

acp-2016-185: Seasonal variability and source apportionment of volatile organic compounds (VOCs) in the Paris megacity (France)

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General/scientific comments

This paper reports the results of a source apportionment by positive matrix factorisation (PMF) of the concentrations of a suite of ambient VOCs measured in urban background air in Paris over a period of several months from January to November 2010. VOCs were measured both by on-line GC and by PTR-MS. In order to help in the assignment of some of the factors emanating from the application of PMF, the authors compared the speciated VOC profiles of the factors with speciated VOC profiles the authors separately measured at three locations where they assume that a single emission source will dominate the ambient VOC, specifically: (1) measurements during busy (and traffic jam) periods in a highway tunnel, to represent vehicle related VOC emissions; (2) measurements close to a domestic gas flue, to represent natural gas source; and (3) measurements at a fireplace facility, to represent residential wood-burning emissions (the authors acknowledge that their measured VOC profile from this source may be less quantitative than for other source profiles). To further assist in the assignment of PMF factors to particular VOC emission sources the authors also make use of additional co-located atmospheric compositional data available to them, such as NO, CO and black carbon (BC).

The authors present results in which the ambient urban background VOC has been apportioned into six sources, each of which has been assigned an identification, albeit that one factor is assigned to be a mixed natural gas/background source. The analysis presented includes speciated VOC profiles for each factor and monthly and average-hourly variation in the absolute contribution ($\mu\text{g}/\text{m}^3$) of each of the six identified factors/sources. The largest contributions in total are from traffic-related activities (through two identified factors: motor-vehicle exhaust, and gasoline evaporation), although all six identified sources have not that dissimilar relative contributions, on average. A noteworthy observation is significant contributions to ambient VOC from wood burning, 18% on average but up to ~50% at times in winter. Biogenic emissions were also reported to be significant, 15% on average but more in summer.

The authors have a large dataset of time-resolved speciated VOC over an extended time period, almost a year. They have used standard, but appropriate, statistical methods to endeavour to decompose the ambient measurements into individual source contributions. These are statistical, rather than dispersion-/chemistry-based source apportionments. These methods have been widely employed to apportion ambient PM, but less so for VOC.

The presentation of results and their discussion are largely descriptive, in the sense that the authors present the details for their factor/source contributions and their monthly and hourly variations, which are rationalised with general text about anticipated behaviour of particular sets of pollutant mixtures in the urban background atmosphere. The authors also present a qualitative and quantitative comparison of their VOC source apportionment with previous literature.

Although in one sense the presentation of this work could be described as 'formulaic' – following previous data presentation and analysis styles – nonetheless the large dataset presented here for a large European city presents a valuable addition to the VOC apportionment literature. A particular feature is the presentation of VOC speciated profiles for three potential VOC sources, although the authors acknowledge some shortcomings in these. The paper is already lengthy and contains much new data, supported by detailed descriptions of the data collection and processing protocols and results. As an overall summary, the content of the paper is suitable for publication in ACP.

Technical comments

The tables and figures are generally clearly presented. The written text is largely unambiguous in conveying its meaning, but it is overly long in places. There are instances where introductory sentences to a section could be substantially abbreviated, or even deleted as repeating what the reader will have picked up from the methods section. The authors should be encouraged to edit text further for conciseness of expression.

The following are more specific comments.

P6, L18: should read “Raw data were corrected using...”?

P6, L24: delete “is” before “analyzer”

P6, L32: delete “at”

P7, L22: should read “except for”

P8, L9: I don’t understand what is being described in the sentence beginning “Finally, these different processings...” Please rephrase this sentence to make clear to what “it” is referring in this sentence and to clarify what the procedure undertaken was.

P10, L26: the phrase “fairly comparable” is not scientifically precise.

P10, L29: “a few flurries” of what? Please write specific statements about the nature of the weather.

P11, L16: the phrase “contribute to the tune of” is too colloquial; please use more direct wording.

P11, L30-L35: there are several instances in these lines of text of negative values for VOC concentrations. These are surely some aberration (albeit repeated aberration) of typing error. Please correct.

P13, L3: I do not understand the scientific sense of the sentence starting “With an atmospheric residence time...” How does the statement at the end of this sentence (about methanol emissions contributing to background levels) derive from, or otherwise relate to, the text at the start of the sentence about methanol residence time? Please reword to clarify.

P14, L24: I do not follow the scientific logic here. The text appears to state that iso-pentane is known to be a key tracer for gasoline evaporation, but also to say that iso-pentane was not present in the speciated profile the authors have assigned in their work to gasoline evaporation.

P14, L32: please explain more clearly what about the monthly change “remains ambiguous”.

P20, L19: The phrasing that the mean temperature was “in the range +/- 20 degC” does not make sense. Either quote the range, or quote the mean and some recognised statistic of the variation about the mean. Likewise for later in this sentence in connection with “+/- 16 degC”.

Caption of Table 1: It would be helpful for the caption to remind the reader with a statement of the time resolution of the raw data from which these statistical summaries are derived, and of the time duration/dates of the total dataset.