

Interactive comment on “Source-receptor matrix calculation with a Lagrangian particle dispersion model in backward mode” by P. Seibert and A. Frank

Anonymous Referee #2

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Overview

The paper presents and 'introduces' (but see below) a methodology to determine source-receptor relationships for the transport and diffusion of atmospheric trace substances. The methodology is outlined and a number of examples provided. The paper is well written and structured and as a whole certainly deserves publication. However, there are two main issues that should be addressed and put into perspective.

Main Comments

1) What is presented as being new ('the method introduced in this paper') is, in fact, not different from footprint modeling as practiced in Boundary Layer Meteorology (and

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in particular: Micrometeorology) for more than tow decades now. In their 'Outlook' the authors even mention this, but refer only to 'flux footprints'. Many available footprint models (and especially Lagrangian models) allow for calculating flux and concentration footprints (e.g. Kljun et al. 2002). First of all this should be stated already in the introduction and the approach should be put into perspective. A recent review can be found in Schmid (2002) [If the authors prefer the 'sr-relationship' as a name that is their choice, of course]. Now, what is indeed new in the present paper is the application and adaptation of the concept to much larger scales and hence a frame of reference within a gridded regional modeling system. This also then allows for attribution of quantitative (not only qualitative) source-receptor relations [note that a footprint function does yield this quantitative information too. However, it is often difficult to deal with small spatial differences between model and observation. If all is averaged over a grid box this seems to be much more convenient].

2) My major concern with the approach presented by the authors concerns the treatment of 'linear processes' acting on the particles. In fact, I find the approach of treating the particles as having a mixing ratio rather than mass attributed, and to allow linear processes acting on the particles too, quite attractive (and it is, according to my knowledge, quite new - what could be stated by the authors). However, there are a few caveats as well with this method.

- First, Lagrangian particle dispersion models are designed with the help of the so-called well-mixed condition according to Thomson (1987). Any model incorporating an additional linear process cannot fulfill the well-mixed condition (wmc), therefore we are lacking a powerful tool in designing the model (in principle). It is clear, that for the time being (i.e., no extended criterion is available to design a model in cases with superposed linear process) it is a reasonable approach to take a model that fulfills the wmc if no linear processes were acting and assuming that the model is useful even with a superimposed linear process. But this should be stated.

- Second, the phrase "Lagrangian particle dispersion models cannot simulate non-

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linear chemical reactionsŠ implies that they can do linear reactions. Now, this is Ű in the framework of the approach of the authors Ű only true if there is no other species involved (or this other one is largely abundant as, say, Oxygen). Hence within air pollution modeling, only photo-dissociation reactions qualify. Otherwise, eq. (1) would contain another concentration (). This then would mean that the particles would have to be ŠcoloredŠ and in order to evaluate the rate of change in the mixing ratio of particles one would have to know the ensemble of the distribution of .

-Finally, there is a principal problem with particles (i.e., in Lagrangian particle models) and chemical reactions in that the former, upon a chemical reaction, will change their properties (molecular weight, shape, etc.), but before and after the reaction they are assumed to behave as fluid elements (which is an idealization of course). This may have prevented some people to use particle models in connection with problems other than passive tracer studies and also might be stated by the authors.

Specific comments

I list a number of minor comments and identify them with their line and page by counting all lines including equations, titles etc. Šc1Š refers to the left column on a page, and Šc2Š accordingly to the right one.

p.3, l. 7ff, c1 Š t' < tŠ: I understand that in the authorŠs implementation, time runs backwards (negative time increment). Therefore, and if tŠ is the variable of the integral and t a specific value, when a particle arrives at time t, any time tŠ between tŠ=0 and tŠ=t will be larger (or equal) than t.

p.3, l.23, c1 ŠŠif the averaging time exceedsŠŠ: also horizontal homogeneity must be required for what follows.

p.4, l. 1, c2 ŠŠdue to the limited number of trajectoriesŠŠ. A good reference to kernel methods would be De Haan (1999).

p.6, l.13, c1 ŠŠdo not simulate the effects of convectionŠŠ. This is probably not true for

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ŠmostŠ of Lagrangian models. For example the models of Luhar and Britter (1989), Weil (1994), Rotach et al (1996) etc. do specifically take into account convection.

Table 2 Number of particles: 1000. This is quite a small number (for a particle model). The authors should comment on this (in connection with the kernel concentration treatment, probably).

p.7, l.. 5, c1 Wet scavenging acts on all particles regardless of their height: a little more information on how this process is parameterized would be desirable.

p.9, last l., c1 ŠĖappears to be sufficient to explain the observed CaesiumĖ.Š This statement should be made more precise. Has any (statistical) test been applied in order to support this statement? What is the resolution of the observations, how many data points etc.? [looking at Fig. 4, for many of the days it appears that both forward and backward simulations yield quite a different value than obs +/- error bars]. Similar: column 2: ŠĖ. Is not likely the sourceŠ: can the authors be a little more quantitative?

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