

Comments to Reviewer # 1:

We appreciate the comments of the reviewer. Please find a detailed reply to all issues.

The authors perform PMF analysis on 7 years of in-situ, daily NMHC measurements from a site in Southern Germany. They perform some tests of the PMF model assumptions to evaluate the robustness of the results and the appropriate number of factors they use. They interpret the resulting factors qualitatively in terms of a combination of source profiles and compound lifetimes. It appears the main findings are 1) the PMF analysis leads to plausible results that bear a general resemblance to various emission sources; 2) the abundance of the longer lived compounds was higher than the shorter lived compounds; 3) anthropogenic NMHC were higher in winter than in summer; 4) isoprene is higher in summer and has high reactivity.

One could certainly learn something useful with this type of long-term dataset ... for instance, emission trends, possible seasonality in emissions for various compounds, etc. But the analysis here doesn't provide any of that. In the end it's not clear that we've learned anything here that wasn't already known. As such I don't believe the paper rises to the level of meriting publication in ACP.

We beg to differ from the assessment that there is basically no new information coming out of this paper:

- PMF has been applied for NMHC in remote areas in some studies, but so far there has been little to no discussion about the applicability. Our paper is the first to discuss the capability of the PMF method to identify source categories of NMHC in remote areas. The concept of strict source apportionment related to emission factors which requires mass conservation was not used, instead, a new concept of PMF usage for identifying the dominant factors contributing to rural site NMHC observations was proposed, and the factors were related to emission sources and aging.*
- It is the first discussion of a method suggested by Sauvage et al. (2009) who treated atmospheric chemical processing by adding uncertainties to the PMF model that so far has not been evaluated at other sites. In addition, another method of Yuan et al. (2012) that looks at photochemical aging and influence on PMF results is now included and discussed.*
- It is the first manuscript on an analysis of a continuous multi-annual dataset of NMHCs for the pre-alpine area of Germany*
- It is the first receptor model applied for this region and compounds.*
- It first discusses the seasonality related to OH seasonality and infers information about the seasonality of sources*

We do not yet see the chance to analyze emission trends. A seven year dataset is too short for such an analysis, a much longer time series would be necessary.

We beg to differ on the lack of seasonality. In our opinion, the seasonality has been analyzed and described in quite some detail within the manuscript. Nevertheless, we extended the analysis by following the specific comments below.

Specific comments.

8156, 15 and subsequent discussion throughout the rest of the paper: 'short-lived compounds emitted at constant rates throughout the year ... would be factors of about 7 higher in winter than summer'. This will clearly depend on the lifetime of the compound at hand. Later on you apply this reasoning to interpret the seasonal cycle for different compound groups in terms of whether there is

a likely seasonal cycle in emissions (i.e. if the winter:summer ratio differs from 7, which is the OH ratio). This is a nice idea, but what is needed here is a simple box model calculation of the summer:winter ratio you would expect as a function of lifetime, given constant emissions. For instance, later (8160, 26) you apply this reasoning to the “remote” factor. However ethane is the main compound for this factor, and it is almost inert in winter (lifetime 450 days at OH = 1e5 molec/cm³). Clearly the steady-state argument is not valid here. It also seems that this type of analysis is much better suited to the compounds themselves than the factors, because of the dependence on lifetime.

A couple other points to consider for that type of analysis: rate constants also vary with temperature (not just OH); and prevailing transport patterns (i.e. air mass origin) will likely differ seasonally.

We agree that our approach of comparing the annual courses of the factors to the OH concentration was oversimplifying actual processes. We now applied a simple box model as suggested and included temperature dependent reaction rate constants both for OH and ozone reactions. This enabled us to perform a better and more sound interpretation of the seasonality of the factors. The factor seasonality was determined by using the factor-loadings as weights in combined seasonality of all contributing VOC compounds. The advantage of the PMF method and receptor models in general is that no additional meteorological or chemical processing data is needed. The performed interpretation with the OH annual course merely constituted additional support for the interpretation and the results as discussed in the revised manuscript support the results of our original apportionment and the applicability (and limitations) of PMF to rural datasets.

8161, 24: ‘long-lived evaporative compounds had factor 7-9 higher mixing ratio contributions in winter than in summer, although their atmospheric turn-over remained the same all over the year’. What is being said here? That alkanes have the same loss rate in summer as winter??

We changed the whole part about seasonality in the revised manuscript; this statement is no longer existent. However, this was a misunderstanding, as it was intended to say that the product of enhanced OH with reduced NMHC in summer yields a constant “turn-over” (= product of OH and NMHC concentration with rate constant) throughout the year.

8146, 10: ‘and in particular NMHC’ ... why in particular? Are NMHC the main category of VOC contributing to ozone production compared to OVOC?

This statement was withdrawn as it certainly cannot be generalized. Among the primarily emitted VOC (as ozone precursors), the NMHC are generally more important than the OVOC, however, in atmospheric chemistry they go through multiple cycles of oxidation with in consequence higher contribution of OVOC to ozone generation. We did not want to go into such details and thus, we changed the text to ‘...and amongst them NMHC...’

8146, 19: ‘expected a strong increase of biogenic NMHC emissions caused by temperature stress’ ... not clear what the context is here.

‘...stress with future increase of global temperatures...’ was added to make it clearer.

8146, ‘while in urban areas they (biogenics) play only a minor but non-negligible role’. Surely this would depend on the urban area, wouldn’t it? What about a city like Atlanta?

We absolutely agree with this statement. Thus, the text has been altered to the following:

‘... while in many urban areas they play a minor role due to high amounts of anthropogenic emissions; however, this is highly dependent on the type and location of the urban area. For major metropolitan areas with large anthropogenic emissions (e.g. Houston, Atlanta) a large impact of the highly reactive biogenic VOC on ozone formation and OH chemistry has been shown (e.g. Chameides et al., 1988;

Leuchner and Rappenglück, 2010; Mao et al., 2010).'

8160, 24: 'ethane appears as an indicator for aged air masses'. This is phrased a bit oddly. Surely ethane is present in pretty much all air masses. It's really the absence or low abundance of other (shorter-lived) compounds that indicate aged air masses ... only the longer-lived compounds like ethane are left.

We absolutely agree with this statement. But nothing different is stated here. We changed the phrase to make it clearer to '...The high abundance in combination with the lack of shorter-lived compounds were indicators for aged air masses. ...'.

8151, 12: 'obtained reasonable agreement' ... what does 'reasonable agreement' mean in this context?

There was no mentioning of 'agreement', only 'obtained reasonable results'. But the phrase '...in terms of plausible factor compositions and contributions of the source categories...' was added to make the statement clearer.

8159, 23: 'aromatics possibly had larger fetch areas during winter due to the longer life-times' ... why 'possibly'? Doesn't this stand to reason?

We agree. Due to changes in the text this statement does not appear in the revised manuscript.

8154, 12: 'CO is a product of incomplete combustion' ... it is also produced photochemically in the atmosphere.

The comment is correct, however, we anyways withdrew this sentence

8154, 14, 'NO and NO₂ are mainly emitted by combustion engines' ... what about power plants etc?

In Germany, the traffic sector is the dominant source for NO_x followed by the energy sector. Going more regional, in Bavaria the contribution of fossil fuel burning power plants is rather small (ca. 20%) and accordingly, only small contribution by power plant to the short-lived NO_x exist.