Atmos. Chem. Phys. Discuss.,
doi:10.5194/acp-2017-90-RC3, 2017

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Exploring sources of biogenic secondary organic aerosol compounds using chemical analysis and the FLEXPART model" *by* Johan Martinsson et al.

Anonymous Referee #3

Received and published: 27 March 2017

Review of Martinsson et al., "Exploring sources of biogenic secondary organic aerosol compounds using chemical analysis and the FLEXPART model"

Synopsis

In this work, the abundance and time variability of molecular compounds identified in filter samples of ambient air measured at the Vavihill site in Sweden is investigated. The authors then use a numerical Lagrangian particle dispersion model (FLEXPART) and statistical analysis (PCA) to identify source regions for the different compounds they have identified. They find that coniferous forests contribute strongly to monoterpene tracers found in organic aerosol samples.

General comments

The authors attempt to use backtrajectory calculations to qualitatively connect exposure of an air mass to land surface types to molecular markers found in organic aerosol samples, thereby investigating their formation formation processes. This is a nice and straightforward idea. The authors struggle, however, to convince the reviewer that their analysis and modeling has been conducted in a knowledgeable way. Their use of beta release software without justification, plus a number of other vaguenesses in the methods description are a warning sign that considerable caution should be taken before this manuscript can be published. Apart from questions regarding the methods used, the final results of the analysis don't seem to provide much new information. The fact that coniferous forest emits monoterpenes which then forms SOA has been shown numerous times. If there are other important findings, they are not apparent to the reviewer.

In summary I am tempted to recommend rejecting the manuscript due to the deficiencies in the methods section and the lack of scientifically new findings. It should be noted that in my review I cannot comment on aspects of the chemical analysis, as this is not my field of expertise. Hence I will recommend 'major revisions' here, as the chemical analysis might contain information that is novel for other readers.

Specific comments

* Use of beta software in analysis

The current stable release version of FLEXPART is 9.02, while the authors (claim to?) use version 10.0. Software in beta versions is considered unstable and for testing purposes only and can surely not be used in a scientific publication.

* HYSPLIT and FLEXPART together

It is unclear why simulations using the HYSPLIT model are 1) done at all and 2) presented as auxiliary analysis which is different from the FLEXPART analysis. Both HYS-

PLIT and FLEXPART solve the transport equations backwards in time. HYSPLIT as used here calculates single, deterministic trajectories, while FLEXPART calculates a large number (100000 in the present case) of trajectories, applying processes like turbulence and convection stochastically. FLEXPART by default delivers mass-weighted center trajectories and clusters (see documentation), which provide information equivalent to HYSPLIT. There is no additional information gained from the use of the HYSPLIT model, unless the authors start and compare the model results in detail. I recommend removing this completely, at most leaving a sentence stating that they evaluated HYSPLIT and it gave similar results.

* Uncertainties due to neglected sources and sinks during transport

On several occasions the authors caution that what they are doing is neither a full source inversion, nor a modeling effort considering (non-linear) effects of chemistry and other sources and sinks in the atmosphere. Statements like : “a formal source apportionment would require precise accounting of these factors, which is extremely complicated and is clearly out of scope of this study” (p 5, l 14-15) leave the reader wondering what this study is about, then, as more then half of the manuscript deals with exactly this kind of analysis on a simple level. This leaves the reader with the uneasy feeling that he/she cannot attribute significance to the findings. How large are those uncertainties? Where do they come from?

* “Surface vertical level” method

The “surface vertical level” definition as half the PBL height for each particle is not a standard FLEXPART output product that I am aware of (at least not in v 9.02). There is no documentation of this feature judging by a cursory look over the available publication (Stohl et al., 2005, ACP) and a quick source code survey. While surely useful, I don’t see how the authors have achieved this without coding it themselves. This would have to be described accordingly, if this is what they did. Furthermore, the choice of 1/2 the PBLH is arbitrary, and the reasoning (“non-linear processes” again) is insufficient.

* AERO-Tracer

Justify the use of the particle diameter used, as this has considerable effect on the lifetime of the particles and hence the exposure calculation. I suggest recalculating for large and small particles.

* source apportionment

Given the description on page 6 (top paragraph), you are simply multiplying the response function output fields (units of s m3 kg-1) by the fractional land cover - did you correct for grid area and level thickness?

* Principal component analysis (PCA) method

The method description of the PCA is insufficient. Citing a commercial, non-free software package is not an appropriate source of information for the reader.

Also: for a PCA to be meaningful, a number of preconditions have to be met, out of which I wonder if two are met: 1) sample size: 38 data points (filter samples) is quite small, can you show that the results are still reasonable? 2) outliers: did you remove them?

* PCA results

The kind of PCA performed should be described in the methods section, see above.

Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-90, 2017.