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Atmospheric
Chemistry
and Physics
Discussions

Interactive comment on "Source-receptor relationships for airborne measurements of CO_2, CO and O_3 above Siberia: a cluster-based approach" *by* J.-D. Paris et al.

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We thank the reviewer for his comments and suggestions for improvements. The main question was on the comparative advantage of clustering of "footprints" based on Lagrangian particle dispersion modelling (LPDM), with respect to forward model sensitivity analysis. It is not entirely clear to us whether the main critique of the reviewer is on the backward simulations with the LPDM or on the cluster analysis of the LPDM output, or both. We therefore provide a line of arguments that shall clearly show the advantages of the new method.

1. Tagged tracer simulations versus backward simulations

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Regarding the issue of tagged tracer simulations versus backward simulations we fully agree that tagged tracer simulations have some attractive features. The most attractive one was actually not mentioned by the reviewer: They can account for non-linear chemical reactions. However, tagged tracer simulations also have two important disadvantages.

Firstly, their limited resolution makes a comparison with in situ measurements often difficult. In contrast, an LPDM can be run backward directly from a measurement point and can in principle resolve structures of any scale and contribute to interpret the vertically layered structure of tracers. Our Lagrangian model captures features present in the high temporal resolution dataset such as sharp layers and frontal redistribution of trace gases, which were encountered on several occasions through the campaigns. Instead, an Eulerian model (also used for this project's data analysis; LMDZ-INCA) was found to be rather applicable to the study of large scale gradients (Paris et al., 2008, Bull Am Met Soc, submitted), which are not within the scope of this paper.

Secondly, with tagged tracer simulations, regions and source types have to be predefined and only a very limited number of tagged tracers can normally be handled. In contrast, with the LPDM calculations in backward mode regions and source types do not have to be pre-specified. In fact, the model is run even without any source information. Backward runs allow propagating more objectively the information from detector to region of origin.

For carrying out tagged tracer runs, as the reviewer indicates, one needs additional information on fires, as well as other biogenic and anthropogenic emissions. The uncertainties associated to these emissions are propagated to the relative source attribution of the tagged tracer. For CO2 where the fluxes are widespread, highly variable and uncertain, this approach will not provide any result. For O3, similarly, which has no surface emissions, tagged forward runs cannot be used, whereas backward runs and clustering can attribute rather simply observed variability to coherent regions or processes. For most trace gases, evaluating the agreement between modelled tagged tracers and observed concentrations is tricky, and may lead to biased interpretation. For instance, the model fit with observations may look satisfactory whereas in fact a small bias might reflects a large error on the tracer origin.

On the opposite, with LPDM, source contributions can then be determined for individual grid cells and source types of an emission inventory, at almost any resolution required. Thus, the LPDM calculation provides more information than even a forward tracer simulation with many thousand tracers. Another advantage is that emission information can be exchanged or updated in the post-processing without having to re-run the model. Thus, for a linear problem, a backward LPDM calculation is certainly the most powerful (and also more elegant) method to identify potential sources. It is more similar to an adjoint simulation with a tracer model. For non-linear problems, the situation is different and there a tagged tracer simulation may be more appropriate.

2. Clustering LPDM output

Regarding the issue of clustering the model output, as noted above the information contained in the model output is huge, equivalent to many thousand tagged tracers! Now, we argue that the sources (and sinks) of most trace substances over Siberia are not well known. There is a large uncertainty on the amplitude (Gurney et al., 2002) and distribution of the Siberian biospheric sources and sinks due to the sparseness of measurements network in Boreal Asia. Fire (either agricultural burnings or wildfires) primary emissions are not well known for Siberian sources (limited amount of data, see e.g. Andreae and Merlet, 2001) and therefore secondary species like ozone photochemically produced in the plumes are difficult to model. Anthropogenic emissions are also not well known. Therefore a method that clearly separate transport classification from actual emissions is useful for the matter at hand.

One method to improve our knowledge of the source distribution is inverse modelling. Such a method using the FLEXPART model output has recently been presented (Stohl et al., 2009). However, this method is complex and directed only towards improving emission estimates. It does not necessarily lead to improved knowledge which sources impact which regions, etc. From the measurement perspective, one often wants to have a more simple way of classifying the data, and cluster analysis is an established method for doing this efficiently. It has been used in many studies for clustering trajectories, and it is still being used by many. One could argue that all these studies are also not necessary because tagged tracer simulations give the same information, which is not entirely true as shown above. Our paper follows the tradition of these trajectory studies. The innovation here is that our study uses the LPDM output which (1) is more quantitative than just trajectory positions and (2) also includes effects of turbulence and convection, which normal trajectories ignore. We emphasize that the clustering is done based on the emission sensitivity, not on source contributions or tracer concentrations. Thus, the method maximizes differences in transport from different regions.

In summary, we think that both tagged tracer simulations and methods like the one presented here are useful for improving our understanding of atmospheric transport and chemistry. Each approach has its own strengths and weaknesses. It would be dangerous to eliminate the diversity of science by claiming that a given problem should be solved only with one particular method because this method has proven to be useful. It may be useful (tagged tracer simulations are) but it may also not be perfect (tagged tracer simulations aren't) and exploring alternatives (such as the approach described in our paper) will eventually lead to better ways of solving the problem.

We finally feel that this method fills a methodological gap between the clustering of back-trajectories (which are not quantitative enough) and the spatially explicit potential emission sensitivity provided by LPDMs (for which no objective classification criteria existed). More quantitative applications are not developed in this paper but are feasible and foreseen to be implemented in upcoming work. It should be noted also that this method has a large potential for very large datasets like the MOZAIC regular aircraft programme where a large-scale tagged Eulerian study would be difficult to implement and to interpret.

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3. Minor comments

The reviewer's comments are reproduced here and addressed one by one, with reply marked by a star.

. . .It would be appropriate in the introduction to refer to some of the earlier measurement programs that provided atmospheric greenhouse gas data over Siberia, e.g. work by Lloyd et al., Tellus 2002, Levin et al., Tellus 2002, and others.

* These references and others have been included in the revised ms

Throughout the manuscript, I suggest to replace the word "aircraft itinerary" by "flight track"

* We agree with this suggestion and will implement it in the revised manuscript

Pg 2, ln 20-26: This paragraph would fit better at the end of the introduction

* We agree with this suggestion and will implement it in the revised manuscript

Pg 2, ln 23: "tomography" suggests three-dimensional domain filling observations, which is certainly not the case for airborne profile measurements using in-situ techniques.

* We change the wording to "extensive sampling" in the revised ms

Pg 3, ln 16: sentence not complete

* If not mistaken the sentence starting with "Pollutant export to or across Siberia. . ." seems to be complete.

Pg 5, ln 22: When precision and accuracy are both 0.15ppm, there is no room for any short-term noise in the measurement

* We agree with the remark of the reviewer as far as this airborne experiment is concerned. The LiCor measurement cell itself has a strong short term noise. The careful design of pressure, flow and temperature control reduces random error, and regular in-

flight calibrations reduce systematic error down to equivalent precision and accuracy of 0.15 ppm.

Pg 6, ln 6: “prior assessment of transport”: this remains unclear. What is the outcome of this assessment?

* FLEXPART was run both based on NCEP and ECMWF fields. When the two runs tend to disagree strongly for a particular ‘retro-plume’, we have less confidence in this ‘retro-plume’ and discard it from further detailed analysis. For the clustering analysis we have selected flight where there was enough confidence in LPDM to reject no ‘retro-plume’. This can be checked on the YAK-AEROSIB FLEXPART results webpage : http://zardoz.nilu.no/∼andreas/YAK/

Pg 6, ln 16: It is unclear which particles are declared as having stratospheric origin. Those that have been in the stratosphere during part of the ten days, or only those that were in the stratosphere 10 days prior to measurement?

* Particles that resided in the stratosphere during any part of the 10 days are declared as having stratospheric origin. This will be made more unambiguous in the revised ms

Pg 9, ln 3-6: Which of the 20 versions was used? Or how have those runs been combined?

* From the 20 realizations of the algorithm (i.e. with 20 distinct initial, random position of clusters’ centroids), the one that yields the better clustering separation (efficiency according to the clusters’ metrics) is recorded for later use, and the other are discarded. The number 20 was chosen as the smaller number that ensures reproducibility of the algorithm on this particular dataset.

Figures 5,7, and 9 do not contain a proper legend for the color scale. Is it the natural log or base 10? What units where used before applying the log?

* The unit is second (of residence time per grid element) to which we apply a log10. We will correct the figures to include a clear colorbar in the revised ms.

Pg 14, ln 11: “: : : excluding data associated to residence time < 100 s/grid”: how does this threshold compare to the range of residence times?

* With this criterion we reject less than 1% of the dynamic range of residence time (see e.g. Fig. 12).

Pg 14, ln 26: replace “seem to has” with “seems to have”

* This is corrected in the revised ms

Pg 15, ln 12: “excepted” –> “excect”?

* The word is appropriate

Pg 15, ln 27: “: : : a novel application of clustering to Lagrangian particle dispersion model footprint” this should be reformulated

* According to suggestion we reformulated this sentence into: “. . . a novel technique based on the clustering of LPDM footprints” in the revised ms

Fig. 12, legend: I would suggest to use month&year throughout the paper instead of campaign names (YAK1 etc).

* We will retain the suggestion as this will improve clarity for the reader. The references to the campaigns will make explicit reference to the month and year of flight in the revised ms.

References :

Stohl, A., P. Seibert, J. Arduini, S. Eckhardt, P. Fraser, B. R. Greally, C. Lunder, M. Maione, J. Mühle, S. O’Doherty, R. G. Prinn, S. Reimann, T. Saito, N. Schmidbauer, P.G. Simmonds, M. K. Vollmer, R. F. Weiss, and Y. Yokouchi, 2009: An analytical inversion method for determining regional and global emissions of greenhouse gases: Sensitivity studies and application to halocarbons. Atmos. Chem. Phys. 9, 1597-1620.

Gurney, K. et al., 2002: Towards robust regional estimates of CO2 sources and sinks using atmospheric transport models, Nature 415, 626-630

Andreae, M. O. and Merlet, P.: Emission of trace gases and aerosols from biomass burning, Global Biogeochem. Cycles, 15, 955–966, 2001.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 6207, 2009.

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