

# INVERSE PROBLEMS IN GREENHOUSE GAS MODELLING

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## 1. INTRODUCTION

The concentrations of radiatively active gases, including  $\text{CO}_2$ ,  $\text{N}_2\text{O}$  and  $\text{CH}_4$ , in the atmosphere have been steadily increasing, as indicated by both direct observations and measurements of gas concentrations in air bubbles trapped in polar ice. The influence of these gases on the earth's radiation budget is predicted to change the earth's climate by an amount greater than any change in recorded history. Because of these effects, the study of the sources and sinks of these gases has taken on considerable importance.

The biogeochemical cycles involving these so-called 'greenhouse gases' are of considerable complexity and have been the subject of many investigations. However much of the information that is obtainable gives only indirect measures of the key processes involved. In mathematical terms, the consequence of this reliance on indirect information is that many modelling studies of greenhouse gases involve ill-conditioned inverse problems. In only a small proportion of such studies has the associated inverse problem been analysed in any detail [1], [2].

The purpose of this report is to draw attention to a number of inverse problems in the area of greenhouse gas modelling, in the hope of stimulating further study in this area. Many of the examples are expressed in terms related to ODEs rather than the PDEs which are the main topic of this volume. However, it must be appreciated that such 'ODE' formulations are almost always 'lumped' representations of multi-dimensional systems whose properties are described in terms of PDEs, particularly those representing

advective and diffusive transport.

## 2. THE ATMOSPHERIC CO<sub>2</sub> BUDGETS

Direct observations of CO<sub>2</sub> in the atmosphere indicate that the concentration is currently around 350 ppmv (parts per million by volume) and is increasing at about 1.5 ppmv y<sup>-1</sup>. This corresponds to rate of increase of carbon mass in the atmosphere of approximately 3 Gt C y<sup>-1</sup>. The release of fossil carbon from fossil fuel use and cement production is currently somewhat over 5 Gt C y<sup>-1</sup>. The conventional view of the atmospheric carbon budget is that the oceans are taking up most of the remaining 2 Gt C y<sup>-1</sup> of the release and that, overall, the terrestrial biomass is in nearly a steady state. There is, however, some difficulty in reconciling this view with estimates, based on analysis of land-use with estimates, based on analysis of land-use changes, that suggest a 2 Gt C y<sup>-1</sup> release of carbon from the terrestrial biota [3]. The discrepancy is exacerbated by the results of Tans et al. [4] who combined the results of modelling of the spatial distribution of CO<sub>2</sub> (see section 6 below) with data on the partial pressure of CO<sub>2</sub> in ocean surface water. They concluded that the net oceanic uptake of CO<sub>2</sub> was in the range 0.5–1.0 Gt C y<sup>-1</sup>.

The usual starting point for modelling studies of the atmospheric carbon budget is the assumption that the processes involving oceanic uptake of CO<sub>2</sub> have been invariant in time over the industrial period. Together with the fact that the processes are nearly linear in perturbations, this property implies that the atmospheric carbon budget can be analysed in terms of a response function formalism. One defines a response function,  $R(t)$ , which represents the amount of excess carbon remaining in the atmosphere at a time  $t$  after a unit input. It then follows that the atmospheric carbon content,  $C(t)$ , is related

to the source function,  $S(t)$ , by

$$(1) \quad C(t) = C(t_0) + \int_{t_0}^t R(t-t')S(t') dt'$$

The source,  $S(t)$ , is the sum of a fossil carbon source,  $F(t)$ , and a net biotic source (or sink if negative),  $B(t)$ . As noted by Enting and Mansbridge [5], relation (1) has a formal inversion relation:

$$(2) \quad S(t) = \dot{C}(t)/R(0) - [C(t) - C(t_0)]\dot{R}(0)/R(0)^2 - \int_{t_0}^t K(t-t')[C(t') - C(t_0)] dt'$$

If  $R(t)$  is approximated by a sum of exponentials then the inversion kernel,  $K(t)$ , can be readily calculated.

The explicit inversion relation shows that the deconvolution is only mildly ill-posed, effectively requiring the numerical differentiation of the concentration history,  $C(t)$ . However this simple relation hides significant difficulties. Firstly, the use of ice-core data to extend the concentration record,  $C(t)$ , back before the time of reliable direct measurements itself involves a deconvolution as described in section 3 below. More seriously, determination of the oceanic response,  $R(t)$ , requires the use of a model of oceanic  $\text{CO}_2$  uptake which must be calibrated indirectly. This problem is discussed in sections 4 and 8. Another serious difficulty is the possibility that ocean mixing processes may have varied in time. This would violate the assumptions underlying the response function analysis presented above and would also violate the assumptions on which the calibration of oceanic uptake using  $^{14}\text{C}$  is based. The issue of the violation of stationarity assumptions is discussed in section 9.

It should be emphasised that in practice, deconvolutions of ice-core data are performed using numerical models directly [6], rather than using the response function formalism. The value of the formal inversion relation (2) is not for performing numerical calculations but rather for revealing which aspects of the inversion lead to the ill-conditioning.

### 3. CO<sub>2</sub> IN ICE

The record of gas concentrations in bubbles trapped in polar ice provides an amazing insight into past changes in atmospheric composition. The interpretation of the record is complicated slightly by the fact that the bubbles in any layer are trapped over a range of times. Enting and Mansbridge [7] formalised this by writing the mean concentration,  $\chi(\tau)$ , within a layer deposited at time  $\tau$  as

$$(3) \quad \chi(\tau) = \int_{\tau}^T R_{\text{trap}}(t - \tau) C(t) dt$$

where  $T$  denotes the present and  $R(t)$  denotes the proportion of trapped gas per unit time at a time  $t$  after the deposition of the snow.

Formally, only a mild ill-conditioning occurs in the deconvolution, the predicted singularity in  $R_{\text{trap}}(t)$  being  $(t_c - t)^{0.55}$  near the 'cutoff age',  $\tau_c$ , based on modelling the trapping by the percolation model from lattice statistics [8]. However while the amplification of errors due to the ill-conditioning of the problem defined in terms of the mean trapping distribution,  $R_{\text{trap}}(t)$ , is comparatively mild, the percolation model predicts that finite samples will exhibit quite large deviations from the mean behaviour of the infinite-size limit. The implications of this behaviour have not been explored in detail.

### 4. CARBON CYCLE MODEL CALIBRATION

In the convolution equation (1) determining the atmospheric carbon budget, the response function,  $R(t)$ , needs to be determined. There have been several attempts to develop ocean models that aim to derive  $R(t)$  from the basic principles of ocean dynamics and chemistry [9]. However these do not yet seem to be sufficiently developed and so, for carbon cycle studies, the determination of  $R(t)$  still relies on indirect methods.

The traditional approach is to construct a very simple model of oceanic mixing and

then use the history of  $^{14}\text{C}$  (radiocarbon) from nuclear testing to calibrate the model. The simplest models consider discrete reservoirs and so model the carbon cycle in terms of ODEs. The widely used box-diffusion model of Oeschger et al. [10] represents the ocean as a horizontally uniform water column with diffusive mixing in the vertical direction. Therefore the calibration involves PDEs (one each for  $^{14}\text{C}$  and total carbon) coupled to ODEs. However this is a highly simplified representation of the real ocean in which tracer transport involves three-dimensional advective and diffusive processes and in which the steady-state assumption is at best only true for statistical averages.

There are however some serious difficulties involved in the use of  $^{14}\text{C}$  to calibrate models. Firstly, the ocean mixing response (and thus  $R(t)$ ) is derived as a modified deconvolution of the atmospheric (and to a lesser extent oceanic)  $^{14}\text{C}$  record. Secondly, some of the corrections that are needed in order to allow for the different behaviours of  $^{14}\text{C}$  and total carbon are themselves rather uncertain.

A preliminary analysis of carbon cycle model calibration from this point of view was given by Enting [1]. Following the carbon budget analysis by Tans et al. [4], Enting [11] extended his earlier analysis, investigating the extent to which calibration was sensitive to the rate at which the terrestrial biota could take up  $^{14}\text{C}$ .

The problem was analysed in terms of an approximate model which compared the  $\text{CO}_2$  response function,  $R(t)$ , to a  $^{14}\text{C}$  response function,  $X_{14}(t)$ . In terms of their respective Laplace transforms,  $r_{14}(p)$  and  $x_{14}(p)$ , these quantities are each related to a response function,  $M(t)$  (with Laplace transform,  $m(p)$ ) describing mixing of tracers out of the ocean surface mixed layer and into deeper ocean layers. In units such that  $R(0)$  and  $X_{14}(0)$  are 1,

$$(4) \quad r(p) = \left( p + \frac{\kappa}{1 + \kappa Z \xi m(p)} \right)^{-1}$$

$$(5) \quad x_{14}(p) = \left( v_{14}(p)^{-1} + \kappa \left/ \left( 1 + \frac{\kappa Z m(p)}{1 + D m(p)} \right) \right. \right)^{-1}$$

where  $v_{14}(p)$  is the Laplace transform of  $V_{14}(t)$ , the response function describing the way in which the terrestrial biota can take up a unit pulse of atmospheric  $^{14}\text{C}$ ,  $\xi$  is a chemical buffer factor ( $\approx 9$ ),  $D$  is the flux of detrital carbon, expressed as a fraction of the mixed layer carbon content,  $Z$  is the ratio of carbon contents of the atmosphere and ocean surface mixed layer and  $\kappa^{-1} \approx 8$  to 12 yr describes the air-sea exchange rate as an atmospheric turnover time.

In these terms, it can be seen that the use of  $^{14}\text{C}$  data to effectively determine  $m(p)$  for use in determining  $r(p)$  is a process which can be very sensitive to errors in the assumptions that are made in the various processes involved, particularly with respect to the biotic  $^{14}\text{C}$  response,  $V_{14}(t)$ .

For studies with numerical models of the global carbon cycle, Enting and Pearman [12], [13] introduced a Bayesian approach to the calibration by constructing a model in which most of the processes could be expressed in forms about which independent prior information. This prior information was then included so as to stabilise what was initially a very poorly conditioned parameter estimation process. In principle this approach can be used to give a comprehensive analysis of the uncertainties in the calibration. However, in retrospect, the re-analysis of the calibration problem [11] suggests that the numerical studies [12], [13] may have underestimated the importance of uncertainties in the biotic uptake of  $^{14}\text{C}$ .

## 5. INTERPRETING CHANGES IN $^{13}\text{C}$

The minor ( $\approx 1\%$ ) carbon isotope,  $^{13}\text{C}$ , provides additional information about the behaviour of the global carbon cycle. In principle it should be able to distinguish between

biotic and oceanic source/sink processes through their differences in isotopic fractionation. However the quantitative interpretation of  $^{13}\text{C}$  changes seems to be strongly model-dependent. Nuclear testing almost doubled the amount of  $^{14}\text{C}$  in the atmosphere; in contrast, only small changes in  $^{13}\text{C}:^{12}\text{C}$  ratios have occurred. This means that small non-linear terms in the exchange processes assume considerable importance. Another consequence of only small changes being involved is the use of the  $\delta^{13}\text{C}$  scale to express isotopic differences where, in units denoted per mil (‰),

$$(6) \quad \delta^{13}\text{C} = \left( \frac{[^{13}\text{C}:^{12}\text{C}]_{\text{sample}}}{[^{13}\text{C}:^{12}\text{C}]_{\text{standard}}} - 1 \right) \times 1000$$

Peng et al. [14] presented an attempt to deduce  $\text{CO}_2$  sources from a deconvolution of the record of  $^{13}\text{C}:^{12}\text{C}$  ratios in tree-rings. However subsequent work [15] suggested that the isotopic fractionation occurring during biotic uptake of  $\text{CO}_2$  is subject to a range of physiological influences related to  $\text{CO}_2$  concentration. Therefore, without independent physiological data, tree-rings do not provide an unambiguous proxy for atmospheric  $^{13}\text{C}:^{12}\text{C}$  ratios.

The problem of characterising the information provided by changes in global mean atmospheric  $^{13}\text{C}$  in terms of reducing the uncertainties in the atmospheric carbon budget remains unresolved. Some preliminary analyses have revealed the difficulties in interpreting atmospheric  $\delta^{13}\text{C}$  changes. If one assumes that there is an atmospheric  $\delta^{13}\text{C}$  response function,  $R_{13}(t)$ , describing the  $\delta^{13}\text{C}$  response to a source whose isotopic composition is taken as fixed, and use  $Q(t)$  and  $Q_{13}(t)$  to denote perturbations of  $\text{CO}_2$  and  $\delta^{13}\text{C}$  from their initial values, then one has the relations:

$$(7) \quad q_{13}(p) = r_{13}(p) s(p)$$

$$(8) \quad q(p) = r(p) s(p)$$

whence

$$(9) \quad q(p) = \frac{r(p)}{r_{13}(p)} q_{13}(p)$$

Actually, because of the non-linearity in isotopic exchange processes and the non-linearity in the definition of  $\delta^{13}\text{C}$  (where a changing  $^{12}\text{C}$  content appears in the ratio) a response function to describe  $\delta^{13}\text{C}$  changes should only be used for illustrative purposes. The oceanic uptake of isotopic perturbations is much faster than for total carbon; for illustrative purposes, we approximate the response by single exponentials,  $A \exp(-\gamma t)$  for  $\text{CO}_2$  and  $A_{13} \exp(-\gamma_{13} t)$  for  $\delta^{13}\text{C}$ . Substituting the Laplace transforms into relation (9) gives

$$(10) \quad q(p) = \frac{A}{A_{13}} \left( 1 + \frac{\gamma_{13} - \gamma}{p + \gamma} \right) q_{13}(p)$$

or equivalently,

$$(11) \quad Q(t) = Q_{13}(t) + \frac{A}{A_{13}} \int_0^t \exp[-\gamma(t-t')] Q_{13}(t') dt'$$

Thus within this approximation,  $C(t)$  is given as an the current  $\delta^{13}\text{C}$  plus a convolution integral over  $\delta^{13}\text{C}$  changes in the past. The same relation connects rate of change of  $\text{CO}_2$  and  $\delta^{13}\text{C}$ . In these terms, a measurement of the rate of change of  $\delta^{13}\text{C}$  gives information in one of two ways. Firstly, the scale factors and time constants involved in the relation will depend on the oceanic uptake parameters, although not in any readily interpretable model-independent manner. Secondly, a breakdown in the relation between  $\text{CO}_2$  and  $\delta^{13}\text{C}$  could be an indication of non-stationarity. There is some indication that the ice-core  $\delta^{13}\text{C}$  data of Friedli et al. [16] are showing some such effect, but the data quality is not adequate to give any great confidence about this.

Numerical studies by Enting and Pearman [12], [13] also indicated that, within the context of a specific carbon cycle model, the the rate of change of atmospheric  $\delta^{13}\text{C}$  was not independent of the other carbon cycle model data used to calibrate the model. However, when exploring the extent to which the model calibration is sensitive to uncertainties in

the biotic  $^{14}\text{C}$  response,  $V_{14}(t)$ , (as described in section 4) more recent modelling studies do show different rates of change of  $\delta^{13}\text{C}$  with different cases of oceanic uptake, but not with any simple dependence that allows simple direct interpretation of the significance of any particular rate of change of  $\delta^{13}\text{C}$ .

Therefore, the question of the utility of  $\delta^{13}\text{C}$  data in carbon cycle studies requires further investigation. Non-linear behaviour, combined with the very small changes involved, limits the validity of simple model analyses like that used for  $^{14}\text{C}$ , and so the problem is one of considerable difficulty.

## 6. SPACE-TIME DISTRIBUTION OF SURFACE SOURCES

In order to reduce the uncertainties in the global budget of atmospheric  $\text{CO}_2$  the space-time variation of sources and sinks has been investigated. The principle is that geographical separation of sources between land and sea should distinguish between biotic and oceanic processes. The inverse problem is one of a space-time deconvolution of concentrations to produce sources. The concentration data are available as time series of 5 or more years at several dozen locations around the world. The sources are related to concentrations by atmospheric transport and so the inversion requires some form of atmospheric transport model.

The observational data are insufficient to support a global-scale inversion and so numerical inversion studies [17], [18] have considered only a zonally-averaged behaviour (i.e. averages over longitude). Enting and Mansbridge [17] considered the conditioning of the inversion and found that the noise amplification exhibited a  $k^1$  growth as a function of the meridional wave-number,  $k$ . The same result was found by Newsam and Enting [19] who investigated the inversion of surface concentrations in some detail, using a highly idealised model of atmospheric transport. They treated transport as purely diffusive and

so were able to find analytic expressions for the relations between sources and concentrations. The reason that the purely diffusive approximation gave the same growth in error amplification as the more realistic numerical model is that in each case it is the most highly dispersive process, diffusion, that determines the degree of ill-conditioning.

The zonally-averaged numerical models [17], [18] represent the atmospheric transport as a combination of advection and diffusion. The zonal averaging leads to transport fields that vary smoothly in space and time. This is in contrast to models using 1 time and 3 space dimensions [20] in which the transport fields show the complex highly variable quasi-random complexity of real atmospheric motions. As well as advective and diffusive motions, expressed as PDEs in standard ways, it is also necessary to model sub-grid-scale convective processes. Such models have not yet been used in inverse calculations because of the lack of inversion strategies for working with such models, especially given the sparsity of the observational data.

In spite of the fact that the determination of surface sources from surface concentration data has been analysed more extensively than most other inverse problems in atmospheric chemistry, a number of important unsolved problems remain.

- a. The characterisation of errors in the estimated sources, from both data error and model error.
- b. Computationally efficient ways of using additional data such as concentration records from non-surface sites, supplementary data such as  $^{13}\text{C}$  data and various constraints.
- c. The design of sampling networks.
- d. The calibration of the atmospheric transport models used to relate sources to concentrations.

With regard to the model calibration problem, Enting [21] has noted conceptual and mathematical analogies between the problem of using concentration data to both estimate sources and to tune the transport and the seismic problem of estimating transport characteristics within the earth from earthquake signals whose time and position of origin are unknown [22].

## 7. SOURCES IN THE FREE ATMOSPHERE

For  $\text{CO}_2$ , most of the sources and sinks occur at the surface of the earth. This is not the case for most other radiatively active atmospheric constituents. Even for  $\text{CO}_2$  there is a minor source from the oxidation of CO in the free atmosphere. This makes a small change in the atmospheric carbon budget derived from the inversion of surface  $\text{CO}_2$  data [23], [24]. Enting and Newsam [25] have analysed some of the inverse problems involved in studying constituents with sources or sinks in the free atmosphere.

These problems are of great importance for  $\text{CH}_4$  for which the sink is through oxidation by interaction with the OH radical in the free atmosphere. Because of the extreme difficulty of measuring OH concentrations, this sink strength is estimated by a combination of forward modelling of atmospheric chemistry and inversion studies which tune calculated OH concentration fields to agree with model calculations for compounds such as methyl chloroform ( $\text{CH}_3\text{CCl}_3$ ) which are oxidised by OH and for which information is available about both source strengths and atmospheric concentrations.

## 8. OCEAN CIRCULATION

As noted above, dynamical modelling of oceanic circulation is not yet sufficiently advanced for it to be possible to deduce oceanic circulations (and thus the uptake of atmospheric constituents) from 'first-principles' dynamical calculations. An alternative approach is to

use the distribution of oceanic constituents in order to constrain the circulations. The majority of such oceanic inversion studies have worked with the dynamically important quantities: temperature and salinity. However these studies have generally been for limited regions [26] and so they do not address the global-scale questions involved in CO<sub>2</sub> studies.

Bolin et al. [27] attempted to obtain a coarse description of the global ocean circulation by inverting the distributions of carbon, oxygen, alkalinity, phosphorus and radiocarbon. Their analysis indicated a number of anomalies that they could not resolve.

The division of the world's oceans into the Atlantic, Pacific and Indian basins, with strong boundary current effects precludes the use of oceanic analogues of the zonally-averaged models used in atmospheric studies. For full three-dimensional (plus 1 time dimension) modelling, the oceanic circulations exhibit the same degree of complexity as the atmospheric transport described above.

From the description in earlier sections it will be clear that an understanding of the atmospheric carbon budget does not require a full understanding of the ocean circulation (at least so long as we avoid consideration of the type of non-stationarity described in the next section). What is required in the first instance is  $M(t)$ , a single functional defined in terms of the ocean circulation. Therefore an important topic for further research is an analysis of the accuracy with which  $M(t)$ , or some equivalent description of oceanic mixing, can be obtained from under-determined inversions of ocean circulation.

## 9. NON-STATIONARITY

The usual assumption that is made in studies of CO<sub>2</sub> over the industrial period is that the oceanic mixing processes have been invariant in time. Possible violations of this assumption are of concern for two main reasons. Firstly, non-stationarity will severely complicate

the interpretation of the atmospheric CO<sub>2</sub> budget over the industrial period. Secondly, most of the possible feedbacks between CO<sub>2</sub> and climate involve non-stationary oceanic behaviour. Such positive feedbacks appear to have been involved in the CO<sub>2</sub>-climate connection over the glacial to interglacial cycles. If similar feedbacks occur in response to greenhouse warming then the warming would be much greater than the predictions of models which omit such feedback [28].

Some of the apparent anomalies in our current understanding of the global carbon cycle may indeed indicate a non-stationarity in oceanic circulation. Enting and Mansbridge [29] analysed the response function relation (1) using  $C(t)$  specified by ice-core data from Neftel et al. [30] and the source,  $S(t)$ , given by the sum of the fossil input,  $F(t)$ , from [31] and the biotic input,  $B(t)$ , estimated by Houghton et al. [3] on the basis of land-use changes. It was found that the specified  $C(t)$  and  $S(t)$  were inconsistent with any response function,  $R(t)$  having the physically reasonable properties:

$$(12) \quad R(t) \geq 0 \quad \dot{R}(t) \leq 0 \quad \ddot{R}(t) \geq 0$$

The preferred suggestion by Enting and Mansbridge was that the biotic releases were in error. However an alternative explanation was that the oceanic uptake was not stationary but rather there was, over the nineteenth century, a recovery from a perturbation associated with the little ice age. This conclusion is supported by more recent work [32] that shows that the discrepancy found by Enting and Mansbridge [29] is increased if the sources incorporate an additional CO<sub>2</sub> sink due to enhanced biotic growth arising from elevated CO<sub>2</sub> levels.

Most of the mechanisms providing a climatic influence on atmospheric CO<sub>2</sub> involve ocean nutrients such as phosphorus. In this regard it is of interest to note that the main anomaly in the biogeochemical inversion calculation of Bolin et al. [27] was the phosphorus

distribution. Therefore it is interesting to speculate that the anomaly represented a failure in the steady-state assumption underlying the inversion calculation and that the same change might explain the inconsistency between  $C(t)$  and  $S(t)$  that was found by Enting and Mansbridge.

## 10. CONCLUDING REMARKS

The previous sections have described a number of important inverse problems that are involved in the study of greenhouse gases. The majority of these problems have not been analysed in any detail. This is especially the case for models involving partial differential equations for which only a small number of highly idealised situations have been analysed [19], [25]. In the hope of stimulating further investigation, some of the most important (in the view of the author) of these are

- a. Refining the determination of surface sources from surface concentrations by making efficient use of additional information such as non-surface data.
- b. Efficient strategies for calibrating (or tuning) atmospheric transport models.
- c. Development of techniques that can detect (and/or be robust in the presence of) model non-stationarity.
- d. More generally, the whole question of the effect of model error needs to be considered in much more detail.

In this regard, efficiency needs to be considered in a context where the ‘forward’ problem is defined in terms of integration of a complex numerical model for which explicit inversion algorithms will often not be available.

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