Atmos. Chem. Phys. Discuss., 12, C12590–C12592, 2013
www.atmos-chem-phys-discuss.net/12/C12590/2013/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Estimation of volatile organic compound emissions for Europe using data assimilation" *by* M. R. Koohkan et al.

Anonymous Referee #2

Received and published: 12 February 2013

Summary.

The authors present an adjoint inversion of European VOC emissions based on an annual cycle of concentration measurements at a network of sites. The mathematical framework for the analysis is thoughtfully presented, and the paper provides an interesting comparison of results obtained using varying statistical assumptions in the inversion. The application is fairly novel, in the sense that there have not been many inverse studies of speciated VOC emissions based on this type of surface network. The topic is appropriate to ACP.

While the statistical assumptions going into the solution are well thought out, the phys-

ical assumptions are less so. My major comments relate to this issue. If these are addressed I recommend publication in ACP.

Scientific comments.

1. The authors examine how their inferred fluxes change with different assumptions regarding the error covariance matrices, etc., which is great. However, they do not test their sensitivity to forward model error, which can often be a larger source of error. How sensitive are your inferred fluxes to uncertainty in a) model transport, b) model OH, c) boundary layer height, and d) boundary conditions? Currently I think this is the biggest shortcoming of the paper.

2. A related point is that the authors do not provide any uncertainty range on their inferred fluxes. That should be fixed. Even if it is difficult to fully characterize the a posteriori uncertainties, some attempt should be made. For instance, do you regard the range between the Case B1, B2 and C fluxes in Table 6 to provide a reasonable estimate of the true uncertainty in the solution?

3. Figure 5 shows that the B2 optimization does not significantly change the OH fields. What about the B1 optimization, does that change OH to any significant degree? Since this is the only scenario where isoprene emissions change appreciably, this is where one might expect to see a notable OH change. (also, just because the OH concentration doesn't change much with the inversion doesn't mean it was accurate in the first place, see comment 1).

4. On page 33238, you make the point that the inversion does not work very well for isoprene because of its short lifetime. Does the surface network provide measurements of MVK and MACR, and if it did would you expect this to help to any significant degree?

5. What do we learn from the inversions that gets at some improved knowledge of process? Are there specific source sectors or regions that appear to warrant specific investigation? What recommendations would you make to inventory developers?

Technical / editorial comments.

1. The first several pages are taken up with mathematical underpinnings of chemical transport modeling and adjoint analysis, much of which is not new. As a suggestion, this section could be shortened by referring to previously-published work.

2. Top of page 33235, ug/m3 is an odd unit for OH, O3, and NO3.

3. Throughout, suggest spelling out the compound names (and/or formulae) rather than using abbreviations, which are not defined in the text. While they are defined in a table, readers might just assume “ACE” refers to acetone or acetaldehyde (for example) rather than acetylene.

Figure 7, consider putting the species names inset in the panels rather than “a”, “b”, etc.? It would make for easier reading.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 33219, 2012.