rely on a reference only (Vrekoussis et al., 2013), which did not include VOCs.

Tunnel measurements

Page 4 L16: The authors should mention the length of the tunnel, whether lanes were for both directions (there could also be dedicated tunnels for one direction only), if there was any artificial ventilation and if there might have been any limitations on traffic through this tunnel (in some cases heavy duty traffic is not allowed). In any case an estimate of the traffic fleet composition (e.g. heavy duty vs light duty vehicles) would be helpful. All these factors have an impact on the NMHCs levels. At what location of the tunnel did the authors make the measurements exactly? I see the measurements were taken on 12 May 2016, which is different from wintertime. Wouldn't the temperature be different from wintertime and wouldn't this have an enhanced impact on NMHC emissions through evaporation, for instance?

Reviewer Remark:
It seems the description of the tunnel is still missing in the manuscript. Also, the authors did not mention explicitly whether this tunnel contains all lanes for both driving directions or whether there were separated compartments for each driving direction. They did not mention either, whether there was any artificial ventilation or not.

Temporal variability of NMHCs

Page 4 L28: I do not understand the concept of data coverage here, as it is not explained. It could refer to the percentage of data above the detection limit vs maximum available data, but this does not make complete sense, as I doubt there were any data of ethane below the detection limit, for instance. However, it cannot be true either that it refers to the data availability vs maximum potential data availability during the time period reflecting instrumental potential instrumental malfunctions and/or failure. This should be clarified. The only thing I understand is that the there has been some interruption of NMHC data contrary to what the authors claim in the abstract of the paper.

Reviewer Remark:
The authors statement of 87% data availability would imply that the instrument did not work uninterrupted (again, I believe that such a species like ethane would always be above the detection limit, if the instrument is working properly). As a consequence I would recommend to replace the statement made in the abstract of the paper (page 1, L16) "..to our knowledge,

time resolved, uninterrupted data of NMHCs..." by "..to our knowledge, time resolved data of NMHCs...".

Page 5, L15: The authors should clarify why the reader should bear in mind differences in sampling methods and analytical techniques. Are some of the sampling methods and/or analytical techniques and associated results listed suspicious and cannot be compared to each other?

Reviewer Remark:
The authors did not change much. The authors still state that "....this decrease has to be seen with cautious considering differences in [...]sampling method and analytical techniques". Are some of the sampling methods and/or analytical techniques and associated results listed suspicious and cannot be compared to each other?

Page 5, L18-20: The same comment as above applies here. As long as there is no more elaborated comparison, the presentation of the data remains generic.

Reviewer Remark:
Why would it be important to note that Paris is located at the mid-latitude and in the northern hemisphere with regard to NMHC emissions? Would it be different for city at the same latitude, but in the southern hemisphere? For instance, would traffic emissions depend on the latitude?
There is not such a thing as oceanic continental climate; this does not exist! Apart from that the climate of Paris is definitely not continental! There are no cold winters in Paris! Also, in Paris, it can be pretty hot during summer time, which cannot be considered "mild". I would rather use that term for the winter period in Paris. The authors want to check an appropriate climate description for Paris.

Page 5, L25-27: The authors neglect to mention the annual variability of other NMHC sources, e.g. evaporation losses.

Reviewer Remark:
With regard to traffic related evaporative NMHC emissions the authors need to consider the work by Kourtidis et al. (1999) who specifically addressed the processes related to these emissions for the same city, Athens. The NMHC mixture might have changed, but not the underlying processes. Unfortunately, the authors removed this reference from the previous manuscript version.

Page 6, L4-7: From Fig 3 I see that B_{ff} increases similarily to B_{wb} at night. Why can the authors make the statement that traffic would not be as important as heating?

Reviewer Remark:
The authors do not consider the nighttime variation of the PBL which would also contribute to a sustained higher level of pollutants, regardless of their origin.

Page 6 L9-10: I disagree. Usually, PBL heights are at a minimum during morning hours before sunrise, unless the authors can show other evidences for their statement.

Reviewer Remark:
I did not see the change the authors may have done in the text (mentioning the line would be helpful).

The role of meteorology on NMHC levels

Page 6 L15-20:
This is a pretty generic description. It is well-known that the concentration of primarily emitted gaseous pollutants will decrease due to dilution regardless of their chemical class. However, windspeeds < 3 m/s alone would not indicate the presence of local sources. This would only be true for calm winds. From the plots it seems like these are skewed distributions with maximum concentration values around 2 m/s or so. This would rather indicate some regional flow impacts, which the authors neglected to consider. It seems a more elaborate analysis of windspeeds and their effect on NMHC levels in the Athens area has already been presented in Rappengluck et al (1998).
With regard to potential long-range transport it is actually interesting to see that there is some acetylene data still around 5 ppb or so at windspeeds around 9 m/s and higher. In fact, those are very high acetylene values despite strong dilution. What is the reason for this?

Reviewer Remark:
As the x-axis of figure 5 shows discrete values, it would be good to have a clear definition of these ranges. Also, are calm wind situations considered, i.e. below the threshold of the wind sensor (both, in figures 5 and 6)?

Identification of NMHC emission sources

Page 8, L28-30: Did the authors also apply the baseline subtraction for the tunnel measurements?

Reviewer Remark:
The authors should mention this limitation in the text.

Page 8, L30-31: How can the authors justify that their tunnel measurements are not influenced by outside air masses?

Reviewer Remark:
There must be some dilution. A tunnel is not a closed box and it seems the length of the tunnel is only 200 m.

Page 8, L31-32: I completely disagree on the authors' statement. The authors neglect to mention what they consider "dominant species", however just looking into NMHCs such as acetylene, benzene, and toluene, the two profiles "Morning Peak Athens" and "Highway Tunnel - Athens" are completely different: while acetylene for the "Morning Peak Athens" is about 6-7 times higher than for the case "Highway Tunnel - Athens", benzene and toluene values are about 2-3 times lower at the same time.

Reviewer Remark:
I agree that the "Morning Peak Thissio" and "Morning Peak Patission" profiles agree with each other within the uncertainties. Still their comparison with the tunnel measurements and both tunnel measurments among themselves are very different. The authors themselves make a reference to various papers which report important differences between tunnel measurements worldwide and also state that there is a possibility that the car-fleet in the tunnel is not representative for the GAA. Why then compare two consistent real-world street canyon measurements with tunnel measurements, whose data is questionable?

Page 8, L32 - Page 9, L1: I disagree here again! I do not see that profiles fit nicely. Instead, there are a lot of significant differences. Also, what do the authors consider "common NMHCs"?

Reviewer Remark:
See my Reviewer Remark above/

Page 8, L1-3: Why should there be higher traffic related butane fraction due to evaporation in ambient air than in the tunnel? Even more surprising, as the tunnel measurements were taken in May, which presumably has warmer temperatures than wintertime. Also, when butanes should be related to evaporation why does propane, another prominent tracer for evaporation, show pretty similar values in the tunnel measurements compared to the "Morning Peak Athens" data?

Reviewer Remark:
The remarks made by the authors in the comments for P8, L31-32 were: "Moreover higher values of ethane, propane and butanes that are depicted in the morning hours at the urban sites relatively to the tunnel measurements, reflect the increased numbers of LPG powered vehicles in Athens and natural gas-powered buses". I deduce again, that the tunnel measurements were not representative for the GAA. For me, it does not make much sense to include the tunnel data then.

Page 8, L12-14: The definition of the background concentration appears odd. How can the minimum value between 12:00-17:00LT be representative for the nighttime period 18:00-05:00LT? Both are pretty long periods (5 and 11 hours, respectively). From Figs. 4 and 5 we learnt that the NMHC concentration critically depends on wind speed and wind direction. How can the authors make sure that such changes in wind speed and/or wind direction would neither occur during the daytime reference period nor during the nighttime period?

Reviewer Remark:
The authors did not address the impact on wind direction. Background NMHC concentrations can critically change with wind direction, in particular at coastal locations. How can one minimum NMHC value observed during mid-day and potentially under one distinct wind direction be representative for all potential wind direction conditions at nighttime from 18:00-05:00 LT? Also, the authors neglect daytime photochemical degradation, albeit limited, but still present in those months.

Page 8, L16-19: Are these differences statistically significant?

Reviewer Remark:
The statement made by the authors should be added in the manuscript.

Page 10, L13: What were those "different meteorological profiles"?

Reviewer Remark:
Still, the term "profile" does not make sense, as long as it is not defined (for instance, there are vertical temperature profiles).

Page 10, L14-16: This is not supported by the data presented in the paper!

Reviewer Remark:
Still, the tunnel measurements would not support this as discussed earlier.

Table 1: Remove the term "worldwide" in the table caption, as Table 1 shows a few selected data from the Mediterranean/European area at the most. What does the second sentence of the table caption refer to? What quantities are compared in this table: means or medians or ....? What do the authors mean by "sampling" frequency: sampling duration or measurement cycle? There is no information given for "sampling frequency" for Baudic et al., Salameh et al., and Durana et al.. Why are the results for the summer 2012 and winter 2013 Athens campaigns reported by Kaltsonoudis et al (2016) not listed in this table? At least, results for isoprene, benzene, and toluene would be comparable.

Reviewer Remark:
The authors should state explicitly that mean values are shown in the table.

Figure 10, figure caption:
I disagree that such a long time period (18:00-05:00 LT; 11 hours!) can be considered a nighttime "peak". Error bars should be included. I do not see that the values shown in the figure add up to 100%.

Reviewer Remark:
I did not argue the term "nighttime", I argued the term "peak", which does not make much sense for a time period of 11 hours.

References:
Kourtidis, K. A., Ziomas, I. C., Rappenglueck, B., Proyou, A. and Balis, D.: Evaporative traffic hydrocarbon emissions, traffic CO and speciated HC traffic emissions from the city of Athens, Atmos. Environ., 33(23), 3831–3842, doi:10.1016/S1352-2310(98)00395-1, 1999