1. INTRODUCTION AND MOTIVATION

Any dispersion model (and even any trajectory model) effectively calculates source-receptor relationships. However, usually these models read in specified source terms (emission rates), and produce fields of concentration as output. The information which part of the source contributes to a specific receptor element (e.g., concentration in a certain grid cell during a certain time interval) is not available in this approach. In inverse modelling, it becomes necessary to consider this information because the inversion consists of finding the value of each source element which leads to the minimum of a cost function measuring the misfit between model output and observations. However, inverse modelling is not the only application of explicit calculation of source-receptor matrices (SRM – definition see below). It could also be used to describe the relative importance of specific subsets of the source (e.g., emissions of certain region or during a certain time) for the impact at a certain receptor site. For linear source-receptor relationships, once the SRM has been calculated different emission scenarios can be used to produce receptor concentrations by a simple (but potentially large!) matrix-vector multiplication. This paper presents some examples of applications, namely inverse modelling of the ETEX-1 tracer release, and work connected to the verification of the Comprehensive Nuclear Test Ban Treaty CTBT.

SRMs can be derived by any method – forward or backward (adjoint), with Eulerian or Lagrangian models. The approach which we have chosen (back-ward with an Lagrangian particle dispersion model [LPDM]) combines the following assets:

- receptor-oriented (backward) – much more efficient for cases with number of receptor elements << number of (potential) source elements
- LPDM – no problems with numerical diffusion
• can handle point measurements (backward mode) without sub-models or artificial mixing when handing over (pseudo-)tracer to grid cells

2. CALCULATION OF THE SOURCE-RECEPTOR RELATIONSHIP

2.1 Definition of the source-receptor relationship

The source-receptor relationship \( m_{il} \) is defined as

\[
m_{il} = \frac{\partial y_i}{\partial x_i}
\]

where \( x_i \) is a source occupying a certain three-dimensional space and time interval, and \( y_i \) is a receptor. The receptor can also be any subset of the 4-dimensional space, but typically it will be a point in space (the sampling location) and a certain interval in time (the sampling period). The dimension of \( y \) is kg m\(^{-3} \) (or Bq m\(^{-3} \)) and that of \( x \) is kg (Bq).

For linear source-receptor relationships, \( m_{il} \) is a constant and \( \frac{\partial y_i}{\partial x_i} \) can be replaced by \( y_i / x_i \). Therefore we speak about the source-receptor matrix (SRM) \( M \) with elements \( m_{il} \). This is the case discussed in this paper. It is relevant for the dispersion of inert tracers and of substances with a prescribed decay (or growth) rate.

\[\text{2.2 SRM calculation with a backward-running LPDM}\]

While running the LPDM forward to produce the SRM would have been straight-forward, the backward approach is novel. There is some previous work with backward-running Lagrangian models, but it was not yet exploited for the formal determination of a SRM. What comes closest is the micrometeorological work of Flesch and Wilson (1995).

There appears to be a conceptual problem with running LPDMs backward because the generation of particles in emission areas is not simply reversible and also nothing for which an adjoint can be constructed. Sometimes, people argue that diffusion as an irreversible processes cannot be present in a backward-mode simulation. This view is only caused by a misunderstanding of what a backward or adjoint simulation really is doing. The backward simulation does not trace backwards the same particles that some forward simulation has used. The forward simulation establishes the SRM between a single source element and many receptors. The backward simulation establishes the SRM between a single receptor and many source elements. For a global coverage mapping a whole (sub-)space to itself, many forward or backward simulations (or one simulation with many species each representing one source or receptor) must be made. Then, the SRMs of both should be identical, apart from numerical errors. To determine a SRM element, forward simulation runs with a certain source and obtains \( m_{il} \) by applying the definition given above. The backward simulation uses something that can be called a pseudotracer – it is only a mathematical quantity, not a real
concentration – and directly evaluates the SRM elements, or ‘influence functions’ as people sometimes put it.

In the following, a short description of the method is given (see also Seibert and Stohl (2000) and Seibert (2001)). For better understanding, let us begin with a characterisation of the usual forward application of the LPDM:

- particles released from prescribed source areas, transported with mean and turbulent velocity field.
- carry mass depending on source strength and particle release rate
- mass can be altered by processes such as deposition or decay
- concentrations: summing up masses of all the particles in a grid cell, divided by its volume.

One performs a run for each source element (or treats each source element as a different species) and collects all the relevant ambient concentrations. For the linear case, the ratio of the two would give the SRM elements.

For the backward simulation, it can be shown that in the case without sinks the SRM elements are simply equal to the residence time spent in the respective source grid cell by particles released from the receptor (Seibert and Stohl, 2000; Seibert, 2001).

The key assumptions made to arrive at this result are

1. only volume sources are considered and no area, line or point sources, and
2. particles carry mixing ratios rather than masses, and these mixing ratios are changed by the volume sources and eventually other processes as represented in the LPDM (scavenging, dry deposition, decay, ...).

To give an intuitive explanation: We consider all particles arriving during the measurement interval at the receptor. We trace them backwards taking into account mean and turbulent motions. Along each trajectory we can integrate the Lagrangian budget equation for the mixing ratio, which means that the mixing ratio at arrival of the trajectory is the sum of the accumulated emissions into this particle, or

\[ \int x(r(t),t) \, dt \]

where \( x \) is the emission rate (here as change of mixing ratio per volume and time) and \( r \) is the location. The mean over the measurement interval gives us the desired average over time and, at the same time, the ensemble average that is typical of a LPDM.

The recipe for the SRM determination in the backward mode thus is:

- use same formalism and computer model, but integrate the particle trajectories backward in time, using a negative time step;
- emit a pseudotracer from each measurement site during one measurement interval, with arbitrary (e.g., unit) source strength;
- new interpretation: particle mass \( \rho \chi \) (in an infinitesimal volume of air); and
- calculate gridded ‘concentrations’ of the pseudotracer \( \bar{c}_i \) as usual and scale them as follows to obtain the residence times per grid cell \( \Delta \tau_i \):
\[ \Delta r_i = \bar{c}_i \frac{V_i \Delta T}{\mu_{tot}} \]

where \( V_i \) is the volume of the grid cell, \( \Delta T \) the averaging time interval for the output, and \( \mu_{tot} \) the total emitted mass.

If there are any first-order processes such as, e.g., scavenging along the trajectory, they will be automatically considered if implemented in the LPDM, as they just change either the real mixing ratio (forward mode) or the signal of a normalised emission (backward mode) by a certain percentage that does not depend from the value itself.

3. APPLICATION TO ETEX

3.1 ETEX key features

ETEX is an acronym for the European Tracer Experiment. We are dealing here only with the first of the two releases of a gaseous, nondepositing, inert tracer. Its main characteristics were:

- Release location: Bretagne (NW France)
- Release time: 23 Oct 16:00 UTC – 24 Oct 3:50 UTC, 1994 (12 hrs)
- Release rate: 7.98 g/s (constant)
- Sampling network: 168 over most of western and central Europe
- Sampling resolution: 3 hours
- Sampling duration: 30 intervals (90 h)

3.2 FLEXPART setup for the backward runs

FLEXPART\(^1\) is a LPDM developed by Andreas Stohl, and tested on various tracer experiments (Stohl et al., 1998). It performed well for the ETEX-1 release \((r=0.59, FB=0.01)\). It was used with the following specifications to calculate SRMs in backward mode:

- Input: ECMWF fields \((1^\circ, 31\) levels, 3 h).
- 30 backward runs (one for each sampling interval)
- 10,000 particle released from each monitoring site
- each monitoring site (release* location) is one species
- output sampled with 1 h intervals on a 41x21 grid with 1° resolution, 9 vertical cells \((0/75/150/300/600/1200/2400/3600/4800/6000 \text{ m})\), 105 time intervals.

The size of source-receptor matrix is \((41x21x6x105) \times (168x30)\) or on the order of \(1 \cdot 10^9\) elements (in sparse matrix format, \(\approx 600 \text{ MB of data}\)). This is too big for direct evaluation. The following simplifications were made for the inversion:

- We take only lowest layer (or vertical integral), as the pseudotracer is well mixed near the release site.
- We assume as given either
  \((a)\) location (sample application: nuclear power plant accident); or
  \((b)\) release time (sample application: nuclear explosion with time known from nuclide ratios); or

\(^1\)see http://www.forst.tu-muenchen.de/LST/METEOR/stohl/flexpart.html
the knowledge that we look for a point (i.e., one-grid cell) source. In this case it is possible to invert separately for the temporal evolution in each grid cell, and check for correlation co-efficient or RMSE between observations and model results \( M \cdot \hat{x} \) for the location giving the best fit. We solve the linear system of equations resulting from the linear source-receptor matrix using a simple regularisation. This regularisation requires that the variance of the solution is small and (optionally) that it is also smooth, meaning that the Laplacian of the solution is small (see Seibert (1999) for details).

### 3.3 Results of inversion

Before going into the results of the inversion, Figure 1 shall illustrate in a more tangible way what the backward simulation looks like, and furthermore it is an example of the capabilities of the SRM approach. In this figure, a quantity \( c^\ast \)

\[
c^\ast_i = \sum_{l=1}^{l_{\text{max}}} m_l y_{l,i}
\]

(which may be called a pseudotracer) is plotted. This means, the SRM elements of each observation have been weighted with the observed tracer concentration \( y_{l,i} \) and then been added. The source index \( i \) is decomposed into a spatial distribution (shown on the maps, for the surface layer only) and a temporal sequence, here illustrated as a sequence of maps. What we can see here is very similar to the pictures obtained with so-called trajectory statistics, only that the single mean-flow trajectories have been replaced by sets of random-walk trajectories and that the ‘trajectory statistics’ is time dependent. It is interesting to notice that at the time of the real release, the pseudotracer cloud is rather compact and its centre is near the release point. However, it does by far not pinpoint the release as well as the inversion shown below.

#### 3.3.1 Inversion of Temporal Evolution of the Release

Figure 2 shows results for the determination of the temporal evaluation for given source location with 1-hourly source resolution. The start of the release was reconstructed well, but its end is not well defined and tends to be too late. This is obviously caused by observed non-zero concentrations at a time when the modelled tracer cloud (in a forward run) had already passed the respective sites. The inversion tends to create two separate emission peaks. If the minimisation of the Laplacian (2nd derivative) of the solution (smoothness condition) is added to the regularisation, a more realistic shape of the curve is obtained. A correlation coefficient between observed and modelled concentrations of 0.5 is reached in both cases (compare with 0.6 for forward run with known emission). The total emission is underestimated by 22% for the simple regularisation and 9% if the smoothness condition is added.
3.3.2 Inversion of spatial location

Figures 3 shows results for the determination of the source location with the temporal evolution given. As an effect of the regularisation requiring a smooth field, the source is smeared out spatially over several grid elements with the maximum about 2 grid elements (= 150 km) west of the real source. Only relative values are given here because the interesting quantity, namely the total release, would be the integral over the whole three-dimensional domain. Other regularisation techniques could be used to avoid the smearing of the peak, but would involve nonlinear constraints and thus require iterations. This does not mean, however, that they would be too costly in terms of computer time, because we only need to apply the source-receptor matrix to the source values of the previous iteration, compute the regularisation terms, and redo the inversion. It is not necessary to repeat the whole simulation as in the case of the adjoint modelling technique without explicit source-receptor matrix. In such iterations, outliers among the measurements could be given a lower weight, so that a more accurate solution would be obtained (reduction of the horizontal location error).

3.3.3 Joint determination of spatial location and temporal evolution

Figure 4 finally shows how location and evolution can be determined together in an efficient manner if the fact that the source covers a single grid cell is used as a priori knowledge. Each grid element was tried out as the location of the source and its temporal evolution was determined by the inversion. The figure shows $r^2$ and the RMSE between observed tracer concentrations and modelled ones, with the reconstructed source term, for each source grid. There is a clear region where the location of the source would be compatible with the observations and an ‘impossible’ region outside, and the grid cell with the smallest error is about 100 km east of the real source location. The resulting temporal distributions of the source for a few such source locations (not shown) are similar to those shown in Fig. 2 for the well-matching source regions.

4. GLOBAL-SCALE SRMS

This work was motivated by the radionuclide network that is being set up for the verification of the Comprehensive Nuclear Test Ban Treaty CTBT (see www.ctbto.org). 79 stations worldwide shall continously take 24 h air samples which are measured for radioactive particles, 39 of them are to measure also radioactive noble gases. Atmospheric transport modelling is to provide estimates of the near-surface regions which could have been the source of any radioactivity found in a sample. Previous work (Hourdin, 2000) shows that transport times of 1-2 weeks could be relevant.

The model set-up is similar to ETEX. We use again $1^\circ$ ECMWF wind fields, now with 60 levels. Because of the low detection threshold of the gamma spectrometric measurements as compared to an expected source term, extremely high dilution ratios are still of interest.

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2 see http://www.boku.ac.at/imp/iws-met-ver/
Therefore, we used 500,000 particles per station, which means quite a high burden for computer requirements. A careful evaluation of sensitivity studies (Seibert and Frank, 2001) indicated that under certain conditions some 10,000 particles would also suffice.

Figure 4 gives an example of the source-receptor matrices belonging to a specific (arbitrarily chosen) sampling interval for a three monitoring sites, as a function of time.

5. CONCLUSIONS AND OUTLOOK

Backward simulations with LPDMs have been established as a tool for source-receptor matrix (SRM) determination in the regional and global scale. No major code changes are necessary in the FLEXPART model. The inversion of the SRM works for ETEX case, and the method appears to be useful for application to nuclear explosions or nuclear power plant accidents. The next applications shall be the inversion of spatially distributed emissions, and climatological studies about the ‘footprint’ of background monitoring stations. The word ‘footprint’ is used here in a similar way as for micrometeorological flux measurements – the areas which contribute to the concentrations measured at the station (we could also speak about the ‘field of view’ of the station).

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REFERENCES


Figure 1: ETEX-1 pseudotracer cloud for 23 Oct 15-18 UTC (upper left), 24 Oct 00-03 UTC (upper right), 24 Oct 15-18 UTC (lower left), and 25 Oct 15-18 (lower right). The real release took place from 23 Oct 16 UTC until 24 Oct 04 UTC. For more explanation, see text.
Figure 2: Temporal evolution of the true (heavy line) and the reconstructed emission. The two reconstructed emission patterns differ in the regularisation (with and without Laplacian included).

Figure 3: Horizontal source distribution (relative units) for given temporal evolution of the release and spatial inversion with minimisation of both length and Laplacian (requiring a smooth field). The real source location is marked by a black dot in a white circle, and the measurement sites by small black dots.

Figure 4: Squared correlation coefficients in per mille [upper] and mean RSMEs [lower] between observed and modelled concentrations for different
assumptions of the location of a 1-grid source whose temporal evolution is determined by inversion of the respective (sub-)SRM. Candidate source grid cells are coloured according to their $r^2$ (RMSE, respectively) values.

Figure 5: Examples of SRMs calculated with the LPDM Flexpart on a global domain. The three receptor sites are indicated by black dots. The receptor time interval is 24 h. The four panels show the SRMs for 24 h source intervals that are 2 days (24-48 h) [upper left], 6 days [upper right], 10 days [lower left], and 14 days [lower right] earlier than end of the receptor interval. FOR unit = 1 means that a $=10^{15}$ Bq source (in one grid cell of 1°x1°) produces a $10^3$ Bqm$^3$ signal at the receptor. After 10 d, areas overlap partially.