Inverse Modeling Approaches to Infer Surface Trace Gas Fluxes from Observed Atmospheric Mixing Ratios

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7.07.1998

1 Introduction

The accurate temporal and spatial quantification of sources and sinks of radiatively or chemically active atmospheric trace gases, constitutes a considerable scientific challenge. However, this quantification is needed for two purposes. Firstly, it provides critical data for the evaluation of process-based prognostic models, which are used to predict the evolution of the atmospheric composition as functions of anthropogenic impacts and environmental changes. Secondly, in the context of international negotiations to curb the emissions of greenhouse gases, an accurate quantification is indispensable to verify reduction targets claimed by individual nations or groups of nations.

One approach to this problem involves the extrapolation of local flux measurements using geographically referenced databases of properties of the surface (e.g. vegetation cover, topography, soil properties etc.) in conjunction with climatic variables (e.g. temperature, precipitation, insolation etc.) and databases of anthropogenic activities (statistics of land-use, energy consumption, population, agricultural practices etc.). As sources and sinks of trace gases are also reflected in the spatial distribution and temporal variation of their atmospheric mixing ratio, an alternative approach consists of inverting atmospheric mixing ratio measurements into a spatial and temporal distribution of the trace gas sources. In order to do this, the atmospheric transport from the source regions to the observation sites has to be described using simulation models of atmospheric transport.

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During recent decades several global networks of monitoring stations have been developed, e.g. by the Climate Monitoring and Diagnostics Laboratory of the U.S. National Oceanic and Atmospheric Administration (NOAA/CMDL), by the Scripps Institution of Oceanography, La Jolla, California, and by the CSIRO of Australia, which monitor routinely the composition of the atmosphere with increasing accuracy and temporal and spatial resolution. These networks are being supplemented by measurements from airplanes, ships and buoys, and by data from satellite-based remote sensing instruments.

The inference of the distributions of sources and sinks and their temporal variations in a consistent way from all the observations in conjunction with a model of atmospheric transport constitutes an inverse problem of considerable complexity. Ultimately, it requires the design and implementation of a global observing system in which a model of the surface sources is optimized in a consistent way by the different observations, including atmospheric concentrations, isotopic composition, surface features observed from satellites. In many respects such a system may be designed similar to the systems that are currently used in the assimilation of meteorological observations in weather forecasting. The main difference is that the aim in meteorological data assimilation is to find more realistic, dynamically consistent fields of the different meteorological variables to define the initial state of the atmosphere from which a forecast is subsequently computed. Contrary, an observing system for the trace gases would be designed to optimize the surface sources which constitute boundary con-

ditions. However, the mathematical optimization procedures to be used in such a system are similar. At present such monitoring systems for the trace gases are not in place. This is mainly caused by three reasons: (i) the interest in atmospheric trace gases is rather recent; (ii) techniques of accurate measurements on a routine basis have only recently become feasible; and the existing monitoring networks are therefore much less dense than the networks of the meteorological agencies.

In spite of these problems there are many pilot studies, in which global and regional scale sources and sinks of atmospheric trace gases have been estimated from a limited number of observational data using a variety of inverse approaches. Most of these attempts, however, have been restricted to long-lived trace gases (i.e. gases with life times longer than one month), trace gases for which atmospheric chemical transformations or removal processes are either absent or relatively well understood. Examples of such gases include carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O), halocarbons, and carbon monoxide (CO). The main reason for this restriction is that the mathematical inverse problem of these gases is either linear or may be linearized. Short-lived reactive species such as NO_x have not received much attention so far because their atmospheric chemistry is too nonlinear and depends on too many other coupled species to make the problem manageable. However, highly nonlinear cases have been addressed in related fields, such as inversions of oceanic biogeochemical processes (e.g. see the proceedings of the "Workshop on Inverse Methods in Global Biogeochemical Cycles, held in Heraklion, Greece, March 18–20, 1998; to be published as AGU monograph in 1999[1]).

The present review is limited primarily to global approaches. At the end we briefly address the extension to the regional problem, where the term 'regional' is defined as the size of a continent such as Europe or larger.

Some important difficulties of the inversion problem include:

1. Current atmospheric transport models are not perfect.

2. The observational network is very sparse, i.e there are only a small number of monitoring stations. Furthermore at some stations the sampling frequency is low, and there are often temporal gaps in the observations.

3. Technically, the 'inversion' of the atmospheric transport model is not trivial and requires much larger computing resources than running the model in the forward mode.

4. Individual measurements are often not representative of the appropriate temporal and spatial scale of the transport model.

5. Individual observations are of limited accuracy and precision and observations from different monitoring networks are often not easily comparable because of differences in measurement techniques and uses of different standards.

Section 2 addresses the problem of modeling atmospheric transport. In section 3 we discuss the mathematical and technical difficulties of the inversion problem. Section 4 describes the results of two case studies to demonstrate the current state-of-the-art, followed by a brief overview of recently proposed strategies to address the regional problem with considerably higher temporal and spatial resolution.

2 Atmospheric transport

2.1 The continuity equation

The link between sources and sinks of a trace gas and its mixing ratio for a specific location and time period is provided by atmospheric transport. In mathematical terms we have a two- or three-dimensional, time dependent field $Q(\mathbf{x}, t, \chi)$ describing the sources and sinks of the trace gas considered. Generally the source may also depend on the mixing ratio of the gas itself and, in chemically more complex cases, on the mixing ratios of other coupled constituents. The atmospheric transport translates the source field $Q(\mathbf{x}, t, \chi)$ into a three-dimensional, time dependent field of mixing ratios $\chi(\mathbf{x}, t)$. The sources and the mixing ratios are related by the continuity equation

$$\frac{\partial}{\partial t}\rho\chi + \nabla \cdot \mathbf{v}\rho\chi = Q(\mathbf{x}, t, \chi) \qquad (1)$$

[1] http://www.mpimet.mpg.de/gbc/heraklion/

where ρ is the air density and $\mathbf{v}$ the wind vector, both, of course, also three dimensional, time-varying quantities.

If trace gas emission and uptake are linearly related to the mixing ratio of the trace gas then the continuity equation is also linear. In the case of the long lived trace gases this is usually correct, or a linear expansion around a mean background state (e.g. in the case of methane, [Hein et al., 1996]) is a good approximation. In this case the continuity equation can be written as

$$\frac{\partial}{\partial t}\rho\chi + \nabla \cdot \mathbf{v}\rho\chi - k(\mathbf{x},t)\chi = Q(\mathbf{x},t) \qquad (2)$$

$k(\mathbf{x},t)$ describes a first order reaction expressed with the spatially and temporally varying reaction constant.

The solution of equation (2) for the atmospheric mixing ratio $\chi(\mathbf{x},t)$ given trace gas sources, $Q(\mathbf{x},t)$, and reaction coefficients, $k(\mathbf{x},t)$, for a finite time interval requires the specification of an initial mixing ratio field $\chi(\mathbf{x},0)$ and boundary conditions at the borders of the spatial computing domain. For example, in the case of a global model for the troposphere suitable boundary conditions, e.g. prescribed trace gas sources or mixing ratios have to be specified at the tropopause and the Earth's surface.

In practice the continuity equation has to be solved numerically in discretized form (see section 2.2 below). Computing the spatio-temporal distribution of the mixing ratio $\chi(\mathbf{x},t)$ from prescribed sources and sinks constitutes a *forward* model run. In the linear case this may be written formally as a matrix equation

$$\tilde{\mathbf{m}}| = \tilde{\mathbf{T}} \cdot \tilde{\mathbf{q}}| \qquad (3)$$

where the column vector of the mixing ratios $\tilde{\mathbf{m}}|$ includes all model grid points in space and time of the simulation, and likewise the column vector of source values $\tilde{\mathbf{q}}|$ includes all grid points of the source in space and time. It also includes any initial and boundary condition terms. The matrix $\tilde{\mathbf{T}}$ represents the transport model code. The tilde symbol "˜" denotes quantities on the full temporal and spatially discrete resolution of the transport model.

In most cases a reduced problem is of interest: We are not interested in the mixing ratio field on the full model resolution at every time step of the model, but in time averaged mixing ratios (e.g. weekly, monthly or annual means) at a finite number of observation locations. Also the sources of interest are often time averaged quantities, possibly also averaged over larger spatial domains. If the averaging operations both in the space of observations and of sources are linear, we have a linear "model equation" similar to (3):

$$\mathbf{m}| = \mathbf{T} \cdot \mathbf{q}| \qquad (4)$$

Here, the elements of $\mathbf{m}|$ represent the observations (in time and space), and the elements of $\mathbf{q}|$ are the "source components" (in time and space) which include the (possibly spatially and temporally averaged) initial and boundary conditions. The elements of the matrix $\mathbf{T}$ may be interpreted as the sensitivities of the observables with respect to the source components. For example, the element $T_{i,j}$ denotes the derivative of the mixing ratio at the time-space location i with respect to the source component j[2].

2.2 Models of atmospheric transport

Numerical models of atmospheric transport solve the continuity equation for a passive atmospheric trace constituent on a discrete model grid. Typical spatial resolutions in present-day global models are on the order of a few degrees latitude and longitude and 10 to 30 layers in the vertical dimension. Likewise the temporal dimension is also discretized with time steps ranging from a few tens of minutes to a few hours.

Atmospheric transport can be computed "online" as part of an atmospheric general circulation model which provides the meteorology (windspeeds, air properties, subgrid scale transport through clouds, convection or diffusion) [Erickson et al., 1996]. Alternatively, the transport may be calculated "off-line" by reading the meteorological fields from stored output of a general circulation model simulation or from analyzed fields of a weather forecast model. In the latter case subgrid scale transport (see below) has to be recomputed in the transport model, which constitutes a non-trivial task (e.g. see Heimann [1995]). By using archived analyses from weather forecast mod-

[2]Formally, the matrix $\mathbf{T}$ is closely related to the Green's function of the reduced problem.

els, the off-line approach has the advantage that the "real" meteorology is used in the simulations, i.e. the model describes the actual atmospheric circulation fields prevailing during the particular time period when the measurements were obtained. In contrast, the on-line general circulation model approach yields simulation results that can only be compared to the observations in a statistical sense, e.g. as monthly or annual means.

The models typically split the transport into two main components: "advection", i.e. the transport resolved on the model grid, and subgrid scale transport which includes the effects of all processes on temporal and spatial scales not resolved on the model grid (e.g. transport through cumulus clouds, thermal convection, diffusion or boundary layer mixing).

Formally this can be written as

$$\frac{\partial}{\partial t}\bar{\rho}\bar{\chi} + \nabla \cdot \bar{\mathbf{v}}\bar{\rho}\bar{\chi} + CONVEC(\bar{\chi}) = \bar{Q}(\mathbf{x}, t, \chi) \quad (5)$$

where the overbars indicate quantities averaged over the model grid. The second term on the left represents the resolved "advection", while $CONVEC$ denotes the subgridscale transport processes.

2.2.1 Advection

Many numerical schemes have been developed in the past for the numerical solution of the part of equation (5) which denotes the grid resolved transport. Important properties of numerical advection schemes are accuracy, linearity, mass conservation, small numerical diffusion, computational efficiency, and positive definiteness. A review of some of the more commonly used Eulerian numerical schemes has been given by Rood et al., [1987]. Examples of more advanced techniques are discussed by Spee et al., [1997] and Spee [1998].

Spectral techniques, which used to be popular in the atmospheric general circulation modeling community, expand the tracer mixing ratio field into spherical harmonics. This approach is not very well suited for tracer transport because the spectral transformation may generate negative concentrations.

There exist also lagrangian schemes, in which the atmosphere is represented by a series of discrete air parcels, each containing an amount of tracer in accordance with the local tracer mixing ratio. These parcels are moved around the atmosphere by the specified wind velocity and keep their identity over the entire length of the model integration time period [e.g. Penner et al., 1991]. Lagrangian schemes have much less numerical diffusion as compared to Eulerian techniques. However, it is difficult to adequately represent subgrid scale mixing processes in such a scheme. Furthermore, because the parcel distribution tends to become non-uniform with time, the schemes need a large number of parcels in order to represent the tracer fields with a sufficient spatial resolution. This makes the schemes in general computationally expensive, except in the situation where the transport of several tracers is to be computed simultaneously, because the air parcel movement has to be computed only once for all tracers.

In semi-lagrangian schemes the tracer is transported during each time step in a lagrangian fashion, i.e. attached to discrete air parcels. After each time step the three-dimensional tracer mixing ratio fields are reconstructed by interpolation from the parcels to the regular model grid. This interpolation tends to have problems with mass conservation [Rasch and Williamson, 1990].

Despite the different numerical techniques employed in present day atmospheric transport models the advection is not considered to be the most critical component in need of improvement.

2.2.2 Subgrid scale transport processes

In the course of the discretization of the basic continuity equation on the model grid the effects of all transport processes on smaller spatial and temporal scales must be described ("parameterized") as functions of the values of the meteorological variables on the resolved scale. Some of these processes include vertical transport through cumulus clouds ("wet convection"), thermally driven dry convection, turbulent diffusion and vertical mixing in the surface and boundary layers. A comparison of several vertical subgrid scale transport parameterizations can be found in Mahowald et al., [1995].

Parameterizations for these processes also exist in atmospheric general circulation models and in weather forecast models. In principle these could be transferred to the transport models. However,

except for water vapor, the transported quantities (energy, momentum) have different properties as compared to trace gases (source distributions, lifetimes) making such a transfer non-trivial.

A very critical subgrid scale transport process is boundary layer mixing over the continents. Most atmospheric observations are being taken close to the surface in the planetary boundary layer, and most of the long-lived trace gas sources are at the earth's surface. The height of the layer, into which the trace gas is emitted, and the exchange processes with the overlaying free troposphere critically determine the simulated mixing ratio in this layer.

In the case of a trace gas with strong diurnal and seasonal sources, such as CO_2, systematic temporal changes of the transport (e.g. height of the boundary layer during day and night and also during the different seasons of the year) are responsible for "rectifier effects", i.e. spatial structures in the mean annual mixing ratio field even if the trace gas source on annual average were balanced at every grid point to zero. These structures arise because of the temporal covariance between source and atmospheric transport [Keeling et al., 1989, Denning et al., 1995, Denning et al., 1998, Law et al., 1996, Rayner and Law, 1996]. Hence, observed spatial structures in the mean annual mixing ratio fields reflect a combination of true sources and sinks and of the "rectifier effects". Because of this an accurate representation of the "rectifier effect" in the atmospheric transport model is an indispensable necessity. Unfortunately, there is at present no way to independently verify the simulated rectifier effect, which, at least for CO_2, constitutes a serious limitation in current inversion studies.

2.3 Validation of atmospheric transport models

An assessment of the realism of present atmospheric transport models is beyond the scope of this review.

A critical quantity of global atmospheric transport models is their portrayal of the meridional interhemispheric transport. This property can be validated by means of simulations with trace gases of known surface sources, such as the radioactive Krypton-85 [Jacob et al., 1987, Heimann and Keeling, 1989], CFC-11 [Prather et al., 1987] or Sulfurhexafluaride (SF_6) [Levin and Hessheimer, 1996]. A consistent intercomparison of global atmospheric transport models with SF6 has recently been undertaken within the phase 2 of the TRANSCOM project [Denning et al., 1998].

As discussed above, at present there is no method available to validate the simulated "rectifier effect". The way the different models simulate this effect has been explored in phase 1 of the TRANSCOM project [Rayner and Law, 1995, Law et al., 1996].

3 Methodological aspects of the inversion problem

3.1 The mathematical problem

The inverse problem consists of inverting equation (4) to find a solution for the source components $\mathbf{q}|$. The structure of (4) shows that this involves two tasks: First, the matrix $\mathbf{T}$ has to be computed and second, the linear equation system has to be solved for the unknown source components.

Depending on the number of observables and the number of source components, the matrix $\mathbf{T}$ may be very large, hence its computation may be very expensive. But also the solution of equation (4) for the source components is not trivial. Typically there are only a limited number of observations in space and time. On the global scale, currently most of the gases under consideration are monitored at less than 10 stations continuously or quasi-continuously and at less than a few hundred stations with a sampling frequency of less than 1-2 observations every week. For some of the gases (e.g. CO) remote sensing observations from space craft are available, but these typically are of limited accuracy and may include only the vertical integral. On the other hand, most source and sink processes are strongly heterogeneous in space and time. To represent this heterogeneity adequately, one is tempted to choose a high resolution in the space of source components, hence the matrix $\mathbf{T}$ becomes rectangular with a much larger number of columns than of rows. In this case the source inference problem is highly underdetermined. But even if one restricts the number of source components to a smaller number than the observations, the equation system might still be "ill-conditioned" because of the diffusive nature of the atmospheric transport. Hence additional restrictions (simplifi-

cations, assumptions, '*a priori*' information from other data sources) are needed in order to make the problem mathematically well conditioned. Some of these are discussed in the subsections below.

3.2 Reducing the spatial resolution

A reduction of the spatial resolution is obtained by dividing the globe up into only a small number of regions (typically 10–100). The surface sources for each of these regions are then prescribed in their spatial and temporal pattern, whereas their overall magnitude is left as an unknown scaling parameter to be determined in the inversion procedure. In the case of a high-resolution, computationally expensive atmospheric transport model, this is the only way to make the determination of the matrix $\mathbf{T}$ manageable.

In this case the matrix $\mathbf{T}$ can be determined by brute force: The atmospheric transport model code is run for each of the source regions separately. $\mathbf{T}$ is then obtained by recording the contributions from the different source components at the observation sites. In this "synthesis inversion" the source regions may be specified as a simple geometric breakdown of the globe, such as the sources in latitudinal bands. This approach has been applied e.g. by Brown [1993, 1995] to deduce the sources of methane on latitudinal bands.

A slightly different variant of the "synthesis inversion" consists of decomposing the source field into several, possibly spatially overlapping components that represent different source processes. This has been applied e.g. in the case of methane by Hein et al. [1996]. Here the different source and sink processes of CH_4 are specified by their global spatial and temporal patterns (e.g. emissions from peats and bogs, from coal mining, oil or gas production, waste disposal, rice paddies, cattle etc.). For CO_2 this approach has been chosen by Enting et al. [1995], Bousquet [1997], and Rayner et al. [1998].

The "synthesis inversion" approach typically yields a relatively stable inversion (possibly even an overdetermined equation system), since the number of source components in most cases is chosen to be less than the number of observations. However, the use of predefined "rigid" spatio-temporal source patterns strongly influences the resulting solution.

3.3 Simplifications in the temporal domain

The atmosphere has a limited "memory". An impulse input of a conservative tracer released at a specific location and time eventually becomes homogeneously mixed. The longest mixing time defines a time horizon, beyond which any source or sink contributes only to the global background mixing ratio of the tracer. Within the troposphere, e-folding times for the decay of pulse input are on the order of up to 1 year [Weiss et al., 1983]. Mixing times into the stratosphere, however, are much longer. Hence, for a tropospheric tracer, a conservative value for the time horizon is 3 to 4 years [Heimann and Keeling, 1989]. For example for the simulation of a transient tropospheric tracer, such as the halocarbons during the 1980's, the history of the F11 sources prior to 1977 is irrelevant, only their cumulative global integral determines the atmospheric background F11 mixing ratio [Bloomfield et al., 1994].

This implies that the elements in the sensitivity matrix T which refer to sources more than 4 years prior to the time of the observation are given simply by the increment in the globally averaged mixing ratio per unit input of the tracer. Hence the atmospheric transport model must be run for only 4 years for each of the source components in order to determine the sensitivity matrix T.

A further simplification arises if one assumes that the large-scale atmospheric transport does not change significantly from year to year. Indeed, this appears to be the case as witnessed by a close inspection of the atmospheric time history of SF_6, which shows that the mixing ratio differences between the hemispheres remained relatively constant over the last decade, when emissions did not change significantly [Maiss and Levin, 1994, Levin and Hessheimer, 1996]. In this case one uses the same wind fields year after year. As an additional check one can repeat the simulation with the meteorology from another year [e.g. Knorr and Heimann, 1995].

Finally, an additional, considerable simplification can be achieved by addressing only the "quasi-stationary" problem: i.e. by assuming that all sources and sinks may include a seasonality, but are invariant from year to year. If also the transport is assumed to be the same year after year, then it is easily seen that after an initial transient

the atmospheric mixing ratio at any location, $\mathbf{x}$, as a function of time, t, can be expressed as an offset, $\chi_0(\mathbf{x})$, a globally uniform linear trend a_t and a seasonal cycle $S(\mathbf{x}, \tau)$:

$$\chi(\mathbf{x}, t) = \chi_0(\mathbf{x}) + a_t t + S(\mathbf{x}, \tau) \qquad (6)$$

where τ denotes the time since the beginning of the year. Many global inversion studies of CO_2 [e.g. Enting et al., 1995] or of CH_4 [e.g. Hein et al., 1996] employed this approximation.

The extension to the interannually varying case has been addressed by Bloomfield et al. [1994] for CFC-11, and Rayner et al. [1998] for CO_2.

3.4 Bayesian approaches

Bayesian approaches to the inverse problem provide a means to include *a priori* information on the unknown source components in the inversion procedure [see e.g. Tarantola, 1987]. They are based on a formulation of the problem in terms of probability distributions in the joint space of sources and concentrations. In practice these probability distributions are assumed to be Gaussian. An *a posteriori* source estimate is derived, which is optimal in the sense that it is as close as possible to the prescribed *a priori* sources, while the resulting simulated concentrations are as close as possible to the observations. Thereby "close" is defined relative to specified uncertainties in both, the observations and the *a priori* source estimates.

In an otherwise underdetermined inverse problem, the Bayesian approach yields a unique solution from all source configurations that are consistent with the observations. In an ill-conditioned inverse problem the Bayesian approach limits the amplification of errors in the observations when inferring source combinations that are badly constrained by the observations (see Enting [1993]).

The necessary *a priori* information, i.e. the *a priori* sources, may be provided by interpolating direct flux measurements, or they may be obtained from prognostic source models. In both cases quantifying the uncertainties of the *a priori* estimates is crucial and should reflect the understanding of the underlying source processes.

Bayesian inversions have been carried out by Enting et al. [1995], Bousquet [1997], Rayner et al. [1998], and Kaminski [1998] for CO_2 as well as by Hein and Heimann [1994] and Hein et al. [1996] for CH_4.

3.5 Technical approaches

In the case of sparse networks, adjoint models provide an efficient tool to compute the matrix $\mathbf{T}$ in equation (4). Instead of computing the partial derivatives $T_{i,j}$ by brute force with forward transport model runs for each source component j, the adjoint model begins at the location and time of the observations (i). Starting with an infinitesimal mixing ratio deviation at the observation point i, the adjoint model works its way backward in time. Thereby it determines the necessary changes in the source components at earlier time points that would induce the infinitesimal mixing ratio deviation at i. In this way the adjoint model essentially propagates the sensitivity of the modeled concentrations from the observational sites i backwards to the sources [Errico, 1997, Corliss and Rall 1996].

Using forward model runs to compute $T_{i,j}$ the required computational resources are proportional to the number of source components, and relatively independent of the number of observations, while for the adjoint model the required computational resources are proportional to the number of observations, and relatively independent of the number of source components. The major difficulty of the adjoint approach is the time consuming construction of the adjoint model. This can be overcome, however, by using tools for automatic generation of adjoint code, which are being developed, e.g. Odysee [Rostaing, 1993] or the Tangent and Adjoint Model Compiler (TAMC) [Giering, 1996]. Using the TAMC, Kaminski et al. (1996) developed the adjoint of the atmospheric transport model TM2 [Heimann, 1995], which allowed an efficient computation of the matrix $\mathbf{T}$ for an observational network of approximately 30 stations with monthly observations and monthly sources on the 8 by 10 degree horizontal model grid; i.e. a matrix for observations at $30 \times 12 = 360$ time-space locations and $12 \times 24 \times 36 = 10368$ source components (a matrix with 3.7 million elements).

3.6 Representativity of individual measurements

In most global studies performed so far, the modeled monthly mean mixing ratio is compared to estimates of the monthly mean mixing ratio at the observing sites. The mixing ratio computed by a model is "representative" for the spatial and temporal resolution of the model grid. On the other hand, individual measurements at a monitoring site in general reflect also local transport and source processes on finer scales than resolved by the model. The comparison between observations and model results hence requires considerable care in order to avoid potentially serious biases.

On the observational side one typically tries to eliminate the influences of local contamination by following a data selection protocol designed to sample so-called "background air" under "baseline conditions". This is achieved, e.g. by specifying the time of day and other requirements (minimum wind speed, particular wind directions, other meteorological conditions) under which air sampling is performed. If possible also the information from other, concurrently measured tracers (e.g. Radon-226 and Radon-222 [Polian et al., 1986]) can be used. An additional elaborate data screening is also used after the sampling in order to eliminate "outliers" believed to represent local ("polluted") samples before computing monthly means. In many cases these procedures may indeed yield observations that are representative to the simulations. However, it is difficult to estimate, to what extent these approaches in fact eliminate the local unwanted source contributions. The data selection procedures by themselves may also induce biases because potentially air is sampled that reflects a different model grid box than the one containing the observing station.

On the other hand, a sampling strategy should also be included in the model that mimics the procedures employed at the observing stations. For example it does not make sense to compute monthly averages in the model by continuous sampling if the model exhibits a diurnal cycle which includes night-time inversions while the observations are only taken in the afternoon when vertical mixing is strongest. Unfortunately, the implementation of adequate sampling strategies in the models is rather cumbersome, as it will be different for each station.

A thorough demonstration of the potential biases incurred by different sampling procedures in the model and the observations has been given by Ramonet and Monfray [1996].

3.7 Inhomogeneity of sampling networks

In many cases, the spatial inhomogeneity of the current observational networks is obvious. As an example Figure 1 shows the CO_2 monitoring network maintained by the U.S. National Oceanographic and Atmospheric Administration's Climate Monitoring and Diagnostic Laboratory [Conway et al., 1994] in which the tropics and in particular the tropical land masses are undersampled. Also, sampling of oceanic regions has been favored by choice of observational sites and by the definition of baseline conditions (see the previous section). On the other hand, in most inversion studies one chooses a relatively small number of unknown source components (see section 3.2). In combination with inhomogeneous sampling, however, a low resolution in the space of sources is likely to yield a biased estimate of the inversion as demonstrated by Snieder [1993]. For inverse problems in seismic tomography, Snieder [1993] and Trampert and Snieder [1996] demonstrate how to reduce the bias at the cost of increasing the uncertainty. For inversion of the global atmospheric transport and the current global networks, the magnitude of this bias is yet to be quantified.

3.8 Calibration problems between different measurement networks

Reported observations from different measurement agencies are often not calibrated well against each other. This is a serious problem, because, in the case of long-lived species, small mixing ratio differences between different stations imply relatively large sources, hence offsets between different networks may induce substantial biases in the inferred source distributions.

In principle the calibration differences among the networks could be reduced by systematic intercomparisons of standards, measurement techniques, sampling procedures and sample handling. Several of such intercomparison efforts are currently underway within the different trace gas species measure-

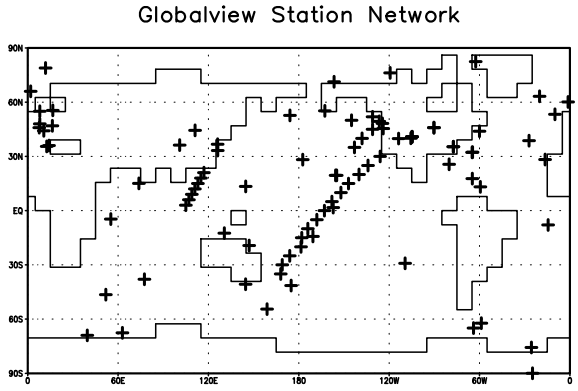

Figure 1: Global station network of the Climate Monitoring and Diagnostics Laboratory of the National Oceanic and Atmospheric Administration [Conway et al., 1994].

ment communities. Unfortunately, these tedious efforts are scientifically not very rewarding and are therefore often lacking sufficient funding support. It must be stressed, however, that such efforts are very important and should receive highest priority.

As an intermediate step it is also possible to short-cut detailed intercomparison procedures by merging the existing datasets in a way that takes possible offsets into account. In the case of CO_2 such an effort has been undertaken by NOAA-CMDL in the creation of the GLOBALVIEW dataset [Masarie and Tans, 1995]. Alternatively, one can of course also use only the observations from a single network. Or, it might also be possible to include the calibration offset between different networks as unknown parameters to be determined in the inversion.

3.9 Uncertainties

In principle, an inversion is subject to two sources of errors: Observations are of finite precision, and models are imperfect. Clearly, these errors cause uncertainties in the source fields that are determined by the inversion. In general, the observations are not representative for the spatial and temporal scale of the model predictions. The associated error in comparison of both quantities, which (depending on how one names the observed quantity) can either be subsumed under model error or observational error, or can be treated separately as a representation error. Computationally, the difference of these errors is unimportant, since they enter the inversion in their sum.

The formal treatment of these errors is in terms of probability distributions. For the sake of computational convenience, usually Gaussian distributions are assumed, and for a linearization of the transport model, one can derive a Gaussian distribution of the source field that is consistent with the sum of the abovementioned errors. Due to this Gaussian assumption, the actual computations are then manipulations of means and covariance matrices. In Bayesian inversions, the *a priori* information is also quantified in terms of a mean and a covariance matrix. The inversion then derives a posterior mean field and covariance matrix. The information in the atmospheric observations then is reflected in a change of this mean and a reduction of the uncertainties.

Practically, errors in the transport model are hard to quantify, potential approaches are either model intercomparisons or checking against observed concentrations for tracers with well known source distributions. Usually transport model errors are neglected. Similar problems complicate explicit inclusion of the representation error. Selection of observations according to their representativeness (see section 3.6) is an approach to minimize this error.

4 Examples

In 1995 Enting et al. presented a Bayesian synthesis inversion for CO_2. Employing observations of atmospheric CO_2 from the global networks of the NOAA-CMDL [Conway et al., 1994] together with $^{13}C/^{12}C$ isotopic ratio measurements from the CSIRO [Francey et al., 1995] , they solved the quasi stationary problem (see section 3.3) for the magnitudes of about 20 unknown source components. Since the study by Enting et al. [1995] a number of research groups have been refining inversion in various directions. As examples, we briefly demonstrate the long-term average annual mean source fluxes of CO_2 inferred in the studies of Rayner et al. [1998] and Kaminski [1998].

By resolving the interannual variability in the sources, Rayner et al. [1998] removed the quasi

stationary assumption. For the period from 1980 to 1995, in a Bayesian synthesis inversion, they inferred the monthly magnitudes of about 30 source patterns from global observations of CO_2, complemented by one station's time series of the isotopic CO_2 composition and the station's linear trend in the oxygen to nitrogen ratio. Figure 2 shows *a priori* and *a posteriori* estimates for the long term annual mean source fields. The figures show only the fluxes of CO_2 between the atmosphere and the ocean, respectively the atmosphere and the terrestrial biosphere; the dominant anthropogenic source, i.e. the CO_2 from the combustion of fossil fuels has been subtracted.

Keeping the quasi stationary assumption, in contrast, Kaminski [1998] removed the simplifications in the spatial domain by means of an adjoint approach (see section 3.5). In principle this approach allows the computation of adjustments to the *a priori* source field resolved on the full horizontal grid of the transport model (8° latitude by 10° longitude). In this study observations of the atmospheric CO_2 mixing ratio from 25 stations of the NOAA-CMDL [Conway et al., 1994] network for the period from 1981 to 1986 were used (i.e. a subset of the stations shown in Figure 1). The highly underdetermined nature of this problem necessitated a Bayesian approach (see section 3.4). The *a priori* source fields were obtained from energy use statistics for the fossil fuel CO_2 source, from statistics of land use change and from spatially explicit carbon cycle simulation models for the ocean and the terrestrial biosphere.

Figure 3 shows the *a priori* and *a posteriori* estimates for the annual mean source fields and their difference. Overall, Figure 3b reveals a similar structure of the non-fossil fuel CO_2 source fields as the *a posteriori* source field of Rayner et al., [1998] (Figure 2): A strong sink for CO_2 in the northern mid-latitudes and a smaller sink in the southern hemisphere oceans. The tropical oceans and in particular the equatorial Pacific reflect regions with CO_2-outgassing.

Despite the considerable spatial and (not shown) temporal resolution of the source fluxes of CO_2 as inferred in the two studies, one must realize, that the *a posteriori* solution is determined considerably by the *a priori* information. The extent to which the atmospheric measurements and the transport model provide additional information can be investigated by analysis of the Singular Vector Decomposition (SVD) [Menke, 1989] of the model matrix $\mathbf{T}$ in equation (4) [Kaminski, 1998].

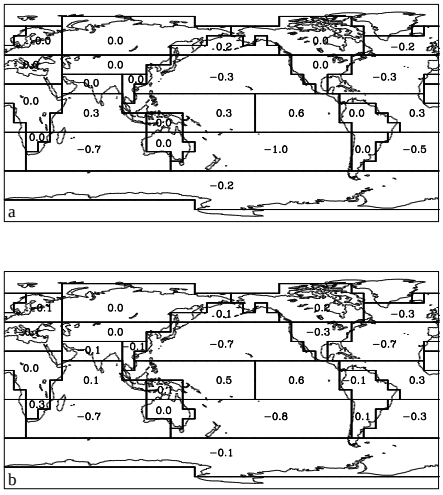

Figure 2: a): *A priori* long-term annual mean source distribution as specified in the inversion study of Rayner et al. [1998]. b): *a posteriori* sources inferred by the inversion procedure. Numbers represent source fluxes of CO_2 in GtC yr^{-1}. Fluxes into the atmosphere are indicated by positive numbers.

Alternatively, the reduction of the *a priori* uncertainty induced by the inversion provides also a measure to assess the relative importance of the *a priori* information. Figure 4 displays the *a priori*, the *a posteriori* uncertainty and the relative reduction for the annual mean CO_2 fluxes from study of Kaminski [1998]. It is seen that in most regions the uncertainty is reduced by only a relatively small amount (at most 20%). Clearly the uncertainty reductions are largest close to the monitoring stations. If averaged over larger regions, e.g. zonally or over continents the percentage uncertainty reduction becomes larger. In particular the large scale features of the resulting solution as described above are found to be significant [Kaminski, 1998]. However, this assumes uncorrelated *a priori* uncertainties between the different gridboxes. This assumption is difficult to assess in the present case since the *a priori* flux fields were partly derived

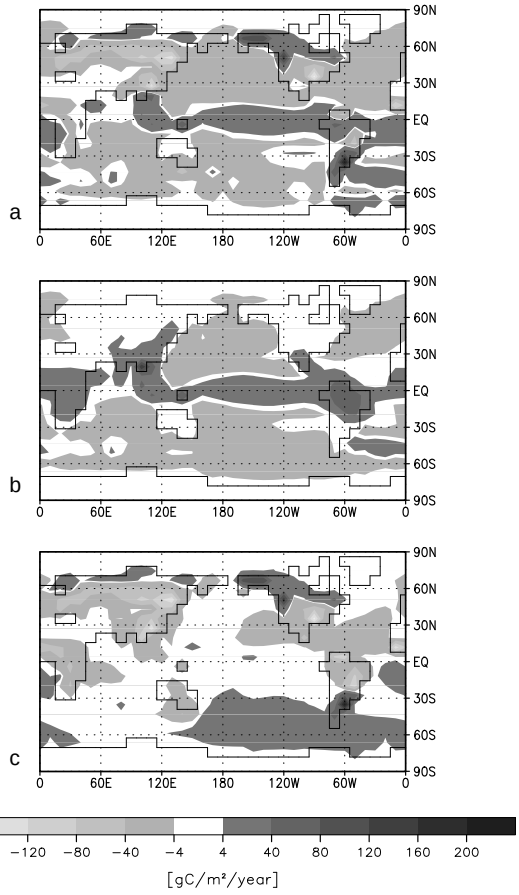

Figure 3: a) *A priori*, b) *a posteriori* and c) difference of the annual mean source flux fields of CO_2 as resolved on the full model grid (8° latitude by 10° longitude) in the study of Kaminski, [1998], using an adjoint model of atmospheric transport. Shown are only the CO_2 fluxes between atmosphere and ocean, respectively atmosphere and the terrestrial biosphere; the anthropogenic source flux from the combustion of fossil fuels has been subtracted. Fluxes into the atmosphere are indicated by positive numbers.

from oceanic and terrestrial carbon cycle models, for which estimates of their errors and associated covariance structure are not readily available.

The relatively small uncertainty reduction provided by the atmospheric observations is sobering. For the case of CO_2 it demonstrates clearly, that the present atmospheric background monitoring networks do not allow a regional determination of the sources and sinks of CO_2 without significant *a priori* information on their spatio-temporal distribution and magnitude. On the other hand, maps such as Figure 4 provide a means to assess the merits of individual stations of the monitoring networks. Using this tool strategies for the optimization of the networks may be devised.

5 New approaches

Recently, a series of new projects have been initiated or proposed to determine the regional sources and sinks with a much higher spatial and temporal resolution than possible with the existing global approaches as discussed in the previous sections ("Carbon America" [Tans et al., 1996], the Large-scale Biosphere Atmosphere (LBA) experiment over Amazonia [Anonymous 1997], COBRA [Wofsy et al., 1997] and "Eurosiberian Carbonflux" [Heimann et al., 1997]).

As discussed above, current observational networks are heavily biased toward oceanic areas. A better and more detailed regional determination of continental sources requires observations closer to these sources. This represents a very difficult problem because of the complex meteorology and the typically strong and heterogeneous terrestrial sources and sinks.

On the observational side these new proposals call for a considerable extension of the monitoring platforms. In particular they include measurements in the vertical dimension by aircraft and, if possible, observations from remote sensing platforms. In addition a multi-tracer approach may be chosen, in which several atmospheric species are measured simultaneously. E.g. in the case of the Eurosiberian Carbonflux project the following tracers are to be measured at several sites on bi-weekly vertical profile flights over a time period of at least three years: CO_2 and its carbon and oxygen isotopes, CH_4 and its carbon isotopes, CO, Radon-

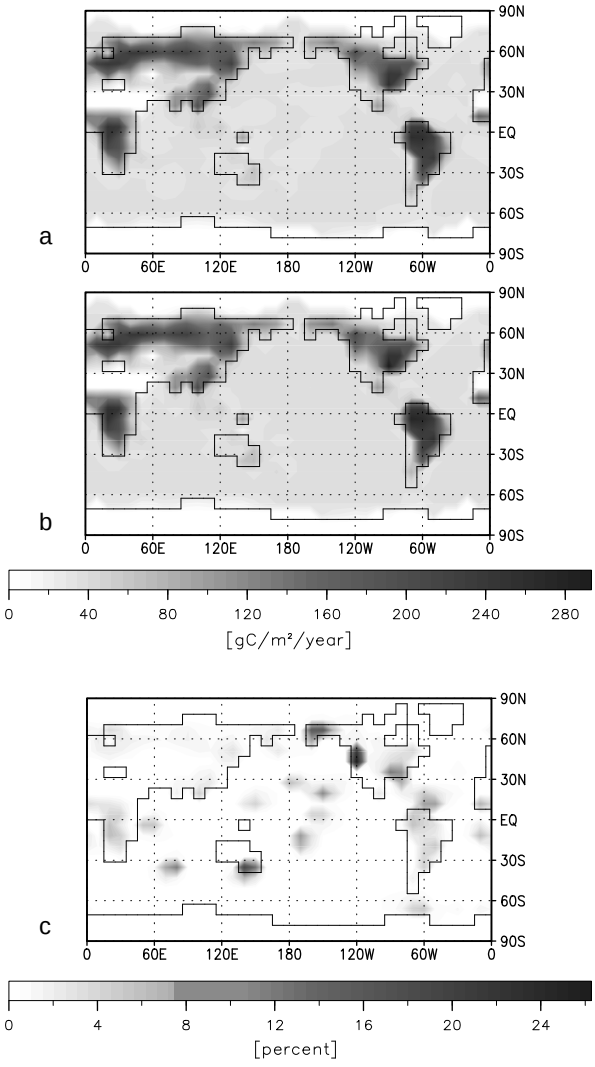

Figure 4: a) *A priori* and b) *a posteriori* uncertainty of the annual mean source flux field of CO_2 corresponding to Figure 3a) and b). c) percentage reduction between the *a priori* and the *a posteriori* uncertainty.

222 (a radioactive noble gas with a half-life of 3.8 days), SF_6 and others. Since all tracers are subject to the same atmospheric transport the problem of representativity of a single measurement with respect to the modeled grid averaged mixing ratio may be addressed in an effective way.

The modeling of the atmospheric transport over the continental sources and sinks also poses a substantial challenge. The heterogeneity of the terrain and the induced complicated meteorology necessitates the use of meso-scale models. Such models must have a horizontal grid resolution of 10–50 km covering an total area of up to 25 10^6 km^2, and need to resolve the diurnal cycle of boundary layer mixing and convection in considerable detail. In order to run these models the large-scale meteorology has to be specified from weather forecast analyses. An additional problem are the boundary conditions for the tracer(s) under consideration, which will have to be specified from the output of a global, low-resolution simulation. An example of such a simulation system has been described for simulations of CO_2 in the arctic region [Engardt, 1997, Engardt and Holmen, 1997] based on the MATCH model [Robertson et al., 1996].

How will these models perform in an inverse approach? Even if the difficulties to realistically model the transport over the continental regions can be overcome, it is not clear if reliable regional source flux estimates may be obtained by this approach. This is because the large and mostly unknown heterogeneity of the source flux distribution might require a sampling density in space and time that is not feasible. Clearly, without a success in these new approaches the second goal of inversion studies mentioned in the introduction, i.e. the quantification of regional flux estimates for the verification of national greenhouse gas reduction targets will remain elusive. Nevertheless, it is hoped, that the new projects will provide some insight into the regional source estimation problem.

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