RECEPTOR-ORIENTED ATMOSPHERIC TRANSPORT STUDIES AND SOURCE IDENTIFICATION

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Summary

Receptor-oriented modelling with a Lagrangian particle model such as FLEXPART is a powerful tool to investigate the areas from which a measurement of atmospheric trace species is influenced as a function of time. This is shown with examples from the CarboEurope site Hegyhátsál in Hungary. About 60% of the potential influence area were found within 500 km in January 1998 and 80% in August 1998; about 90% are within 1000 km in both seasons. Another example is related to the identification of point sources in the context of the Comprehensive Nuclear Test Ban Treaty. It could be shown that a new method to include the uncertainty of the atmospheric transport calculations, as expressed by a multi-model ensemble, leads to substantial improvements of the results.

1 Introduction

The origin of observed atmospheric trace constituents has traditionally been investigated mainly with trajectory models. Given the improvements in available computing power, nowadays Lagrangian particle dispersion models can be used instead, with a much more accurate simulation of relevant processes. FLEXPART is such a model developed at BOKU-Met (Stohl et al., 1998; Stohl et al., 2005), based to some extent on our older trajectory model FLEXTRA (Stohl et al., 1995). Both models can be used more or less for the same purposes, though traditionally FLEXTRA gives a sequence of trajectory points as the output and FLEXPART gridded concentration fields. However, it is possible to convert trajectory data to residence times and, with the assumption of a mixing height, further into source-receptor relationships (Seibert, 1999). On the other hand, FLEXPART output can be organised in a way which resembles traditional trajectories (Stohl et al., 2002). Trajectories have always been used in forward and backward mode. We have started early to use FLEXPART also in backward mode (Seibert and Stohl, 2000; Seibert, 2001a; Seibert, 2001b). The most important fact to notice for backward calculations is that the output is a source-receptor relationship (sensitivity), closely related to residence times, instead of a concentration.
Two application examples are shown here, the influence region for the CO_2 concentrations at the Hegyhátsál tower in Western Hungary, and source determination for the Comprehensive Nuclear Test Ban Treaty CTBT.

2 Influence Region of the Hegyhátsál Tower

2.1 Introduction

Hegyhátsál in western Hungary is a site where greenhouse gas concentrations and local fluxes (determined by eddy covariance methods) are monitored. Fluxes are also estimated from concentrations with a bulk boundary layer model. However, influence regions (footprints) for local (EC-measured) fluxes and concentrations are of completely different scales. Here we present calculations of the influence region for the concentrations. They were obtained with the FLEXPART Lagrangian particle dispersion model in backward mode. For each observation hour, 10,000 particles were released at 115 m agl (highest measurement level), and tracked back for 6 days. The potential sources were resolved with 1 h temporarily, and horizontally with 1° over Europe and (not shown) 10 km in a nested domain around the site.
We tested both the fast (pure random walk) and slow (accurate) option of FLEXPART for the data of August 1998 and found that the fast option agrees well for the shape of the fields, but it causes systematic underestimation (here the mean over the whole domain is 0.13 instead of 0.18). Therefore, the following results are based on the accurate, slow option.

2.2 Potential influence region for Hegyhátsál

In order to answer the question how far the Hegyhátsál tower can 'see', the source-receptor-matrix has been calculated, and integrated as a function of distance from the tower. First results for January and August 1998 are presented, characterising the two seasons. It should be noted that these results refer to potential sources only.

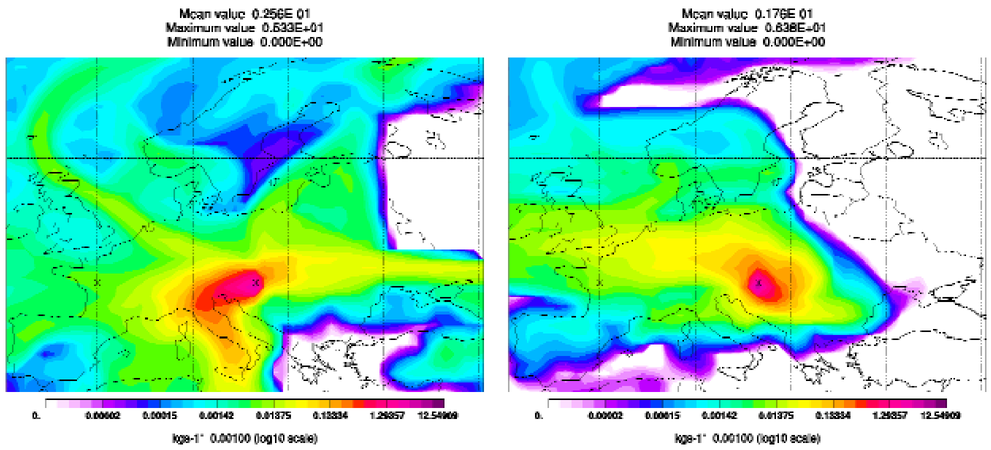


Figure1. *Source-receptor relationship for Hegyhátsál tower. Left: January 1998. Right: August 1998.*

The area of potential influence is larger in January 1998 than in August 1998, because in Summer high pressure systems prevail, blocking the intrusion of fronts and thus the measurements are influenced stronger by the local phenomena. Nevertheless, the potential influence of the tower extends over large parts of Europe. 50% of the total potential influence was found within 180 km in August and within 350 km in January, 80% within 600 and 720 km, respectively. For very large distances, the differences between the seasons become very small. On different days or even in different months, the tower 'sees' quite different parts of Europe, e.g., more areas to the south or to the north of the Alps.

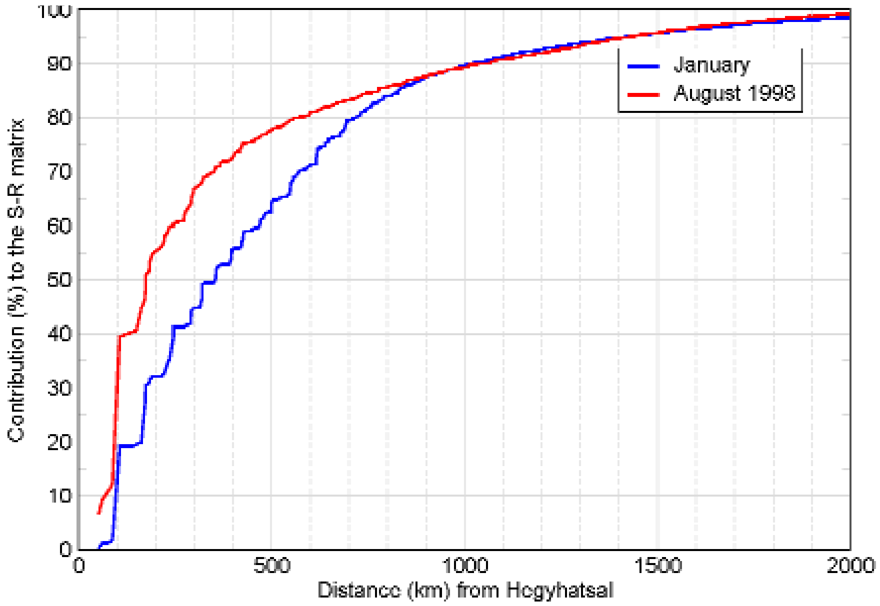


Figure 2. *Cumulative, integrated source-receptor relationship as a function of distance for January and August 1998. The curves converge towards 100% by definition when the whole domain is covered (approx. 3000 km)*

2.3 Actual influence region for CO_2 concentrations at Hegyhátsál

If the question is which sources of CO_2 actually cause the variance in the CO_2 concentration signal observed, the source-receptor-relationship fields alone are not sufficient. They have to be multiplied by the CO_2 fluxes per grid cell at the respective time. Gridded anthropogenic fluxes of CO_2 (resolution 1°) with 1 hour temporal resolution were made available to us by IER Stuttgart. Results are presented here for one day in January (when the biogenic fluxes are small), illustrating also the diurnal course of the long-range source-receptor relationship.
The agglomerations of the Po basin dominate the influence according to our calculations, and this long-range transport is strongest during daytime. We can also observe a shift from western to southern influence regions during the day. It remains to be checked if the model is able to reproduce the observed diurnal course of the concentrations.

3 CTBT Source Determination

3.1 INTRODUCTION TO THE PROBLEM

The CTBTO is currently building up a network of stations to monitor the occurrence of nuclear explosions anywhere on the Earth. It is based on seismic, infrasound, hydroacoustic and radionuclide measurements. The radionuclide network shall comprise 80 stations with daily samplings of particulates, and a subset of stations measuring also noble gas concentrations. The International Data Centre (IDC) of the CTBTO collects these data in real-time. If the radionuclide network finds suspicious radioactivity, the question arises where source of the radioactive material is located. This is a task for atmospheric transport modelling and inversion (Wotawa et al., 2003). The IDC operates its own model (FLEXPART based on ECWMF meteorological fields), but has also an agreement with the World Meteorological Organisation (WMO) that certain national meteorological centres (RSMCs) would deliver source-receptor calculations with their respective models on request for events of special interest.

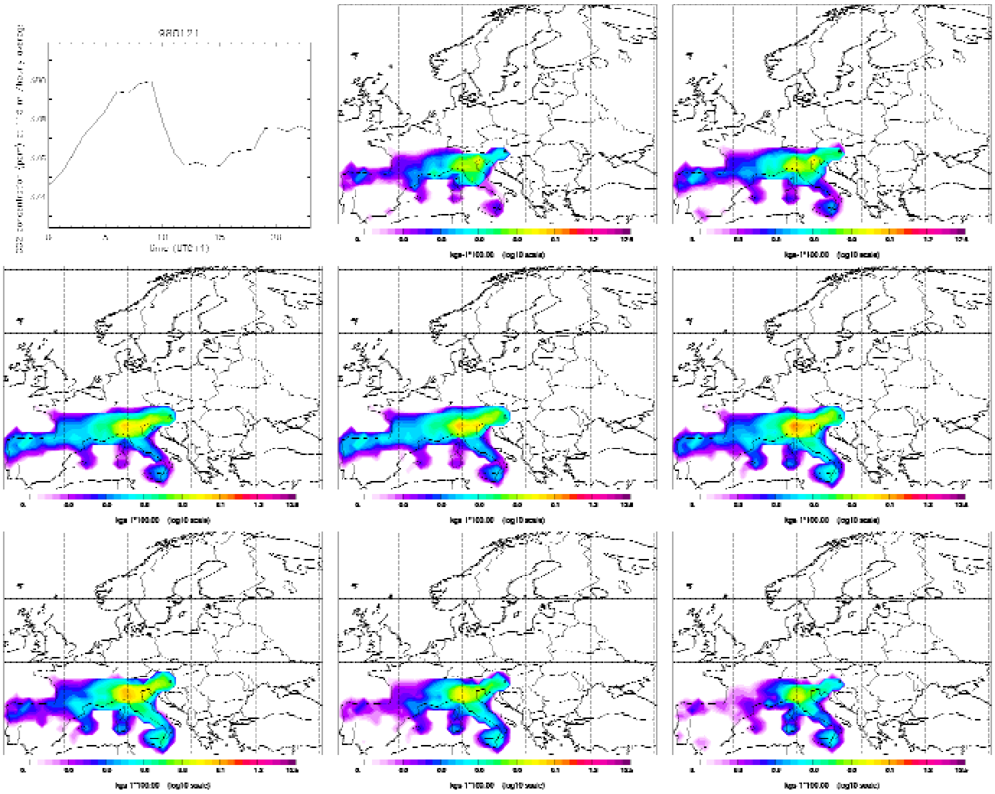


Figure 3. *Observed CO_2 at Hegyhátsál tower on 21 Jan 1998 (top left), and actual influence region for CO_2 concentrations at Hegyhátsál tower on 21 Jan 1998, 00/03 UTC (top row), 06/09/12 (middle row), and 15/18/21 UTC (bottom row).*

Exercises are being conducted where several groups around the world, including the RSMCs, perform receptor-oriented transport and dispersion calculations. In January 2005, such an exercise was carried out to test this collaboration. A fictitious scenario of radionuclide detections was created by a forward run (with FLEXART and NCEP meteorological fields), and the participants were asked to perform source-receptor calculations for 50 samples, taken at several stations and days. The backward transport had to be calculated 120 h (5 days) back from the collection stop of the last sampling. A total of 12 different models / model setups was available. This provides multi-model ensembles of the source-receptor matrix. As a step in our activities to quantify uncertainties in large-scale atmospheric transport modelling and use them to improve in inverse modelling, we used this data set to develop and demonstrate a new method to make the cost function used for the inversion more robust and results more accurate by considering the model uncertainty as given by the multi-model ensemble.

3.2 Outline of the inversion method with uncertainties

The principle of the method is to use each grid cell and 3-hourly time interval as a potential delta-shaped source, and to minimise a cost function which is the weighted sum of all the mean-square errors (MSE) between observed and modelled (using the source hypothesis being tested) concentrations to determine the magnitude of the source. Then we search for the source hypothesis with the smallest cost function value, which is considered to be the desired solution. In order to include the modelling uncertainty as expressed by the multi-model ensemble, a weighting was introduced for each observation-simulation pair which is the inverse of the RMSE averaged over all the models for an prior source estimate.

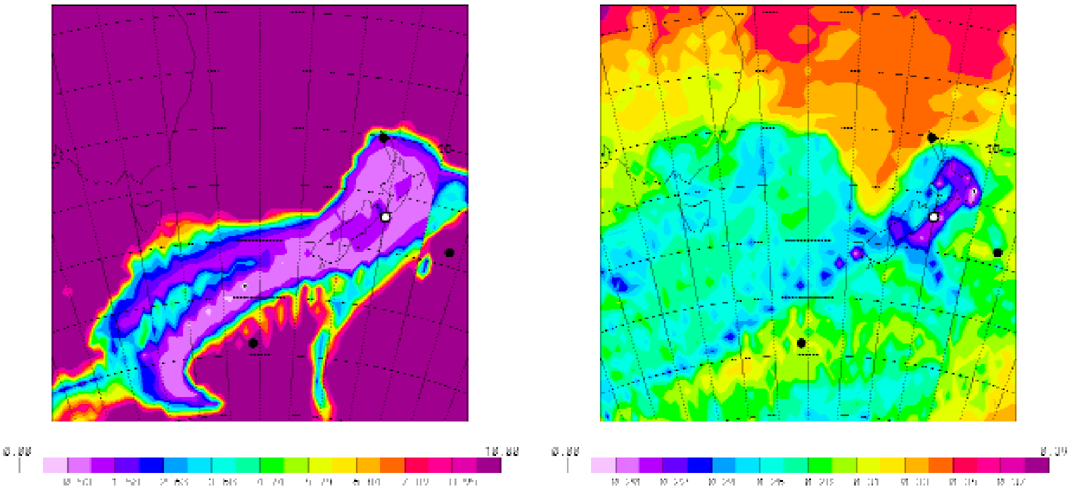
Results

Fig.3 shows that the minimum of the prior cost function is more than 15° SW of the source. Its form is roughly like an U-shaped canyon in a plain, indicating the high uncertainty of the solution. The posteriori solution has a much better shaped cost function (no flat bottom) and a more accurate solution, indicating that the weighted cost function greatly improves the result.

4 Conclusions

FLEXPART is suitable to calculate source-receptor relationships for concentration measurements at sites such as the Hegyhátsál tower with high temporal and spatial resolution. These simulations reveal many details in the influence functions, and confirm the concept of measuring greenhouse gas concentrations at tall towers to be able to constrain their fluxes over large regions by means of inverse modelling. More detailed studies involving longer time periods, biogenic fluxes, and comparisons between modelled and observed concentrations are needed.

A method has been introduced which allows to consider the dispersion model uncertainty in source determination (inverse modelling). The source location is improved considerably and the uncertainty in the solution is much reduced. Further developments like iteration and inclusion of off-diagonal covariance terms will be tested and should easily improve the results even more. Also, the simple approach without multi-model ensemble, just using the MSE of one model, will be tested for comparison. We believe that quantifying uncertainty in atmospheric dispersion modelling, case by case, is important for the future, especially to improve the quality of inversions.



(Please excuse the plotting problem at the date line.)

Figure 4. *Geographic distribution of the cost function. 40 different source times have been tested for each grid cell, and the minimum value is displayed. The true source location is marked by a white dot, the monitoring stations by black dots, and the location of the minimum of the cost function is marked by a tiny black dot in a white circle. Left: A priori results without weighting. Right: A posteriori result obtained by weighting using the multi-model ensemble uncertainties.*

Acknowledgements

This work is supported by FWF project P17924. The work of Klára Tarczay at BOKU-Met was carried out as a scholarship holder of the Marie Curie Training Site AFIP (HPMT-2001-00400). Y. Scholz from IER (Univ. of Stuttgart) kindly made available the anthropogenic CO_2 flux data. The cooperation with G. Wotawa and A. Becker (CTBTO/PTS/IDC) and the participants of the 2005 CTBTO-WMO source location experiment is gratefully acknowledged. BOKU-Met is a member of the ACCENT Network of Excellence funded by the European Commission under FP6.

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