Inverse Problems in Atmospheric Constituent Transport

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CAMBRIDGE
UNIVERSITY PRESS

PUBLISHED BY THE PRESS SYNDICATE OF THE UNIVERSITY OF CAMBRIDGE
The Pitt Building, Trumpington Street, Cambridge, United Kingdom

CAMBRIDGE UNIVERSITY PRESS
The Edinburgh Building, Cambridge CB2 2RU, UK
40 West 20th Street, New York, NY 10011-4211, USA
477 Williamstown Road, Port Melbourne, VIC 3207, Australia
Ruiz de Alarcón 13, 28014 Madrid, Spain
Dock House, The Waterfront, Cape Town 8001, South Africa

http://www.cambridge.org

First published 2002

Printed in the United Kingdom at the University Press, Cambridge

Typefaces Times 10.25 / 13.5 pt and Joanna *System* LaTeX 2_ε [TB]

A catalogue record for this book is available from the British Library

Library of Congress Cataloguing in Publication data

Enting, I. G.
Inverse problems in atmospheric constituent transport / I. G. Enting.
p. cm.
Includes bibliographical references and index.
ISBN 0 521 81210 0
1. Atmospheric diffusion – Mathematical models. 2. Dynamic meteorology – Mathematical models.
3. Inverse problems (Differential equations) I. Title.

QC880.4.A8 E68 2002
628.5′3′015118 – dc21 2001052977

ISBN 0 521 81210 0 hardback

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Chapter 1

Introduction

> One cannot possibly study the disease unless one understands what it means to be healthy. We probably only have a few more decades to study a 'healthy' earth.
>
> R. F. Keeling: Ph. D. Thesis [251].

1.1 Overview

Human activity is changing the composition of the atmosphere. This goes beyond the often obvious problems of local and regional pollution – even in remote locations there are changes in concentrations of minor atmospheric constituents such as carbon dioxide, methane and nitrous oxide. These and other long-lived gases affect the balance of radiation of the earth – they are the so-called greenhouse gases. Other long-lived gases are implicated in the decrease in concentration of ozone in the stratosphere.

The ability to understand the current atmospheric budgets of these trace gases is essential if we are to be able to project their future concentrations in the atmosphere. This book concentrates on one group of techniques that are being used to improve our knowledge – the interpretation of spatial distributions of trace-gas concentrations. An important theme of this book is the use of a statistical approach as being essential to obtaining realistic assessments of the uncertainties in the interpretation of trace-gas observations.

Modelling of the atmospheric transport of carbon dioxide (CO_2), methane (CH_4) and other greenhouse gases is used to interpret the observed spatial distributions of these gases. The spatial distribution of trace-gas concentrations represents a combination of the effect of spatially varying sources and sinks and the effect of atmospheric transport. Therefore a model of atmospheric transport is needed if the source/sink distributions are to be deduced from observed concentrations. The main reason for

deducing the source/sink distributions is to help identify and quantify the processes responsible. We define 'tracer inversion' as the process of deducing sources and sinks from measurements of concentrations. We also consider a number of related inversion problems involving trace atmospheric constituents. This use of modelling is termed 'diagnostic' – the model is being used to interpret observations. The alternative use of models is in 'prognostic' operation, in which the model is used to make projections of future conditions.

Modelling of global atmospheric change has progressively widened its scope from the physical properties of the atmosphere to include atmospheric chemistry and biogeochemistry and is progressing to the currently emerging area of 'earth-system science'. This increase in scope has been motivated by the recognition of causal links between the components of the earth system. Realistic projections have to consider these connections and model their evolution in time. In contrast, diagnostic modelling is able to analyse components of the earth system, defining the linkages in terms of observations. Therefore, we can expect that inverse modelling in general (and inverse modelling of the atmosphere in particular) will become an increasingly important part of the development of earth-system science and the validation of earth-system models.

Recognition of the information present in global-scale spatial differences in concentration of CO_2 came soon after the establishment of high-precision measurement programmes at Mauna Loa (Hawaii) and the South Pole in 1958. The CO_2 records revealed a persistent difference and this mean spatial gradient has increased over the subsequent decades. Much of this difference is due to fossil-fuel use, which occurs mainly in the northern hemisphere. It is a measure of the difficulty of interpretation that there remain competing interpretations for the residual.

In order to achieve local air-quality objectives, many jurisdictions have established regulations controlling emissions. On a larger scale, cross-border transport of sulfur compounds has led to international agreements in some cases. On a global scale, the two objectives have been the control of ozone-depleting chemicals through the Montreal Protocol (see Box 16.3) and the restrictions on emission of greenhouse gases through the still-to-be-ratified Kyoto Protocol (see Box 6.1). The existence of these agreements creates a need to ensure that they are based on sound science, in order to ensure that the prescribed actions achieve the objectives of the agreements.

This book aims to capture my own experience and that of my colleagues in using atmospheric-transport modelling to help understand the global carbon cycle and other similar biogeochemical cycles. Since this activity is composed of so many interlinked parts, this introduction is designed to serve as a road-map to what lies ahead. The main division of this book is into Part A, which surveys general principles, and Part B, which reviews recent applications.

The components of tracer inversions are (i) a set of observations, (ii) an atmospheric-transport model and (iii) a set of mathematical/statistical techniques for matching observations to model results. This book is mainly about the matching process. It takes its context from the specific issues raised by the nature of atmospheric transport, the types of observations that are available and what we would like to learn about trace-gas fluxes.

Two important issues that we identify in developing practical inversion calculations are

- ill-conditioning, as introduced in Section 1.3, whereby the inversions are highly sensitive to errors and uncertainties in the inputs and assumptions; and
- the use of a statistical approach to the assessment of uncertainty.

1.2 Atmospheric inversion problems

As noted above, this book is divided into two parts, covering principles and applications, respectively. Nevertheless, principles need illustrative examples and most of the developments of techniques of trace-gas inversion have been in response to specific problems. The main classes of trace-gas inverse problem are the following.

Estimation of atmospheric transport. Inversion calculations to determine atmospheric transport have played a relatively small role in trace-gas studies. An exception is early studies of ozone as a tracer of atmospheric motion. A few tracer studies have concentrated on estimating key indices of transport, such as interhemispheric exchange times. Some of these are reviewed in Chapter 18.

Estimation of sources and sinks of halocarbons. Studies of the various halocarbons have mainly been motivated by their role in ozone depletion. Initially, studies of chlorofluorocarbons (CFCs) concentrated on estimating the loss rates, expressed in terms of atmospheric lifetimes. Studies of methyl chloroform (CH_3CCl_3), for which there are good concentration data and quite good estimates of emissions, also aim to estimate the loss rate. CH_3CCl_3 is removed from the troposphere by reaction with the hydroxyl radical (OH) and so the CH_3CCl_3 loss rate can characterise the loss by reaction with OH of other trace gases, particularly methane [377]. More recently, studies of halocarbons have attempted to estimate the strengths and locations of unreported emissions. Inversions of halocarbon distributions are discussed in Chapter 16.

Estimation of sources and sinks of CO_2. The key issue in studies of CO_2 is the atmospheric carbon budget and, in particular, the partitioning of CO_2 exchanges between oceanic and biospheric processes. Atmospheric CO_2 inversions aim to use the spatial distribution of CO_2 to infer the spatial distribution of surface fluxes, the objective being to obtain sufficient detail to distinguish terrestrial from ocean fluxes. (Note that this book uses the term flux to mean both (i) exchange of mass per unit area, generally in the context of partial differential equations, and (ii) area-integrated exchange of mass, in contexts involving finite areas.) CO_2 inversions are discussed in Chapter 14.

Estimation of sources and sinks of CH_4. As with CO_2, the important questions for CH_4 are those concerning the atmospheric budget. Consequently, the main atmospheric inverse problem is that of estimating the spatial distribution of methane fluxes, mainly from surface-concentration data. The sink in the free atmosphere is an additional complication. Methane inversions are discussed in Chapter 15.

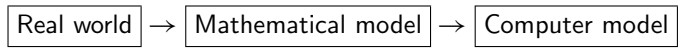

Figure 1.1 A schematic diagram of the relation among the real world, the mathematical model and the computer model. We adopt the terminology [423] of using 'validation' for testing the mathematical model against the real world and 'verification' for testing the computer model against the mathematical model.

Global-scale inversions of other trace gases are noted in Section 16.4 and regional-scale inversions are discussed in Chapter 17.

We consider the most common tracer inversion problem, that of deducing sources and sinks from concentration data. As noted above, this requires the use of a model of atmospheric transport. Figure 1.1 represents the relation among (i) the real world, (ii) a mathematical model of (some aspect of) the world and (iii) a computer implementation of the mathematical model. Identifying the mathematical model as an explicit intermediate step in model building allows us to use a wide range of mathematical techniques to analyse the modelling process. Much of this book is written in terms of such mathematical models.

The general mathematical form of the transport equation for a trace constituent describes the calculated rate of change with time of $m(\mathbf{r}, t)$, the (modelled) atmospheric concentration:

$$\frac{\partial}{\partial t} m(\mathbf{r}, t) = s(\mathbf{r}, t) + \mathcal{T}[m(\mathbf{r}, t), t] \tag{1.2.1}$$

where $s(\mathbf{r}, t)$ is the local source and $\mathcal{T}[.,.]$ is a transport operator. Equation (1.2.1) expresses the rate of change of a trace-gas concentration at a point, $\mathbf{r}$, and time, t, as the sum of the net local source-minus-sink strength at that point, plus a contribution due to trace-gas transport from other locations. The transport is usually modelled with an advective component, $\nabla \cdot (\mathbf{v}m)$, often with a diffusive component to represent sub-grid-scale processes.

We can identify two main classes of inversion, which we denote 'differential' and 'integral'. The former works with equation (1.2.1). The latter uses Green's functions obtained by (numerical) integration of the transport equations. There are also various 'hybrid' techniques.

(a) **Differential inversions.** These are based on rewriting the transport equation (1.2.1) as

$$\hat{s}(\mathbf{r}, t) = \frac{\partial}{\partial t} \hat{m}(\mathbf{r}, t) - \mathcal{T}[\hat{m}(\mathbf{r}, t), t] \tag{1.2.2}$$

where $\hat{s}$ and $\hat{m}$ denote statistical estimates. The most common application is deducing surface sources from surface observations, so (1.2.2) is used at surface grid points, while (1.2.1) is numerically integrated throughout the free atmosphere. Equation (1.2.2) is applied with $\hat{m}(\mathbf{r}, t)$ as a statistically smoothed version of the observed concentration field, $c(\mathbf{r}, t)$ (hence the notation $\hat{m}$). This technique is described as a 'differential' form because of the $(\partial/\partial t)\hat{m}$ term – it

is often referred to as the 'mass-balance' technique since the transport equations both in the original and in transformed forms are expressing local conservation of mass. Mass-balance inversion techniques are reviewed in Section 11.1.

(b) **Green-function methods.** These are expressed formally through the Green function, $G(\mathbf{r}, t, \mathbf{r}', t')$, relating modelled concentrations, $m(\mathbf{r}, t)$, to source strengths, $s(\mathbf{r}, t)$,

$$m(\mathbf{r}, t) = m_0(\mathbf{r}, t) + \int_{t_0}^{t} G(\mathbf{r}, t, \mathbf{r}', t') s(\mathbf{r}', t') \, \mathrm{d}^3 r' \, \mathrm{d}t' \tag{1.2.3}$$

where $m_0(\mathbf{r}, t)$ describes the way in which the initial state, $m(\mathbf{r}, t_0)$, evolves in the absence of sources. Of necessity, actual calculations are performed using some discretisation of (1.2.3). This is expressed as the generic relation

$$c_j = \sum_{\mu} G_{j\mu} s_\mu + \epsilon_j = m_j + \epsilon_j \tag{1.2.4}$$

where c_j is an item of observational data, m_j is the model prediction for this item of data, ϵ_j is the error in c_j, s_μ is a source strength and $G_{j\mu}$ is a discretisation of $G(\mathbf{r}, t, \mathbf{r}', t')$.

The discretisation is based on decomposing the sources as

$$s(\mathbf{r}, t) = \sum_{\mu} s_\mu \sigma_\mu(\mathbf{r}, t) \tag{1.2.5}$$

so that the $G_{j\mu}$ are the responses (for observation j) to a source defined by the distribution $\sigma_\mu(\mathbf{r}, t)$. (Often, for convenience, $G_{j\mu}$ includes pseudo-sources defining the m_0 of (1.2.3), which is assumed to be constant for each species.) The sources are estimated by using (1.2.4) to fit the coefficients, s_μ. For this reason, these Green-function methods that work in terms of pre-defined components, $\sigma_\mu(\mathbf{r}, t)$, have been termed 'synthesis' calculations [165], since the source estimate is synthesised from these pre-defined components.

The most important point for the development of Green-function methods is that (1.2.1) defines a linear relation between the concentrations, $m(\mathbf{r}, t)$, and the sources, $s(\mathbf{r}, t)$, so the full machinery of linear algebra can be applied to solving (1.2.4). Green-function techniques are discussed in Chapter 10.

(c) **Hybrid techniques.** These techniques lie between the differential (mass-balance) and the integral (synthesis) inversions. Generally, they take the form of synthesis inversions over a sequence of relatively short time-steps. Examples of this are the techniques of Brown [56], Hartley and Prinn [197] and Ramonet *et al*. [394]. These and other similar techniques are reviewed in Section 11.2. In addition, there is an exploratory discussion in Chapter 12 of techniques involving non-linear estimation.

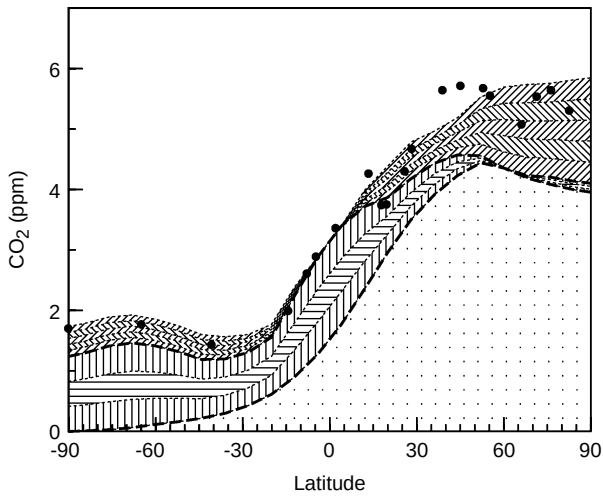

Figure 1.2 A schematic representation of synthesis inversion as analogous to a jigsaw, fitting an unknown number of differently shaped components (fossil, ocean and terrestrial) to find the best fit to observations (solid points).

The emphasis given to Green-function, or synthesis, techniques in this book primarily reflects the scope for error analysis. This also underlies the second reason, which is the extensive experience of synthesis inversions in our research group. In its simplest form, the synthesis approach corresponds to multiple regression: a function $c(x)$ is expressed as a linear combination of specified functions, $G_\mu(x)$, in terms of unknown coefficients, s_μ, by fitting a set of observations at points x_j as $c(x_j) \approx \sum_\mu s_\mu G_\mu(x_j)$.

A visual illustration of the technique can be obtained by regarding the fitting process as a 'jigsaw'. Figure 1.2 gives a schematic representation of the regression (in terms of latitudinal variation only) for CO_2 distributions. Rather than fit pieces of unknown size but known shape, the 'jigsaw' analogy approximates this by fitting an unknown number of pieces of fixed shape and size, shown by alternating hatching. Figure 1.2 demonstrates fitting an unknown number of land pieces (five in this example, above the upper dashed line and shown with diagonal hatching) and an unknown number of ocean pieces (three in this case, between the dashed lines) plus a fairly well-known fossil piece (below the lower dashed line with dot fill) to fit the observed spatial distribution (solid points). (Note that the higher concentrations in the 'land' pieces at high southern latitudes reflect the transport of northern air southwards through the upper troposphere.)

1.3 Uncertainty analysis

A key focus of this book is the estimation of uncertainties. It is particularly important in ill-conditioned problems that are subject to large error-amplification. Uncertainty analysis is required on general grounds of scientific integrity and the needs of policy-related science, as well as for input into further levels of scientific (or policy-related) analysis. In addition, as described in Chapter 13, we have used a systematic uncertainty analysis as the basis of experimental design.

The underlying principle is that *any statistical analysis requires a statistical model.* Generally we need to *assume* such a model. We can test (but never prove) the validity of the assumptions. In discussing the various types of error that can affect trace-gas inversions, Enting and Pearman [141] noted that "any variability that cannot be modelled $[\cdots]$ must be treated as part of the 'noise' $[\cdots]$". In other words, the noise in the statistical model is whatever is not being modelled deterministically. Statistical estimation is described in Chapter 3; the special case of time series is described in Chapter 4.

This book follows Tarantola [471] in being firmly based on the use of prior information. This is both an optimal use of available information and an essential part of stabilising ill-conditioned problems. The standard Bayesian formalism has proved adequate for the problems that we have encountered in practice, without the need to adopt the extensions proposed by Tarantola (see Box 12.1).

The emphasis on the statistical modelling of uncertainty leads us to express the results from inversion calculations in the terminology of statistical estimation. The results of inverting (1.2.4) are *estimates*, denoted $\hat{s}_\mu$, of the source components, s_μ, or more generally estimates, $\hat{x}_\mu$, of parameters, x_μ. As noted above, the essential requirement for any statistical analysis is that one must have a statistical model of the problem.

The word 'model' has been used both for the transport model and for the statistical model. This multiple usage needs to be recognised since both types of model are needed for tracer inversions. The transport model represents a deterministic relation between the sources and the concentrations. However, our overall knowledge of the sources and concentrations is incomplete. Most obviously, our knowledge is not infinitely precise. This incompleteness in our knowledge is expressed in statistical terms. With this terminology, the quote from Enting and Pearman [141] needs to be reworded as *any variability that cannot be modelled deterministically* $[\cdots]$ *must be treated as part of the 'noise' and modelled statistically.*

In order to address these issues of uncertainty, inversion studies need to use an overall model with both deterministic and statistical aspects. A common form of the combined model is one in which the statistical aspects appear as noise added to the outputs of a deterministic model and (when one is using a Bayesian approach) finite uncertainties on the inputs. With such a structure, there can be an apparent distinction between the deterministic (transport) model and the statistical model. The deterministic model never 'sees' the statistical model – all the statistical analysis occurs somewhere outside. Conversely, the statistical model sees the deterministic model as a functional relation and need take no account of the immense complexity that may lie inside a functional representation of atmospheric transport. This convenient separation between deterministic and statistical models becomes rather less tenable when we wish to apply a statistical approach to considering uncertainties in the deterministic model itself. State-space modelling (see Section 4.4) provides one framework in which statistical and deterministic aspects of modelling can be integrated.

The simplest statistical model of the observations is to assume independent normally distributed errors. For the case of linear relations between observations and

parameters, this leads to a weighted least-squares fit giving the optimal (minimum-variance) estimates. Conversely, adopting a least-squares fitting procedure and associated error analysis is equivalent to assuming, whether implicitly or explicitly, that the errors are normally distributed. Errors with a multivariate normal distribution lead to a weighted least-squares fit with 'off-diagonal' weights (see Section 3.3) as the optimal estimates. Other error distributions (and associated non-linear estimation) have been considered in a highly simplified zonally averaged inversion [130]. Additional discussion of non-linear estimation is given in Chapter 12.

Many of the inversions use a Bayesian approach, i.e. independent prior estimates of the sources are included in the estimation procedure. Detailed discussions of applications of Bayesian estimation in carbon-cycle studies have been given for global model calibration [140], for synthesis inversion [143, 145], for methane [242, 206] and in subsequent work.

Some of the key aspects of error analysis are the following.

Measurement error. Most error analyses in trace-gas studies have assumed that the errors in the various observations, c_j, are independent. To the extent that 'error' includes the effect of small-scale sources that are omitted from the model, the assumption of independence of distinct sites can easily fail. Probably, a more important omission is the time correlation in the errors for a single site. Most inversion studies have ignored this problem. Two early exceptions are the three-dimensional model study reported by Bloomfield [34], in which an autoregressive error model was used, and the synthesis inversion by Enting *et al*. [145], in which the issue of autocorrelation in the data was avoided because time-dependence was expressed in the frequency domain.

Model error. The problem of determining the effect of model error remains largely unsolved. The difficulty is particularly great in ill-conditioned inverse problems with their large sensitivity to errors. Tarantola [471] describes a formalism in which model error becomes an extra component added to observational error (see equation (9.1.2)). Enting and Pearman [141] considered such a formalism with particular reference to the 'truncation error' when the small-scale degrees of freedom are excluded from the process of synthesis inversion (see also Section 8.3). One difficulty with this approach is that these errors are unlikely to be independent and there is little basis for defining an appropriate error covariance.

Numerical modelling involves an initial discretisation of the spatial and temporal variations both of sources and of concentrations. Further discretisation of source distributions may be needed because of the limited information contained in a sparse observational network and the loss of information associated with the ill-conditioning of the inversion. The 'synthesis-inversion' approach defined by equation (1.2.4) is usually based on a coarse discretisation of source/sink processes.

In recent years there has been a collaborative project of the International Geosphere–Biosphere Program (IGBP) known as TransCom, which compares some of the atmospheric-transport models used to study CO_2 [398, 277; see also Section 9.2 and Box 9.1]. These studies have confirmed the importance of the problem but have not yielded a 'magic-bullet' solution. A further aspect of 'model error' that must be considered is that of errors in the statistical model.

Source statistics. The Bayesian approach requires, as one of its inputs, prior statistics of the source strengths. One of the most critical issues is the time correlation of these prior source estimates in time-dependent Bayesian inversions. Initially, Rayner *et al*. [402] used time pulses whose prior distributions were assumed to be independent. Later calculations (e.g. those in Figure 14.9) used 'mean-plus-anomaly' representation of data error and prior estimates. Mulquiney *et al*. [332] used a random-walk model for the prior statistical characterisation of sources.

In addition, consideration of the spatial statistics of the sources is required in order to assess the discretisation error inherent in the synthesis approach. The particular importance of spatial statistics is quite explicit for inversions using adjoint calculations [240], in which very large numbers of source components are involved. Assumptions of independent uncertainties for a large number of small regions could imply an unrealistically small uncertainty in the total. Similar issues of spatial statistics are implicit in synthesis inversions based on a small number of highly aggregated components.

The process of deducing trace-gas fluxes from concentration data is severely hampered by a mathematical characteristic known as ill-conditioning. Figure 1.3 gives a schematic representation of the source of the difficulty: dissipative processes in the real world lead to a loss of detail in the information available for analysis. In cases in which the model calculations (or, more precisely, the model-based inferences) are in the opposite direction to the real-world chain of causality, the attenuation of detailed

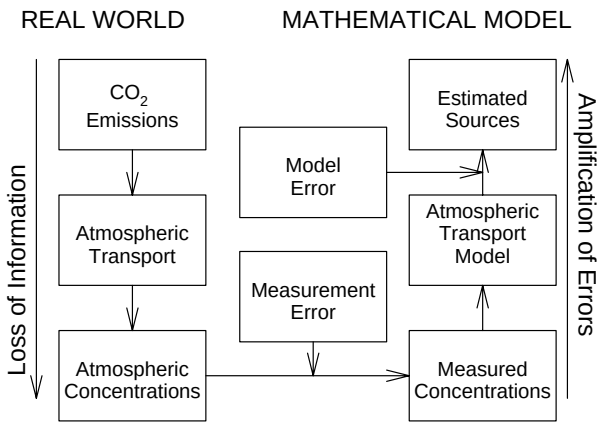

Figure 1.3 The origin of ill-conditioning in the tracer inversion problem. Figure from [132].

information requires a corresponding amplification to try to reconstruct the source distribution. This will also amplify errors introduced by the observational process and errors in the transport model.

The loss of information in ill-conditioned inverse problems can be quantified in terms of how rapidly the attenuation in the forward problem changes as the length-scale decreases. In terms of an inverse length-scale specified by a wave-number, k, the low-frequency response of the surface concentration to a surface flux behaves as k^{-1}. The asymptotic behaviour of global-scale transport can be (and was) derived from an analytical purely diffusive model, because the limiting behaviour is determined by the most dissipative process. Calculations with numerical models show that the k^{-1} behaviour applies quite accurately for $k > 1$ [132, 136]. The ill-conditioned nature of the atmospheric-tracer inversion problem has been known since the work of Bolin and Keeling [37], although the k^{-2} response that they found applies to vertically averaged concentrations rather than surface concentrations.

Similar ill-conditioned inverse problems are common in many areas of the earth sciences. Much of our work has drawn on analogies from seismology, particularly the network design study [400] noted in Chapter 13. Chapter 8 revisits these issues of ill-conditioning. For the present, the important point is that the limited ability to recover details from indirect information is an important reason for using Bayesian estimation.

1.4 Toy models

This book makes frequent use of 'toy models' to illustrate aspects of tracer inversion. Toy models are highly simplified models that capture only a few important attributes of a system. The value of these models comes from the insights that can be obtained from analytical or semi-analytical solutions and/or the ability to explore solutions for a large number of different conditions quickly. For example, identifying the sensitivities to key model parameters is an important application of toy models. In this book, one of the most important applications of toy models is to illustrate the differences between forward and inverse modelling with various types of model.

In addition, toy models may often give useful estimates of uncertainty. For ill-conditioned inverse problems, estimates of the form $\hat{x} = \sum a_j c_j$ will frequently involve cancellations and so accurate estimates require accurate values of the 'inverse-model' coefficients, a_j. In contrast, for independent data, errors of the form $\operatorname{var} \hat{x} = \sum |a_j|^2 \operatorname{var} c_j$ involve sums without cancellations and so are much less sensitive to errors and approximations in the model.

There are five classes of toy model that are followed in a sequence of boxes and exercises throughout the book, refining the models and/or the analysis, to elucidate aspects of the general theoretical treatment. We classify them as three groups of toy transport model, a group of statistical models and a class of toy chemistry model. Suggested values for the model parameters are given in Appendix B.

Box 1.1. **Toy transport model A.1. The one-reservoir atmosphere**

In representing the atmosphere as a small number, N, of reservoirs, we start with $N = 1$. Transport is irrelevant and the only changes in concentration are due to the net effect of sources and sinks.

We write the rate of change of concentration as

$$\frac{\mathrm{d}}{\mathrm{d}t}m = \lambda(t)m(t) + s(t) \tag{1}$$

where $s(t)$ is an 'external' source and $-\lambda$ represents an atmospheric decay rate (or inverse 'lifetime') for a decay process, such as chemical reaction or radioactive decay.

For a specified initial condition, $m(t_0)$ at time t_0, equation (1) has the formal solution

$$\begin{aligned} m(t) &= m(t_0)\exp\left(\int_{t_0}^{t}\lambda(t')\,\mathrm{d}t'\right) \\ &\quad + \exp\left(\int_{t_0}^{t}\lambda(t')\,\mathrm{d}t'\right)\int_{t_0}^{t}\exp\left(\int_{t_0}^{t''}-\lambda(t')\,\mathrm{d}t'\right)s(t'')\,\mathrm{d}t'' \\ &= m(t_0)\xi(t) + \int_{t_0}^{t}\xi(t)/\xi(t')s(t')\,\mathrm{d}t' \end{aligned} \tag{2}$$

where

$$\xi(t) = \exp\left(\int_{t_0}^{t}\lambda(t')\,\mathrm{d}t'\right) \tag{3}$$

is termed an integrating factor. Relation (2) is a special case of the Green-function formalism

$$m(t) = m_0(t) + \int_{t_0}^{t}G(t,t')s(t')\,\mathrm{d}t' \tag{4}$$

(see Section 10.1) whereby an inhomogeneous differential equation is 'solved' in terms of an integral operator that is the inverse of the differential operator.
For constant λ, (2) reduces to

$$m(t) = m(t_0)\mathrm{e}^{(t-t_0)\lambda} + \int_{t_0}^{t}\mathrm{e}^{(t-t'')\lambda}s(t'')\,\mathrm{d}t'' \tag{5}$$

A. **Few-reservoir representations of atmospheric transport.** These models allow us to look at broad-scale features at a low level of discretisation. From a mathematical perspective, the models are defined by sets of ordinary differential equations (ODEs). This simplifies some of the analyses.

B. **Diffusive transport models.** At the opposite extreme to the highly discretised models, the purely diffusive model uses a continuum view of the atmosphere. Tracer distributions are modelled with partial differential equations (PDEs).

Box 1.2. **Toy transport model B.1. The diffusive atmosphere**

The first representation of an atmosphere with purely diffusive transport distorts the geometry in order to simplify the mathematics. We consider anisotropic diffusion in a rectangular region. Schematically it represents a zonal average of the atmosphere. The flux across the lower boundary is specified in terms of a flux-gradient relation and zero flux is prescribed on the other three boundaries. The lower boundary is at $y = 1$ and the upper boundary is at $y = 0$ so that y is acting like a pressure coordinate and x is analogous to $0.5[1 + \sin(\text{latitude})]$.

The diffusion equation is

$$\frac{\partial m}{\partial t} = \kappa_x \frac{\partial^2 m}{\partial x^2} + \kappa_y \frac{\partial^2 m}{\partial y^2} \tag{1}$$

in the domain $x \in [0, 1]$ and $y \in [0, 1]$ and subject to

$$\frac{\partial m}{\partial x} = 0 \quad \text{at } x = 0 \text{ and } x = 1$$

$$\frac{\partial m}{\partial y} = 0 \quad \text{at } y = 0 \qquad \text{and} \qquad \frac{\partial m}{\partial y} = s(x) \quad \text{at } y = 1$$

For the steady-state case, the boundary conditions imply solutions of the form

$$m(x, y) = \sum_{n=0}^{\infty} m_n \, \Xi_n(x, y) \tag{2a}$$

with

$$\Xi_n(x, y) = \cos(n\pi x) \cosh(\gamma_n y) \tag{2b}$$

and the differential equation implies

$$\kappa_x n^2 \pi^2 - \kappa_y \gamma_n^2 = 0 \quad \text{whence} \quad \gamma_n = n\pi \sqrt{\kappa_x / \kappa_y} \tag{2c}$$

From the boundary conditions, the sources are

$$s(x) = \sum_n s_n \, \Xi_n(x, 1) \quad \text{with} \quad s_n = \gamma_n \tanh(\gamma_n) \, m_n$$

The $m_n \sim s_n / n$ attenuation illustrated here remains a common feature of the problem of deducing surface sources from surface data as the diffusive model is refined (Box 2.2) and also characterises realistic advective–diffusive models [136, 132].

This model is of historical interest because of its early use (1963), averaged over height and longitude, by Bolin and Keeling [37]. However, the greatest importance of this model is that it gives a realistic estimate of the rate at which information about small-scale details of the fluxes is attenuated by atmospheric mixing.

Box 1.3. **Toy transport model C.1. An advective 'atmosphere'**

The model domain is from $x = 0$ to $x = 1$ and $y = 0$ to $y = 1$. The transport is purely advective and is expressed as a time-varying stream function:

$$\chi = \sin(\pi y)\,[\sin(2\pi x) + \alpha \sin(\pi x)\cos(\omega t)] \tag{1}$$

This represents a system of two counter-rotating cells with a time-varying modulation of the relative sizes of the cells.

The transport in this system is not amenable to analytical solution – indeed the purpose of this toy model is to illustrate how simple time-varying flow fields can lead to chaotic transport of matter.

The stream function, $\chi(x, y)$, defines the velocity components as

$$v_x = \frac{\partial \chi}{\partial y} = \pi \cos(\pi y)\,[\sin(2\pi x) + \alpha \sin(\pi x)\cos(\omega t)] \tag{2a}$$

$$v_y = -\frac{\partial \chi}{\partial x} = -\pi \sin(\pi y)\,[2\cos(2\pi x) + \alpha \cos(\pi x)\cos(\omega t)] \tag{2b}$$

This transport formalism can be used in two ways, either in a Lagrangian mode to advect individual particles with velocity $[v_x, v_y]$ or in an Eulerian mode to describe the evolution of a concentration field, $m(x, y, t)$, as

$$\frac{\partial}{\partial t} m(x, y, t) = -\frac{\partial}{\partial x}(v_x m) - \frac{\partial}{\partial y}(v_y m) = \frac{\partial \chi}{\partial x}\frac{\partial m}{\partial y} - \frac{\partial \chi}{\partial y}\frac{\partial m}{\partial x} \tag{3}$$

C. **Atmospheric transport modelled by advective-stirring.** This is also presented as a continuum view of the atmosphere. The transport representation is the opposite of that in model B: model C is purely advective, whereas model B is purely diffusive. The role of model C is to explore the statistics of chaotic processes and the conditions under which chaotic transport can appear as diffusive.

D. **Statistics of 'signal plus noise'.** These models are used to illustrate the underlying statistical principles. The common framework is one of a deterministic signal plus random noise. The standard problem is that of estimating the signal in the presence of the noise. Initially the 'signal' is simply a mathematical function. Later refinements of the model incorporate deterministic components specified by the 'toy' transport models.

E. **Toy chemistry.** This is a 'whole-atmosphere' representation of the CH_4–CO–OH balance, with lumped production/loss rates characterising all other reactions.

Box 1.4. Toy statistical model D.1. Linear trend plus white noise

The sequence of statistical models is of the general form signal plus noise. We start with N data values, c_n for $n = 1$ to N modelled with a simple signal: a linear trend and simple noise: independent normally distributed white noise, ϵ_n, with zero mean and known variance, Q. Thus the statistical model is

$$c_n = \alpha + n\beta + \epsilon_n \tag{1}$$

with $E[\epsilon_n] = 0$ and $E[(\epsilon_n)^2] = Q$.

Ordinary least-squares (OLS) estimates of α and β are obtained by minimising

$$J_{\mathrm{OLS}} = \sum (c_n - \alpha - n\beta)^2 \tag{2}$$

giving the estimates

$$\hat{\alpha} = \frac{\sum(n^2)\sum c_n - \sum n \sum(nc_n)}{N\sum(n^2) - (\sum n)^2} \tag{3a}$$

$$\hat{\beta} = \frac{N\sum(nc_n) - \sum n \sum c_n}{N\sum(n^2) - (\sum n)^2} \tag{3b}$$

Using (1), these can be rewritten as

$$\hat{\alpha} = \alpha + \frac{\sum(n^2)\sum \epsilon_n - \sum n \sum(n\epsilon_n)}{N\sum(n^2) - (\sum n)^2} \tag{4a}$$

$$\hat{\beta} = \beta - \frac{\sum n \sum \epsilon_n - N\sum(n\epsilon_n)}{N\sum(n^2) - (\sum n)^2} \tag{4b}$$

which emphasises that, once the data are modelled as random variables, estimates derived from these data will also be random variables. Since the ϵ_n have zero mean and appear linearly in (4a) and (4b), the estimates $\hat{\alpha}$ and $\hat{\beta}$ are unbiased, i.e. $E[\hat{\alpha}] = \alpha$ and $E[\hat{\beta}] = \beta$, where $E[.]$ denotes the mean over the error distribution, i.e. the expected value averaging over an arbitrarily large ensemble of realisations of the random noise process, ϵ_n.

Equations (4a) and (4b) also give the starting point for calculating the variance of these estimates as $E[(\hat{\alpha} - \alpha)^2]$ and $E[(\hat{\beta} - \beta)^2]$ (problem 2 of this chapter) as well as the covariance, $E[(\hat{\alpha} - \alpha)(\hat{\beta} - \beta)]$.

This regression can also be solved by a recursive technique that goes back to C. F. Gauss [525]. This is a special case of the Kalman filter (see equations (11.2.3a)–(11.2.3c) and Box 4.4).

Box 1.5. **Toy chemistry model E.1**

Several workers have used highly simplified models of the major balance of tropospheric chemistry among methane (CH_4), carbon monoxide (CO) and the hydroxyl radical (OH). These were expressed in terms of the global totals, m_{CH_4}, m_{CO} and m_{OH} of CH_4, CO and the OH radical. Several of the models are special cases of the general form

$$\frac{d}{dt}m_{CH_4} = -k_1 m_{CH_4} m_{OH} - \lambda_{CH_4} m_{CH_4} + S_{CH_4} \tag{1a}$$

$$\frac{d}{dt}m_{CO} = -k_2 m_{CO} m_{OH} + \zeta k_1 m_{CH_4} m_{OH} - \lambda_{CO} m_{CO} + S_{CO} \tag{1b}$$

$$\frac{d}{dt}m_{OH} = -k_1' m_{CH_4} m_{OH} - k_2' m_{CO} m_{OH} - \lambda_{OH} m_{OH} + S_{OH} \tag{1c}$$

where k_1 and k_2 are the rate constants for the reactions for oxidation (by OH) of CH_4 and CO, respectively, and using $k_1' = k_1/\xi$ and $k_2' = k_2/\xi$ allows m_{OH} to be expressed in mm^{-3} and equation (1c) to have time units of seconds. The factor ζ represents the proportion of oxidation of CH_4 that generates CO. For CH_4, CO and OH, S_{CH_4}, S_{CO} and S_{OH} are the respective rates of production and λ_{CH_4}, λ_{CO} and λ_{OH} are the respective loss rates, in each case excluding the reactions described by k_1 and k_2. Because of its very short lifetime, the concentration of OH can be treated as being in a steady state defined by

$$m_{OH} = \frac{S_{OH}}{\lambda_{OH} + k_1' m_{CH_4} + k_2' m_{CO}} \tag{2}$$

Guthrie [186] considered the case $\zeta = 1$, $\lambda_{OH} = \lambda_{CO} = 0$ and found that the system was unstable unless $2S_{CH_4} > (\xi S_{OH} - S_{CO}) > 0$.

Prather [366] considered the case $\lambda_{CH_4} = \lambda_{CO} = 0$, $\zeta = 1$ in his analysis of adjustment times for methane perturbations.

Krol and van der Woerd [265] considered the case $\lambda_{OH} = 0$ and $\zeta = 0.8$, with S_{CO} including indirect sources from non-methane hydrocarbons.

The units that we use for numerical examples generally follow those used in the IMAGE-2 description [265], time is in years, so that the λ_η are in yr^{-1}. The concentrations m_{CH_4} and m_{CO} are in ppm while, unlike the IMAGE units, we use m_{OH} in mm^{-3}. Thus k_1 and k_2 are in $mm^3\ yr^{-1}$. The sources are expressed as in IMAGE in terms of emissions in mass units per year multiplied by conversion factors of 0.202×10^{-3} ppm/(Tg CO) and 0.353×10^{-3} ppm/(Tg CH_4).

Further reading

As I noted in the preface, one of the joys and difficulties of studying biogeochemical cycles is the need to know about so many fields of science. The following list of suggested reading reflects my own experience of what has been useful. In most cases the references are chosen for depth rather than breadth.

Atmospheric chemistry. A comprehensive account of atmospheric chemistry has recently been produced by Brasseur *et al*. [51]. Although inverse problems are given only a brief discussion, many of the chapters parallel some of the 'background' sections of the present book. In particular, the chapter *Atmospheric dynamics and transport* [168] gives an account that goes well beyond the overview in Chapter 2, *Observational methods* [301] expands on Section 5.1 and *Modeling* [50] goes beyond Chapter 2. Additional overview accounts of atmospheric chemistry are given in the IPCC assessments [369, 370] and WMO ozone assessments [516].

Atmospheric circulation and dynamics. There are many books. For the purposes of understanding tracer distributions, that of James [230] gives a good balance between the description of the circulation and the dynamical processes that cause it. The books by Holton [212], Gill [177] and Green [184] are notable examples of books on dynamical meteorology.

Bayesian statistics. The classic textbook on Bayesian estimation is Box and Tiao [47]. As an older book, Jeffreys [231] has the interest of being written from a more defensive position, taking note of the foundations of mathematics and theories of knowledge. It justifies the Bayesian formalism as being that which answers the questions that scientists actually ask. Tarantola [471] takes the Bayesian approach as a necessary 'given' and extends the formalism (see Box 12.1).

Biogeochemical cycles. Schlesinger [424] reviews biogeochemistry firstly in terms of processes and then by considering the respective cycles of water, carbon, nitrogen, phosphorus and sulfur. Broecker and Peng [54] consider the carbon cycle with an emphasis on the role of the oceans. Lovelock [292] introduced an entirely different perspective with his 'Gaia hypothesis', proposing that the biogeochemical system is self-regulating, with living organisms acting to provide negative feedbacks that maintain the earth in conditions suitable for maintaining life.

Carbon cycle. The global carbon cycle is an active area of research, since CO_2 is the most important of the anthropogenic greenhouse gases. The assessments by the Intergovernmental Panel on Climate Change (IPCC) include assessments of the state of science for the greenhouse gases. CO_2 and the carbon cycle have been reviewed in the Radiative Forcing Report [422] and, most recently, in the Third Assessment Report [371]. Several volumes from 'summer schools', e.g. [200, 507], provide valuable introductions to the field.

Computer programming. Computer-programming techniques have evolved considerably since the days when I initially trained in Fortran and Cobol. I have found Michael Jackson's book [225] to give a lot of valuable concepts about program structure. In addition, *Elements of Programming Style* [255] gives a number of rules (with informed justification), of which my favourite is 'Make it right before you make it faster'.

Earth-system modelling. The 'earth-system' view goes back at least to the NASA report [334]. Harvey [198] has written a textbook that uses simple modelling to quantify influences contributing to global change. Integrated assessment models tend to include at least the faster-acting components of the earth system, although often in a highly parameterised manner. The IMAGE model volume [4] describes one such model. The volume by Martens and Rotmans [306] covers both model formulations and the implications of model-based decision making.

Inverse problems. For a comprehensive presentation of the mathematical formulation of inverse problems, Tarantola's book [471] stands out. In the same way as that in which all western philosophy has been described as footnotes to Plato, the whole of the present book can be regarded as footnotes to Tarantola – identifying the specific quantities to insert into one or other of Tarantola's estimation techniques to solve a biogeochemical inverse problem.

Numerical techniques. The book *Numerical Recipes* by Press *et al*. [373] has become extremely popular and editions are now available for Fortran-77, C, Pascal and Fortran-90. In this case, the coverage reflects breadth rather than depth. For depth, the 'references therein' provide a starting point. There has been a series of discussions on the internet, criticising the quality of some of the algorithms. My overall assessment is that, generally, using a 'numerical recipe' is better for a non-specialist than writing your own routine from an abstract mathematical formulation. Certainly they should be adequate for the exercises in this book. For problems that are large, or otherwise difficult, routines from a specialised mathematical subroutine library may be needed. Acton's book [1] *Numerical Methods That Work* gives advice on pitfalls in some common calculations. For the specific area of numerical optimisation applied to inverse problems, Tarantola [471] describes the main algorithms.

Time-series analysis. There are many books on time-series analysis. Priestley's book [374] stands out for comprehensiveness, although achieving such coverage in 'only' 890 pages makes for a generally compact presentation. Even within the field of atmospheric science, there are many books on time-series analysis; that by von Storch and Zwiers [455] is both comprehensive and recent. In the specialized area of Kalman filtering Gelb's book [173] is a classic. Young's book [525] is also a good introduction to recursive estimation.

Spatial statistics. Cressie's book [84] is the best general reference that I know of for spatial statistics. Techniques relevant to climate research are covered by von Storch and Zwiers [455]. Kagan [238] describes the statistical aspects of

averaging observational data. Mandelbrot [300] provides something entirely different. First, there is the style (which he describes as a scientific essay: dealing with a subject from a personal point of view). Secondly, there was the highly innovative topic of self-similar probability distributions (fractals), with arguments for the ubiquity of such distributions in nature.

Atmospheric tracer inversion. Writing this book was motivated by the lack of books covering this field. There are, however, some review papers: Mulquiney *et al.* [331] give a summary of the main inversion techniques, with a little background on applications; Prinn and Hartley [376] concentrate on state-space modelling; and Heimann and Kaminski [203] give a more recent overview. The AGU monograph [244] from the Heraklion workshop contains a number of articles that review particular inversion techniques [375, 133, 176, 429, 97, 58, 20, 449]. In addition there are several other descriptions that focus on applications of tracer inversion [430, 460, 356]. The present book grew from my own contribution to the Heraklion conference [133]. My aims in writing this book have been the following: to achieve a more unified presentation; to emphasise the inversions as problems in statistical estimation; and to explore the boundaries of current practice as a guide to possible future developments.

Others. A number of other books are important references for topics less closely related to tracer inversions. Morgan *et al.* [324] discuss uncertainty, including aspects of uncertainty that have little relevance to atmospheric science but very great relevance to policy-related analyses. Daley's book gives a comprehensive coverage of the assimilation of meteorological data into operational weather-forecasting models [94]. Trenberth's account of climate modelling [485] is a comprehensive introduction. The *Concise Encyclopedia of Environmental Systems* [526] is a useful reference to a range of statistical techniques.

Applied mathematics. Finally, in the case of 'applied mathematics', rather than selecting particular books, I will try to identify the level of mathematics required. The key requirements are a knowledge of vectors and matrices, the calculus of vector fields and an understanding of probability distributions.

In most cases, additional suggestions for further reading are given at the ends of the chapters.

Notes on exercises

Exercises illustrating various aspects of the presentation are included at the end of each of Chapters 1–13, i.e. those chapters dealing with techniques. Many of them are mathematical in nature. For those that require numerical values, Appendix B gives values of real-world constants and toy-model parameters. It also gives information

about electronic access to observed concentration data and emission inventories. Other exercises involve writing simple computer programs, particularly to implement the Kalman filter, at least for the special case of recursive regression (see Box 4.4). Other useful generic components are a Monte Carlo inversion unit and a singular-value-decomposition (SVD) routine. Routines for SVD (and its use in regression analysis) are included in *Numerical Recipes* [373] and these versions are included in IDL (Interactive Data Language, from Research Systems Inc.). In a class situation, instructors may be able to provide such components.

Exercises for Chapter 1

1. By how much does the annual burning of 6 Gt (1 Gigatonne $= 10^{15}$ g) of fossil carbon change the mass of the atmosphere if 50% of the carbon remains in the atmosphere and the rest is taken up by the oceans?
2. Use the expressions in Box 1.4 to obtain an expression (valid for large N) for the variance of the estimated trend β, in terms of R, the variance of the ϵ_n. How much does this change if (as is suggested for monthly mean CO_2 data – see Section 5.4.3) the covariance of consecutive errors is $R/2$?
3. (Numerical problem, no solution given) Write a computer program (preferably with graphical display) to implement two or more particles advected according to toy model C.1 of Box 1.3. Note the pattern of residence times in each hemisphere. If multiple colours can be displayed, simulate the evolution of an initial colour gradient.