

Reviewer comments

Overall:

The manuscript presents observations of VOCs at the Ersa site in Cape Corsica over a two-year period and provides a comprehensive description and analysis of their behaviour during this time. PMF analysis of the data is presented along with comparison to other station across Europe. The plots are clear and generally well presented. The length of the manuscript, however, is something of a problem with lots of repetition throughout. I am sure there is an interesting story here, but it is difficult to assess what that is from the current article. Careful consideration of each section, it's findings and their relevance is required in order to make the manuscript worthy of publication. I therefore recommend a major revision of the manuscript. I do however, urge the authors to continue to work on this as I feel it can be a very nice piece.

The authors should consider writing the manuscript in terms of the features observed at the site and then use the data and plots to explain that behaviour. In its current format, each compound or group is considered separately and methodically (which leads to a comprehensive, but repetitive narrative) whereas, the behaviour of these different compounds can often be explained by the same phenomena (e.g. a changing boundary layer or temperature difference).

I have included suggested changes to individual sections, but these may not be relevant to the newly written article.

Abstract

Page1, Line17:

"... The VOC speciation was largely dominated by oxygenated VOCs ..."

Should this be the VOC abundance or mass? I'm not sure how speciation can be dominated.

P1, L18:

"VOC temporal variations are then examined..."

Past tense, should be "were examined"

P1, L19:

"... and solar radiation ones."

Delete "ones"

P1, L20:

"Anthropogenic compounds have shown an increasing concentration trend in winter (JFM months) followed..."

This reads as though the concentrations increase between these months – ie March is bigger than February which is bigger than January – this doesn't appear to be the case from figure 6

"Anthropogenic compounds showed increased concentrations in winter (JFM months) followed..."

P1, L21:

"... and different concentration levels in winter periods of 2013 and 2014."

These are inevitably different, but the question is by how much are they different?

Suggest including "by up to XX% in the case of compoundY"

P1, L21:

“OVOC concentrations were generally higher in summertime, mainly due to secondary and biogenic sources, whereas their concentrations during fall and winter were potentially more influenced by anthropogenic primary/secondary sources.”

This sentence seems a little confusing to me. I agree that the secondary sources of OVOCs will be increased during summertime and that the contribution from biogenic sources will also be greater during summer. As it is written though, it sounds like the anthropogenic secondary sources only contribute to the OVOC concentrations during the winter months. This is not the case, the anthropogenic secondary sources will also increase during the summertime.

P1, L26:

When listing the PMF factors, I suggest that these be listed in order of significance in terms of relative contribution.

P1, L30:

at the receptor site are also

Suggest changing “receptor site” to ERSA station or observatory

P2, L2:

“... winter 2014 ones could ...”

Delete “ones”

2.2.1. VOC measurements

Where there any compounds measured by the multiple measurement techniques used in the study? If so, were there any comparison exercises performed to ensure consistency?

P10, L14:

“... and the lowest ones in winter ...”

Delete “ones”

P10, L22:

Paragraph beginning “On one hand, western European ...” and ending “...across the north Atlantic toward the British Isles (Kendon and McCarthy, 2015).” on P11, L7 seems excessively long. Of key relevance here (to VOC observations) is that the temperatures were different (lower in 2013) and a short statement/sentence to say the lower temperatures were observed across Europe, with relevant citations, would suffice.

P11, L15:

“Relative humidity globally followed opposite seasonal variations than temperature and solar radiation ones.”

Should read:

“Globally, relative humidity followed opposite seasonal variation to temperature and solar radiation.”

P11, L15:

“Relative humidity globally followed opposite seasonal variations than temperature and solar radiation ones.”

P11, L15:

“In June 2012, air was dryer compared to June 2013 and 2014 mean relative humidity values ...”

Delete:

“mean relative humidity values ...” they’re not need ed here since these are described in the parentheses.

P11, L15:

“The wind speed did not show a clear seasonal variation over the two years studied, except maybe higher wind speeds in April and May that could induce higher dispersion of air pollutants and could advect air pollutants from more distant sources to the receptor site.”

Suggest changing to ““The wind speed did not show a clear seasonal variation over the two years studied. Slightly higher wind speeds in April and May 2014 which could induce higher dispersion of air pollutants and advect air pollutants from more distant sources to the receptor site.”

P11, L19:

“May 2014 encountered particularly windy conditions.”

I don't think this sentence is warranted (only 1.5 m/s higher than April) and would suggest removing it this sentence, it is not needed

P11, L31:

Air masses spending longer periods over the ocean will indeed have undergone more atmospheric processing, but they may also have more influence from oceanic sources of VOCs. While these are likely insignificant compared to the anthropogenic inputs from Continental Europe, I feel they should be mentioned here.

P12, L4:

“showed relatively close transport times”

Change to “... short transport times ...”

3.3 VOC mixing ratios

P12, L8:

The statement “Descriptive statistical results for a selection of 25 VOCs, which showed significant concentration levels during the 2-yr studied period, are summarized in Table 4” implies that more VOCs were measured during the period, but are not reported here because they were below some threshold value decided upon by the authors. If this is the case, there should be a statement describing the selection criteria used to define the “significant concentration levels”.

P12, L24:

“On the contrary, larger α -terpinene contribution was noticed during the summer intensive campaign than the 2-yr observation period.”

Were these observations made using the same technique or could there be some instrument bias associated with this result? It is important to clarify and state that in the text here.

P12, L28:

“... dispersion, dilution processes ...”

are these the same thing?

Section 3.4.1. Biogenic VOCs

P13, L2 - 18:

The authors state “Surprisingly, isoprene and α -pinene concentrations were drastically lower in July 2012...” and then go on to state that the temperature and solar radiation during July were lower, therefore, I fail to see the surprise here.

The bigger surprise here seems to be that the July 2013 isoprene falls below the July 2012 level despite the temperature and solar radiation being higher (increasing emissions) and the wind speed being lower

(increasing dispersion) during that period. Perhaps including the wind direction in figure 4 may help to explain this?

P13, L10:

“... which may be related to the fact that temperature and solar radiation were more favourable to enhance biogenic emissions in June 2012 compared to June 2013 and 2014 meteorological conditions ...”

There is also the effect of relative humidity to consider here. Figure 4 shows the relative humidity was lower in June 2012 and 2014 compared to 2013, see the work of Ferraci et al. for the effect of drought conditions on the emissions of isoprene. Links to this research would be useful here.

P13, L16:

“This finding could be the result of a weaker degradation of α -pinene due to lower ozone concentrations observed from October to December compared to summer ...”

Emissions of isoprene are light and temperature dependant while monoterpenes are thought to be solely temperature dependant. I'd suggest that the difference in seasonal cycles of isoprene and alpha-pinene is due to the difference between the solar radiation and temperature profiles: solar radiation falls much quicker than temperature which may have the effect of “switching off” the isoprene emissions before the alpha-pinene emissions.

3.4.2 Anthropogenic VOCs

P13, L16:

“... characterized by almost the same seasonal variation ...”

Replace with “similar”: “... characterized by similar seasonal variation ...”

P13, L27:

“... with the highest atmospheric lifetime ...”

Replace with “Longest”: “... with the longest atmospheric lifetime ...”

P13, L27:

“... considering its low photochemical reaction rate with OH radicals ...”

“... due to its low photochemical reaction rate with OH radicals ...”

P13, L28:

“... It is typically emitted by natural gas use and can be also considered as a tracer of the most distant sources.”

Transport and storage of natural gas are also important sources here.

P14, L2:

“... four to ten times higher than ethane one (Atkinson, 1990; Atkinson and Arey, 2003)”

Remove “one”, it's not required here.

P14, L4:

“... e.g., Leuncher et al., 2015).”

Should be “Leuchner”

P14, L6:

“... winter 2014 ones.”

Remove “ones”, it's not required here.

P14, L11:

As a result, winter variations of concentration levels concerned at a time close sources and more distant ones and will be more investigated thereafter (Sect. 4.2)."

I don't think this sentence makes sense. Just a statement that the winter period will be investigated later in the manuscript would suffice.

3.4.3 Oxygenated VOCs

P14, L21:

"Nevertheless, acetaldehyde is only produced as a second or higher-generation oxidation product of isoprene for all its reaction pathways with atmospheric oxidants (Millet et al., 2010)."

I don't know what this means – further clarity in the text is needed here.

P14, L30:

"... with air temperature one,"

Remove "one", it's not required here.

P14, L2:

"... which can denotes that"

Replace with "denote": "... which can denote that"

P14, L32:

"These findings are in agreement with a large result on BVOC oxidation on the local photochemistry.!

Remove this sentence, not required.

P14, L32:

"... remained relatively significant during fall ..."

significant to what?

P15, L9:

and since meteorological conditions in August 2013 were more favorable to photochemical processes

What "meteorological conditions" do the authors refer to here? If it is just the higher solar radiation, then state this in the text.

P15, L17:

"Acetone is the OVOC of the selection with generally the highest atmospheric lifetime, considering its photochemical reaction rate ..."

Unclear, suggest changing to: "Of the measured OVOCs, acetone has the longest atmospheric lifetime, considering its photochemical reaction rate ..."

P15, L22:

"Acetone showed similar seasonal variations than formaldehyde and acetaldehyde, ..."

Replace with "to": "Acetone showed similar seasonal variations to formaldehyde and acetaldehyde, ..."

P15, L25:

"... remained significantly high during winter ..."

Significant to what? Either include a parameter or remove "significantly" from the sentence.

P15, L28:

"... than summer 2013 one, ..."

Remove “one”, it’s not required here.

P15, L30:

“... than winter 2014 one, ...”

Remove “one”, it’s not required here.

P15, L30:

“... but admitted low enough to allow advection to the receptor site ...”

Delete admitted”, not needed here.

P16, L2:

“... from other OVOC ones ...”

Remove “ones”, it’s not required here.

P16, L2:

“Indeed, MEK concentrations did not show seasonal variations except an increasing winter trend ...”

Increased concentrations in winter sounds very much like a seasonal variation. From figure 7, it appears that there is a weak seasonal cycle during 2013, but this is not replicated (or at least not so clear) in the other years.

P16, L6:

“... in February 2013 was by 0.2 $\mu\text{g m}^{-3}$ higher than ...”

Remove “by”, it’s not required here.

3.4.4 Comparisons with other VOC measurements performed at Ersa

I don’t think this section is needed as it doesn’t say a lot. Perhaps the link to the supplementary material could be included in one of the earlier discussion sections.

3.5 VOC factorial analysis

Perhaps some further explanation of the reasoning behind choosing a subset of the measured compounds is required here? Along with a discussion of whether limiting the number of species that are included in the PMF analyses may well affect the result and the number of factors. Perhaps a discussion of how the results here compare to the shorter, intensive campaign results published earlier would help here?

P16, L27:

“... should rather be explained as aged profiles originating from several sources assimilating to several source categories ...”

Should this be: “... should rather be explained as aged profiles originating from several source regions comprising several source categories ...”?

3.5.1 Biogenic source (factor 1)

P17, L6:

“The relative load of this VOC for the factor 1 is 70%.”

Clarify what is meant by this statement.

P17, L6:

“This latter is mainly consisted of primary anthropogenic ...”

What is meant by “This latter”?

3.5.2 Short-lived anthropogenic sources (factor 2)

The description of Factor2 and its influences is rather vague and contains a number of potential contributing sources. This is a result of this type of analysis, but the authors need to be wary of making contradicting statements, for example describing “slightly higher contributions during fall” (P17, L27), then “factor 2 contributions were also significant in spring and summer” (P17, L32) and then “mean monthly factor 2 contributions (Fig. 10b2) pointed out no clear seasonal variation over the study period” I think this is due to the differences observed between different years and so care should be taken not to generalise here.

P17, L6:

“... with an average contribution to the sum of measured VOC concentrations from this factor of 66%.”

Is this correct? Looking at figure 9(b), factor 2 does not appear to ever be 66% of the total.

P17, L31:

“... winter, conducting to less dilution of emissions, ...”

suggest changing to “leading to”

P17, L32:

“However, factor 2 contributions were also significant in spring and summer ...”

This only appears to be the case in 2013.

P18, L1:

“... which could illustrate an enhanced evaporation of gasoline, solvent inks, paints and additional applications during these months as a result of higher temperatures.”

This is contradicted by the temperature data shown in figure 4(b1) which shows lower temperatures in June 2013 compared to 2012 and 2014 which have smaller factor 2 contributions shown in figure 10(b2). The authors go on to give explanation of these differences, but I feel it's important to highlight this anomaly here.

3.5.5 Regional background (factor 5)

P20, L17:

“... probably related to photochemical decay and dilution processes.”

Earlier in this section the authors state that natural gas may be an important source for factor 5 so presumably a summer decrease in emissions may also contribute to the observed seasonal variation?

P20, L21:

“Mean factor 5 contributions in function of air mass origins were in the same range, except that more elevated contributions were noticed under the influence of European air masses (especially those potentially connected to distant contributions; Fig. 11) compared to the ones related to others continental origins.”

This is a confusing sentence; can it be re-written for improved clarity?

4.1 Determination of controlling factors

This whole section appears to re-cap the information given in section 3.5. In order to reduce the size of the manuscript, I would suggest these sections be combined to give a more concise explanation of the observations at the site. This could be by either including extra information in section 3.5 (and removing section 4

P22, L5

“... favouring phenomena of vertical dispersion.”

Delete “phenomena of”, not required here: “... favouring vertical dispersion.”

4.2 The particular case of winter

Figure 13, referred to in the text needs further explanation and a legend describing the colour scheme and the meaning of C1 – C5. These are described elsewhere, but should be included again here in the figure.

P24, L2:

“... compared to winter 2013 ones ...”

Remove “ones”, it’s not required here.

P24, L34:

“As a consequence, this finding also point out that shorter observation periods (i.e., up to two months) may be reflected the variability of the identified parameters under the specific meteorological conditions of the studied period.”

Sentence is poorly written and doesn’t make sense, needs to be re-written for clarity.

5. VOC concentration variations in continental Europe

Figure 15 is referred to in the text. The ERSA site should be highlighted in the caption to identify the station under study here.

P25, L17:

“... observed in most continental Europe ...”

“... observed in most of continental Europe ...”

P25, L20:

“... were globally lower and ...”

not globally, but European wide

P25, L23:

“... suggesting a high importance of photochemistry processes and vertical dispersion phenomena in regulating concentration levels.”

I would suggest that temperature (linked to boundary layer height) is the main driver here. As the authors state earlier in the manuscript, the majority of these compounds (with the exception of ethylene) have relatively long lifetimes and so photochemistry will likely be limited.

P26, L5:

“Then, at stations located ...”

Delete “then”

P26, L22:

to normal values

How do the authors conclude which is “normal”?

P27, L6:

“Then, VOC concentrations ...”

Delete “then”

P27, L11:

Sentence containing “... was not as warmer-than-average as ...” is poorly written, please re-write for clarity

6. Conclusions

This section is far too long and needs to be re-written more concisely.