

Modelling the Airborne Dispersion of Pollutants from Coal Seam Gas Extraction

Peter J. Rayner and Steven Utembe

December 17, 2013

1 Introduction

Unconventional gas extraction has the potential to release various pollutants into the atmosphere. These could have direct impacts on the environment and health or indirect effects on the climate, offsetting the relative benefit of gas as a lower-carbon fuel. These impacts need to be both assessed and monitored; requiring a combination of measurements and models. For example, if a pollutant is released into the atmosphere, we wish to calculate the exposure of people or the environment as a function of meteorological conditions and the chemical properties of the pollutant. Alternatively we wish to quantify the emissions from incomplete extraction, leaks or other mishaps in order to quantify the complete greenhouse gas implications of a switch from coal to gas. This document aims to describe the available capabilities, both in measurement and modelling, for these tasks.

1.1 Purpose of document

This document will describe the suite of measurements and modelling tools for simulating the dispersion of pollutants in the atmosphere and quantifying emissions. It will:

1. Describe the classes of measurements and their applications;
2. Introduce the general principles of atmospheric dispersion models;
3. Describe the various classes of models, their applications and requirements;
4. Make recommendations for the measurement and modelling of direct environmental and climatic impacts of unconventional gas extraction.

1.2 Scientific Background

The principle underlying requirement of atmospheric models is the conservation of mass. This states that the change in concentration¹ of some atmospheric constituent is the sum of sources, transport in the atmosphere and chemical transformation. In principle then, we can simulate the concentration by writing equations (or numerical code) to describe these processes. These models are called chemical transport Models (CTMs). They are used to predict how much pollutant

¹We use concentration to denote the amount of the constituent per unit volume. The other common term, mixing ratio, refers to the relative amount of the constituent to that of air.

will arrive at a point given a previous release. This predictive modelling is also called forward modelling since we run the model forward in time. It is used to model everything from air quality at street level to the generation of the ozone hole. The other use for the same models, usually called inverse modelling, starts with measurements of a pollutant in the atmosphere and tries to localise and quantify the release that produced it. Put roughly we run the model backwards. So while forward and inverse models are closely related, they solve different problems; forward models estimate exposure to pollutants while inverse models estimate emissions.

There is a strong overlap between CTMs and modelling the physical behaviour of the atmosphere (weather prediction, climate and the like). One practical reason for this is that the most common constituent modelled in the atmosphere is moisture. The movement of moisture by winds and other atmospheric processes is similar to chemical constituents so we can borrow the numerical techniques for this. Many of the transport processes also require highly detailed simulation of changes in wind and temperature so that CTMs are often attached to state-of-the-art physical models to ensure the best possible simulation.

1.3 History of Use

Simple models of pollutant dispersion have existed since the 1940s (e.g. *Sutton*, 1947). As questions of urban air quality and transboundary air pollution rose in prominence, atmospheric models of trace gases became important research and policy tools. The first working grid-based model for the study of urban and regional air pollution was developed by *Reynolds et al.* (1973, 1974), with specific application to the Los Angeles air pollution problem. *Carmichael et al.* (1991) developed the the regional-scale Sulfur Transport Eulerian Model (STEM-1) to study sulfur pollution episodes that were associated with stagnant high-pressure systems. Examples of historical models down the years include the Acid Deposition and Oxidant Model (ADOM) of *Pleim et al.* (1991), the Regional Acid Deposition Model (RADM) of *Walmsley and Wesely* (1996), the Regional Oxidant Model (ROM) of (e.g. *Roselle et al.*, 1991). Australia has been a world leader in providing practical models for pollutant dispersion, (e.g. *Hurley et al.*, 2003; *Hurley*, 2005) which have been used to study dispersion on local and regional scales from, oil refineries and smelters. Another example of Australian based dispersion models is AUSPLUME (*Lorimer*, 1986).

Most major cities use air quality forecasts to underpin either advice to the population or, in extreme cases, policy intervention to limit pollution at the source.

The other common policy application is emergency response. This is usually a very efficient model which can be switched on to calculate exposure for a single source such as a volcanic eruption or industrial accident. The Australian Bureau of Meteorology maintains such a capability. Examples include the Chernobyl (e.g. *Kolb et al.*, 1989) and Fukushima (e.g. *Christoudias and Lelieveld*, 2013) nuclear accidents, the Buncefield explosion and fire (http://uk-air.defra.gov.uk/reports/cat05/0606201126_Buncefield_report_vF3_text2.pdf) and the Eyjafjallajökull volcanic eruption (*Vogel et al.*, 2013).

Inverse studies have long been used to infer the large-scale patterns of CO₂ sources and sinks (*Tait et al.*, 1990) and methane emissions (*Fung et al.*, 1991). More recently they have provided the key information on how CO₂ sinks on land respond to climate extremes (e.g. *Peylin et al.*, 2005; *Rayner et al.*, 2008). They have recently been applied to regions such as the Les Landes region of southwest France (*Lauvaux et al.*, 2009) and the intense agricultural area of the U.S. Upper Midwest (*Schuh et al.*, 2013). An emerging trend is the use of inverse models to deduce emissions from point sources. This was tested on a power-station near Port Augusta by *Utembe et al.* (2013).

2 Measurements

We wish to determine the exposure of people or the environment to any pollutants released by gas extraction or to determine the emissions. Both of these can be measured directly although with limitations on scale and accuracy.

Measurement systems exist for most of the major pollutants potentially released in gas extraction (e.g. *Hammer et al.*, 2002). For many more conventional chemicals (e.g. methane itself) measurement is possible on-site and continuously at a deployment cost of around \$70,000. More exotic chemicals involve instrumentation which is too expensive or fragile to leave in the field long-term. Finally air can be captured in flasks and returned to laboratories to measure a very wide range of species. These last two classes of measurements are used to check on exposure in field campaigns and to validate CTMs.

Emissions can be measured directly in one of two ways. If the location is certain (down to about 100m) we can use a technique called eddy covariance in which fluctuations of wind and concentration are measured by a pair of instruments mounted on a mast (usually about 30m high). Well-known techniques (*Aubinet et al.*, 2012) can then be used to calculate the emission. If the emission is very diffuse, such as seepage through soils, we can sample it by putting a sealed chamber on the surface and measuring the build-up of concentration in the air inside. This is a direct measurement of the emission but over only a fraction of a square metre. If the emission is spatially variable we may need many such measurements to observe it well. Finally in Section 5 we will describe how to use concentration measurements to infer emissions.

3 Forward Models

3.1 Classification

Atmospheric models can generally be classified either as Eulerian or Lagrangian. In Eulerian models, the domain is split into fixed grid cells and the tracer mass in every grid cell is calculated directly. By contrast, in Lagrangian models, particles or puffs of pollutant are tracked from the source so no calculations are performed where there is no pollutant. Both models calculate the concentration of pollutant so the emission and concentration data used is the same for either model.

Both types of models can be applied either "online" or "offline" depending on how the meteorology is coupled with the tracers or chemical species in the model. Online models are true weather models where the physical changes in the atmosphere (wind temperature etc) are simulated along with the chemistry. This means that pollutants can also affect the meteorology (e.g. *Baklanov*, 2010). Offline models take the output of physical models computed by a previous model run. Offline models are more common since they are much cheaper to compute.

3.2 Common requirements

The main requirement for any chemical transport model is a good representation of the physical state of the atmosphere (wind, temperature etc). Atmospheric constituents are advected by winds and mixed by turbulence in the atmosphere and may be transported by the internal motions of clouds. Much of the important chemistry in the atmosphere is driven by sunlight so accurate knowledge of clouds is also important. Thus the winds, temperature and clouds in a model must be as realistic as possible. This makes forecasting air quality particularly difficult since some of these physical processes are the most difficult to forecast. Forward modelling also needs good

knowledge of emissions. For pollutants like ozone the relevant emissions are for precursors such as oxides of nitrogen. For the major greenhouse gases, CO₂ and methane, we need the emissions of the species themselves.

Finally computational constraints mean we often run highly detailed simulations over small domains (typically hundreds to thousands of square kilometres in regional scale models). These are not closed in the real atmosphere so we must estimate the influence of pollutants arriving from outside the domain.

3.3 Problem of small-scale physics

Both weather models and CTMs are run on a mesh of points. The finer the mesh the greater the range of processes we can capture but the more computationally expensive the model. There are always processes we can't cover explicitly. We have to estimate these from variables we do compute. This is called parameterization. Good parameterization, appropriate to the scale of the model and the processes important in a particular region is fundamental to good models. Managing the trade-off between the size of the region we wish to model, the detail we wish to capture and the computational resources available is an early decision in setting up a modelling system. The choice of parameterizations in a model depends on the region, the dominant meteorology and the model resolution. This requires experience from the modeller during set-up but is usually not changed after that.

In the following sections we discuss the major classes of model, their differences and common features.

3.4 Common Processes

Several processes are modelled the same way in Eulerian and Lagrangian models. These include:

- Emissions. These may be gridded (e.g. emissions from a forest) or a series of point sources such as power-plants or gas wells. In general Lagrangian models are better suited to point source emissions such as power plants plumes whereas the well-defined mesh-like domain of Eulerian models means that they are better suited to area sources.
- Chemical transformation. This is treated similarly whether the transformation happens in a mesh cell in the atmosphere (Eulerian) or to an air parcel as it moves (Lagrangian).
- Deposition: Pollutants are ultimately removed from the atmosphere by being deposited to the surface of the earth. The process by which this occurs is either by dry deposition (through impact with the surface) or by wet deposition (through incorporation into water droplets or ice crystals and subsequent removal in precipitation). This process is not important for methane but is important for many other pollutants.

3.5 Eulerian Models

3.5.1 General principle

Also known as grid models, Eulerian models typically consider the model domain to be made up of three dimensional boxes or grids at a given resolution. Fluid properties are monitored as functions of time as the flow passes fixed spatial locations. Air movement through these boxes is

used to calculate the fluxes of pollutants in and out of the grid cells. In general, this approach mathematically gives rise to partial differential equations as the variation of the flow quantities occur in both space and time. Eulerian models are close to weather models since they share the same model structure (a mesh of points) and many of the same processes move heat and moisture in physical models and pollutants in CTMs.

Table 1: Strengths and weaknesses of Eulerian Models

Advantages	Disadvantages
Sample whole domain	Have fixed resolution
Computationally expensive	Easy to visualize and process results.
More physically complete	Difficult to generate source-receptor relationships

Although Eulerian models can be applied across a wide range of scales, they are mostly often used in applications that do not need extremely fine grid resolution such as in global models and regional models. The advantages and disadvantages of Eulerian models are summarised in Table 1.

Examples of commonly used Eulerian models for simulation of pollutants include Community Multi-scale Air Quality model (CMAQ)², The Air Pollution Model (TAPM)³, and the Weather Research and Forecasting Chemistry Model (WRF-CHEM)⁴

3.6 Lagrangian Models

3.6.1 General principle

Instead of a mesh of points, Lagrangian models represent the pollutant as a series of points or particles. Each particle has an exact position. So Lagrangian models are not limited to the mesh-size of a particular grid but they may need many particles to sample the pollutant distribution.

There are different types of Lagrangian models. The simplest are the forward or backward trajectory models. As the name suggests, forward trajectory models are run forward from a given source to evaluate its impact on receptor areas. Alternatively they can be run backwards where the trajectory path is defined in backward steps from a given receptor. In this approach the impact of emissions along the trajectory path can be assessed. Other types of Lagrangian models consider further treatment of the air parcel under consideration.

Table 2: Strengths and weaknesses of Lagrangian Models

Advantages	Disadvantages
Avoiding numerical diffusion	Fail in some meteorological conditions
Low cost	Difficult to visualise or treat results.
Can be used to analyse source-receptor relationships	

Their simplicity and efficient computation makes Lagrangian models popular at all scales. They are particularly well suited to very fine scales where the mesh of an Eulerian model may fail to

² <http://www.epa.gov/AMD/Research/RIA/cmaq.html>

³ <http://www.cmar.csiro.au/research/tapm>

⁴ http://ruc.noaa.gov/wrf/WG11/Users_guide.pdf

capture the shape of the pollutant plume. Lagrangian models do not work well in extremely windy or unstable environments such as thunderstorms.

Strengths and weaknesses of Lagrangian models are summarised in Table 2

Examples of commonly used Lagrangian models include CALifornia PUFF model (CALPUFF)⁵ and the Hybrid Single Particle Lagrangian Integrated Trajectory model (HYSPLIT) (e.g. *Draxler and Rolph*, 2013).

The relative strengths and weaknesses of the two modelling approaches suggest that Lagrangian models are well-suited to environmental assessment such as the impact of CSG releases.

3.7 Combined Models

Eulerian and Lagrangian models are not mutually exclusive. Eulerian models often compute all the fields needed to run a Lagrangian model. Thus they are frequently used in the one system (e.g. *Brandt*, 1998; *Brandt et al.*, 1998; *Pillai et al.*, 2012). This allows the same modelling system to work near the source where the high-resolution of Lagrangian models is an advantage and far from the source where the complete coverage of Eulerian models is better. It also allows the same system to be used for forward and inverse modelling. We recommend adopting the Weather Research and Forecasting model with the Stochastic Time-inverted Lagrangian Transport model (WRF-STILT) as described in *Pillai et al.* (2012).

4 Using Forward Models

In this section we describe a number of practical considerations for using models. We develop these in the context of forward models but most will also hold for the inverse models discussed in the next section.

The choice of which model to use depends on a number of factors such as choice of scale, operational requirements, data and infrastructure requirements, computational requirements and required expertise.

4.1 Choice of scale

Setting up a model for a particular domain and resolution can be time-consuming so this choice requires care. It is a trade-off between computational cost and the quality of the simulation (*Law, A. M.*, 1991). The pollutant under study affects the size of the domain since different pollutants will survive longer in the atmosphere and hence be transported further. As an example, Figure 1 shows the correspondence between temporal and spatial scales in the atmosphere and the group of pollutants that correspond to the local, regional, continental and global scales (*Whelpdale*, 1983). Belonging to the local scale (scale distance of up to 10km are heavy particles or highly reactive species (e.g. H_2S) or species that are rapidly deposited (e.g. large Pb-containing particles). Regional scale considers distances of the order of 100km. In this category, pollutants with lifetimes of several hours (e.g. O_3 , NO_x etc) can be transported tens of km downwind of precursor emission sources. Pollutants (e.g. SO_x , fine particles) in the continental scale category have lifetimes of several days and can be transported up to thousands of kilometres. Global-scale problems involve long-lived species such as CO_2 , CH_4 and the perfluorocarbons.

⁵<http://www.src.com/calpuff/calpuff1.htm>

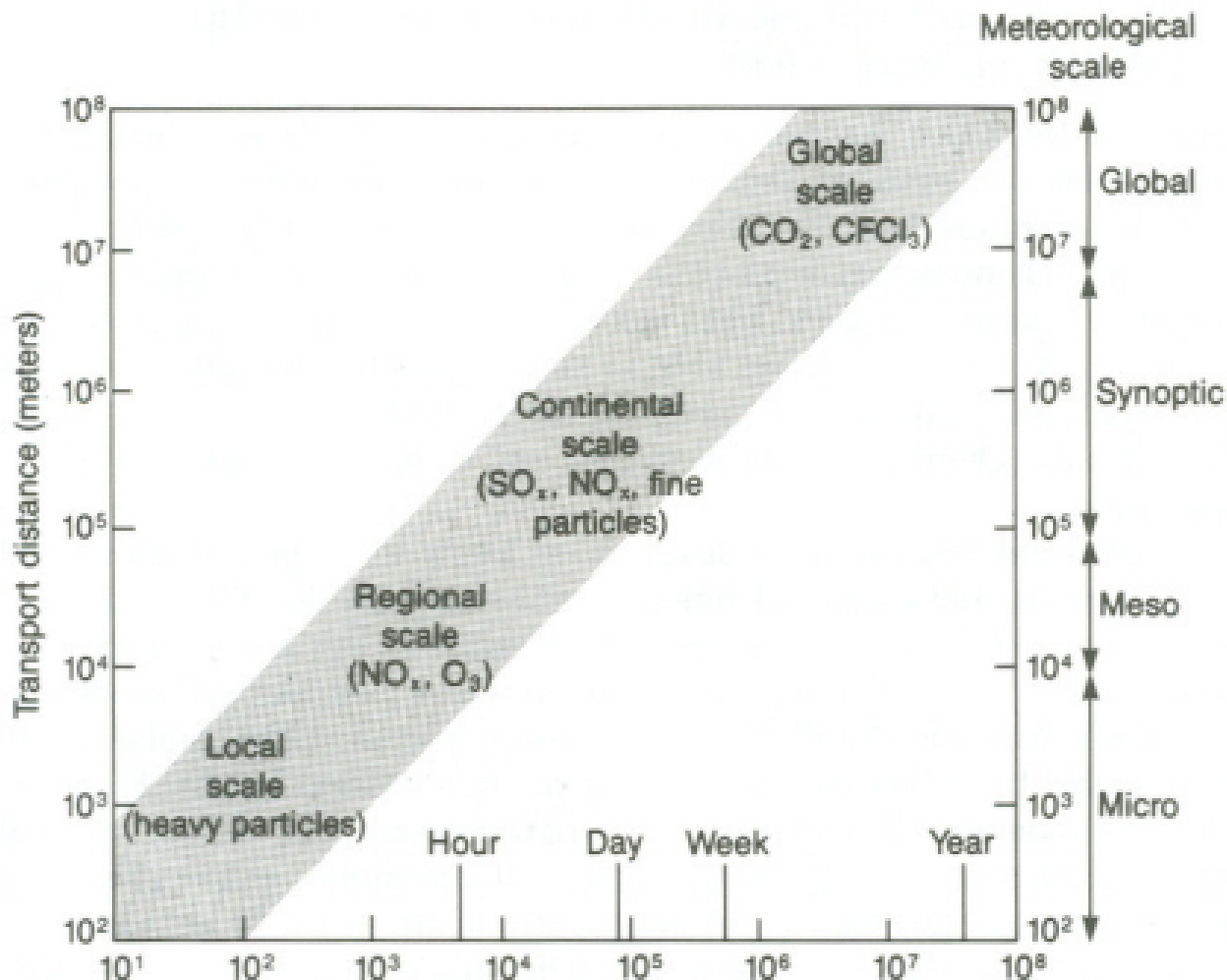


Figure 1: species simulated at various atmospheric scales

4.2 Operational requirements

There are a number of model operational requirements that need to be considered. These include data and infrastructure requirements, computational requirements, as well as required expertise.

4.2.1 Data and infrastructure requirements

Atmospheric models require considerable input data which is itself often generated by other models. Typical required data include:

Emissions Input to the model as surface fluxes mass per unit area per unit time), emissions can be from anthropogenic sources (e.g. CH_4 emissions from coal seam gas wells) or biogenic (e.g. volatile organic compounds (VOCs) such as isoprene) from vegetation. Emissions may be measured directly (see Section 2) though this is usually only possible from a limited number of pre-identified point sources. This includes gas-wells but excludes leaks in complex pipeline

infrastructure. In the absence of direct measurements one will use maps of potential sources and rely on the inverse methods described later to improve the information.

Boundary conditions regional models require lateral and vertical boundary conditions. These are the concentrations or inflows at the edges of the domain. They are needed since pollutants may enter the domain as well as be emitted within it. Only the side boundaries usually matter because horizontal winds are much stronger than vertical winds. Typically, boundary conditions are generated from global model runs. An example is in WRF-Chem, where simulations from the global Model for Ozone and Related Chemical Tracers (MOZART-4) are frequently used as boundary conditions (*Emmons et al.*, 2010). The accuracy of boundary conditions can also be improved by satellite observations (*Hollingsworth et al.*, 2008).

Meteorology Needed to drive the transport of the model, meteorological fields are usually obtained from operational agencies such as the Bureau of Meteorology.

Geophysical data This include surface parameters such as topography, land use categories and vegetation types.

4.2.2 Computational requirements

Thanks to the increased power of modern computers, computational limitations to running atmospheric models are not prohibitive. Running them operationally requires access to large amounts of data which usually places them within operational agencies. Research and assessment experiments can be run on modern computing clusters (provided one chooses a model set up for this) and emergency response calculations can be run on a desktop provided the driving data is available.

4.2.3 Required expertise

The problem of forward modelling for environmental exposure is fairly well understood. Furthermore modern models are not particularly difficult to set up and the most popular have a range of on-line and class-based tutorials. For example the most widely used model globally WRF-Chem has an annual tutorial run by the model developers. Similarly the CSIRO and various private providers will train and certify users of their TAPM model. Experienced modellers are still required to interpret the results. In particular an understanding of which meteorological situations are difficult and understanding the sensitivity of the results is required to understand potential uncertainties. Thus, while assessment teams do not need to consist entirely of experienced atmospheric chemistry modellers, we recommend having one available to assess the results.

4.3 Knowledge gaps

- A forward atmospheric model can only be as accurate as the description of the emissions in the model domain. Currently there are significant gaps in knowledge concerning fugitive emissions from CSG and other unconventional natural gas resources (*Tait et. al.*, 2013). This is usually treated by running multiple scenarios to bracket likely responses.
- Although a well-studied subject, the sea breeze is still hard to forecast. This degrades the qualities of current simulations of evolution of pollutants in coastal areas.

- The trapping of pollutants near the ground at night is a difficult phenomenon to model. For continuous fluxes (i.e those that continue at night) this trapping can lead to high peak exposure. Special care and even some ad hoc correction must be made to estimate peak nighttime exposures.
- The transport of pollutants in clouds and thunderstorms is an active research area although it is less important for questions of peak exposure.

5 Inverse Modelling

5.1 What are They For?

In many situations we observe the outcome of some physical process and wish to infer something about its cause. Such inferences are made in the opposite direction to physical causality and are hence termed inverse approaches. To make such an inference we need a model mapping the (unknown) cause to the observed effect. Speaking loosely we “invert” (run backwards) this model to gain information of the cause from observations of the effect. In our situation, the cause is an emission of some pollutant and the observations are measurements of atmospheric composition.

5.2 General Principle

This class of problems is treated exhaustively by *Enting* (2002). It is a branch of statistical inference in which we estimate the emissions given measurements and, optionally, prior estimates of emissions. The statistical nature of the problem means that both the measurements and prior estimates are associated with uncertainties. The methodology subsumes both verification of prior estimates and generation of improved emissions estimates. Without presenting the mathematical development we can describe the algorithm as follows:

1. Use the prior estimates of emissions as input to a CTM to simulate concentrations at the times and places of the measurements;
2. Calculate the differences between these simulated concentrations and measurements;
3. Weight these mismatches by the uncertainty in the measurements;⁶
4. Make an optimal correction to the prior estimate of emissions; this will depend on how uncertain we think the emissions are.

The optimal correction in step 4 is the heart of the process. It requires knowledge of how each simulated concentration will change as we change each component of the emission. This sensitivity of simulated concentrations to emissions is usually called the Jacobian \mathbf{J} .

5.3 Requirements relative to Forward Modelling

The main requirement is the feasible calculation of \mathbf{J} . This can be done with many forward runs e.g. for each possible emission point. Thus it is extremely useful to have an inventory of all possible emissions before starting the calculation. This not only reduces computational cost

⁶This must also account for the md’s ability to simulate the particular measurement

Emissions	Measurements	Choice
Few	Any	Forward Eulerian
Many	Few	Backward Lagrangian
Many	Many	Adjoint
Many	Path-integrated	Adjoint

Table 3: Conditions and choices for Inverse Model.

but increases the ability of the measurements to quantify emissions. We recommend that such inventories are made wherever possible. If, however, we need to calculate a map of emissions this forward computation rapidly becomes infeasible (it can require millions of hours of computing). With care we can also calculate \mathbf{J} running tracer transport backward, directly representing the reversal of causality mentioned earlier. This is easiest for Lagrangian models where we move particles backwards along trajectories but is also possible with specialised models called adjoints (*Kaminski et al.*, 1999a,b) derived from forward models. These are computationally efficient but difficult to generate. The choice depends on the number and type of measurements and the number of emissions we are trying to estimate. Table 3 lays out the various conditions. The

5.4 Regional vs global

The need for high accuracy and hence high resolution suggests the use of regional rather than global models for detailed inversion studies. *Geels et al.* (2006); *Law et al.* (2008) have shown that these models capture better the detailed variations in concentration we use to locate and quantify emissions. A problem is the influence from outside the boundaries of our domain. These can be included by including the boundaries as part of the unknowns in the problem (e.g. *Lauvaux et al.*, 2008) or by nested approaches (e.g. *Rödenbeck et al.*, 2009). It is convenient if the signals that come from the boundaries are easily distinguished from those from the emissions we seek. This will happen if the boundaries are very far away so that signals arriving at our observing sites are smooth and diffuse while those from nearby emission sources vary rapidly as winds change. This requirement conditions the setup of a regional inverse study and also suggests that certain classes of problem are especially hard because the boundary and internal influences are hard to separate. Fortunately the detailed regional problem we are discussing appears not very sensitive to boundaries provided we use a reasonable domain.

5.5 Characterizing Uncertainty

The emissions and measurement uncertainties we prescribe are as important as the data themselves (*Raupach et al.*, 2005). Prior uncertainty is calculated by comparing our prior estimate with independent data (e.g. *Chevallier et al.*, 2006, 2010). Data uncertainty arises from the characteristics of the measurement. For most in-situ measurements in greenhouse gases it is negligible for the regional problem. Model uncertainty is calculated from the differences between simulated and concentrations *for cases with exactly known emissions*. The problem is similar to the validation discussed below. We can also use an ensemble of different models as realisations of the correct model either for forward models (e.g. *Law et al.*, 1996) or inverse models (e.g. *Gurney et al.*, 2002).

5.6 State of the Art at Various Scales

Inversion modelling is at different stages of development for different scales. At the global scales there are systems at or near routine production. The CarbonTracker system (more information at: <http://carbontracker.noaa.gov>) delivers annual updates of estimated carbon fluxes for subcontinental regions over the globe with a more detailed focus on North America. Similar systems are under development for Australasia, East Asia and Europe. NASA's Carbon Monitoring System (<http://carbon.nasa.gov>) and the European GMES Atmospheric Service (<http://gmes-atmosphere.eu>) both aim to produce more frequent updates using satellite measurements of greenhouse gases.

Work at regional scale ($1000 \times 1000 \text{ km}$) is limited to intensive research projects (e.g. *Lauvaux et al.*, 2009, 2012; *Schuh et al.*, 2013) which have demonstrated success when comparing their proposed net uptake to direct measurements or inventories. Such efforts have been greatly boosted by the reduction in measurement cost and more intensive regional deployments are planned, some with commercial funding (<http://www.earthnetworks.com>).

There are no operational systems at city or regional scale but several research projects aimed at urban emissions mapping using atmospheric measurements, e.g. for Paris, Indianapolis and Los Angeles. Some of these are likely to evolve naturally into long-term monitoring projects. There is also funding to instrument the Latrobe Valley under the Carbonet initiative of the Federal Government's Education Infrastructure Fund

Monitoring of point source emissions has been studied for purposes of verifying geosequestration. *Loh et al.* (2009) demonstrated the ability to detect and locate releases from a single source from a distance of about 1 km. Checking whether this technique could isolate which of many potential gas-wells was leaking is an important research task.

Industrial emissions at smaller scales (such as gas fields) have only been studied during intensive campaigns. This is a very recent development based either on the proximity of research labs to significant sources (e.g. *Petron et al.*, 2012) or the availability of portable technology (*Karion et al.*, 2013).

5.7 Simplified Approaches to Inverse Modelling

So little is known about emissions from unconventional gas extraction that various simplified, exploratory approaches have been used to detect emissions. In Australia a team at Southern Cross University have surveyed the Tara Gas Field in SE Qld. http://scu.edu.au/coastal-biogeochemistry/download.php?doc_id=12515&site_id=258&file_ext=.pdf This was an exploratory survey designed to discover whether there were elevated methane concentrations that might arise from local sources. The Picarro Surveyor (<http://www.picarrosurveyor.com>) is a commercial product using a similar approach. It uses vehicle-mounted instruments that can measure rapidly concentration and wind direction. If a concentration spike is detected, the wind direction measurement allows it to be roughly located. By driving a grid pattern one can roughly map emissions. Recently reported work demonstrated such leaks at a gas-field in Texas and showed that most were caused by engineering failures which were easily repaired (<http://www.esrl.noaa.gov/gmd/annualconference/slides/99-130416-A.pdf>). Because it relies on spikes in concentration, it is likely to miss diffuse sources which do not produce spikes but large-scale changes in concentration. These sources (such as soil seeps) may be large when integrated over an entire gas-field. These techniques are good for identifying sources but less so at quantifying them. They should, however, form a part of any monitoring system. We recommend that such a capability be developed and readily available.

5.8 Instrument and Network Considerations

Instrumentation and modelling requirements for determining emissions depend on the scale of the problem. If we know the location of any potential emission we can make a direct measurement of the flux from the surface at that point (*Aubinet et al.*, 2012). The field of view of such a measurement is perhaps $100\times 100\text{m}$ depending on the height of the measurement so it is impractical for diffuse emissions or complex infrastructure.

The atmospheric techniques discussed here make use of two classes of measurements, either in-situ measurements made by analysing air at a point or path-integrated measurements that measure the total amount of tracer along some path (often the path between the instrument and the Sun). The in-situ measurements are highly precise and give good local detail. Path-integrated measurements cannot localise sources as well as in-situ measurements but can observe larger areas since tracer arriving anywhere along the path will be observed. Optimum strategies combine the two classes of measurement. There are techniques of network design that can suggest an optimal network of measurements (e.g. *Rayner et al.*, 1996; *Lauvaux et al.*, 2008). Modern instrumentation is making it easier to maintain networks of instruments but regular inspection is still required. This means the staffing requirement is set by both the distances involved and the number of instruments. A trained technician can probably support 10 instruments provided they can inspect them monthly. Given usual up times of 90% we should expect that one of these 10 instruments was under repair at any time.

Once an emission is noted, the survey technique is unequalled at locating it. Thus we recommend a standing capability for surveying CSG fields.

Another particularly promising technique is to use an aircraft to sample the concentration upwind and downwind of a gas-field. The difference in concentration is very likely to come from emissions from the field and some simple calculations can quantify it. *Karion et al.* (2013) used the technique to estimate emissions from a gas-field in Utah. There are research aircraft in Australia capable of similar studies provided they were equipped with appropriate instruments. We recommend investigating such a capability for Australia.

5.9 Knowledge Gaps

The knowledge gaps identified in Section 4.3 which pertain to atmospheric transport also affect inverse modelling. For inverse modelling they generally limit the times and places where we can use data. The early stage of research into using inverse models for CSG problems means it is unclear how confident we should be in their estimates. *Utembe et al.* (2013) was quite successful in estimating an isolated source but further research is required to extend this to complex sources like a CSG field. We recommend that such a study be undertaken for a small field for which we can develop a reliable inventory.

6 Validating Models

Both forward and inverse models are validated in two ways: directly by comparing with measurements or indirectly by comparing with other models. For inverse models, the task of validation and the task of quantifying uncertainty are similar since both aim to quantify the confidence we should have in their results.

6.1 Direct validation

Direct validation involves comparing modelled and measured concentrations to test the model. This requires two things:

- Measurement data suitable for comparison with models
- Good knowledge of the inputs, particularly the emissions (since we must rule out an incorrect emission strength as a source of error).

Direct validation of forward models can be done by conducting a planned tracer release experiment whereby an inert tracer such as Radon-222 is released and measured at various locations in the model domain. It can also be done by taking advantage of data from industrial accidents or simple natural or anthropogenic sources. The European Tracer Experiment (ETEX) is a widely used intentional release experiment of an inert pollutant near the ground (*vanDop et al.*, 1998). It is hence highly relevant to fugitive CSG emissions. Many CTMs have simulated the case. Their performance is generally good close to the release point with the quality of long-range transport mainly controlled by the quality of the meteorological data used in the simulation. We recommend that the use of any forward model should be preceded by a detailed screening program to identify major characteristics of meteorology of the area.

There are very few direct validations of inverse estimates because there are few cases where we know the emissions well enough to test the model. Tests using isolated power-stations (e.g. *Utembe et al.*, 2013) have shown good agreement with reported emissions provided the CTM used has good meteorological data to drive it. Tests of small cities (e.g. *Turnbull et al.*, 2011) have compared atmospheric measurements with simulations driven by the best available inventories and also shown good agreement.

6.2 Indirect Validation

Indirect validation of an atmospheric model is achieved by verifying with related dynamical models, conducting inter comparisons and sensitivity studies with other already well-established models, and by conducting peer review of the science of the model. Indirect validation is easy to do as it does not require measurements but requires caution since it does not directly assess the difference between the model and reality.

For inverse models the statistical characterization of the uncertainty can be expanded to include uncertainties in the model itself, usually by using an ensemble of models. For experiments at regional scales (*Schuh et al.*, 2013) this extra uncertainty was smaller than uncertainty arising from insufficient data. This is likely to hold at the scales of CSG fields as well but this is untested.

7 Modelling Summary

We attempt here to draw together the information in the preceding sections into a concise summary. The references direct readers to relevant sections in the document.

8 Examples of Model Applications

In Section 1.3 we described some of the uses of CTMs. CSG, however, is a recent enough development that there is, to our knowledge, no published work on their application to this problem.

Aspect	Forward	Inverse
Required Inputs	<ul style="list-style-type: none"> • Meteorological Data 3.2 • Emissions Inventories 3.2 	<ul style="list-style-type: none"> • Meteorological Data 3.2 • Concentration Measurements 5.8 • Boundary conditions for the domain 5.4, 4.2.1 • Optionally emissions estimates which we will either verify or improve
Outputs	3-dimensional grids of concentrations for pollutants	Maps of emissions
Algorithms	<ul style="list-style-type: none"> • eulerian models (3.5) solve the atmospheric transport equation on a mesh of points • Lagrangian models (3.6) represent the pollutant field as a series of particles and transport each particle separately. 	<ul style="list-style-type: none"> • Inverse models use the same transport algorithms as forward models • Estimating emissions (5.2) uses Bayesian estimation. • Inverse models reverse the effects of transport; easy for Lagrangian models but requiring an adjoint for Eulerian models.
Skills and training	<ul style="list-style-type: none"> • Training is widely available • experience either in atmospheric chemistry modelling or meteorological assessment needed to interpret results (4.2.3) 	<ul style="list-style-type: none"> • Requires same skills as forward modelling • Also requires working knowledge of Bayesian statistics.
Modelling providers	<ul style="list-style-type: none"> • Methods widely used in research institutions • Many private consultants in environmental assessment 	<ul style="list-style-type: none"> • Still a research problem • CSIRO and several university research groups have expertise
Best practice	<ul style="list-style-type: none"> • Eulerian model with embedded Lagrangian capability for handling close and distant exposure 	<ul style="list-style-type: none"> • Same models as for forward case • Well-developed Bayesian inverse system for estimating emissions

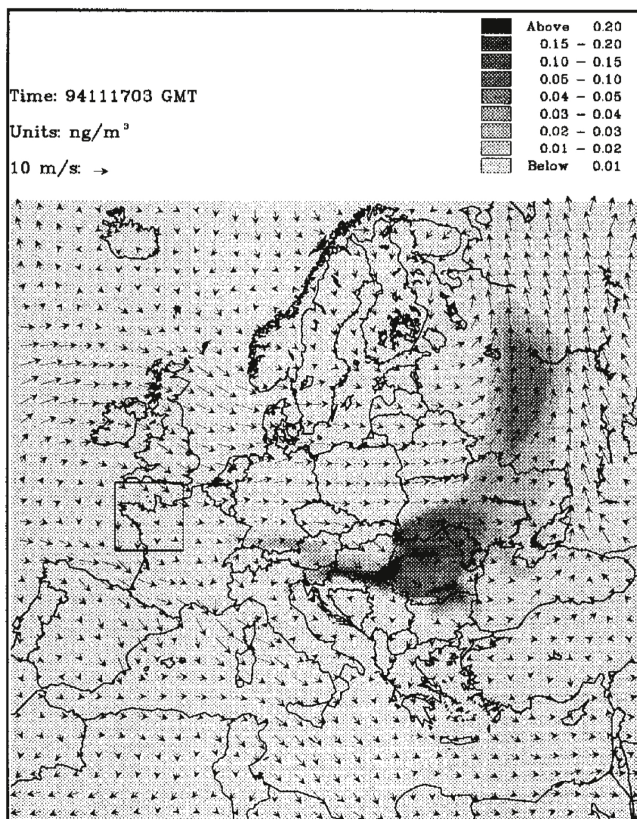
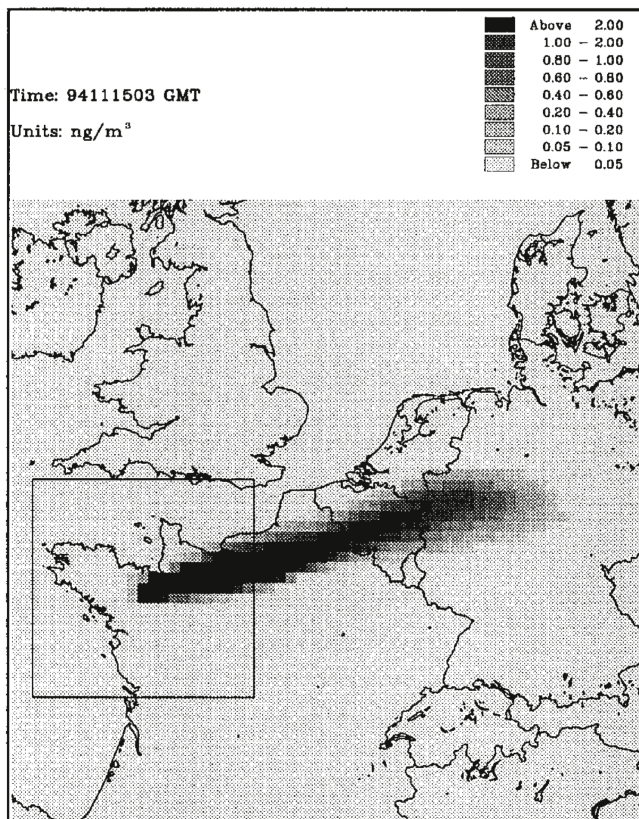
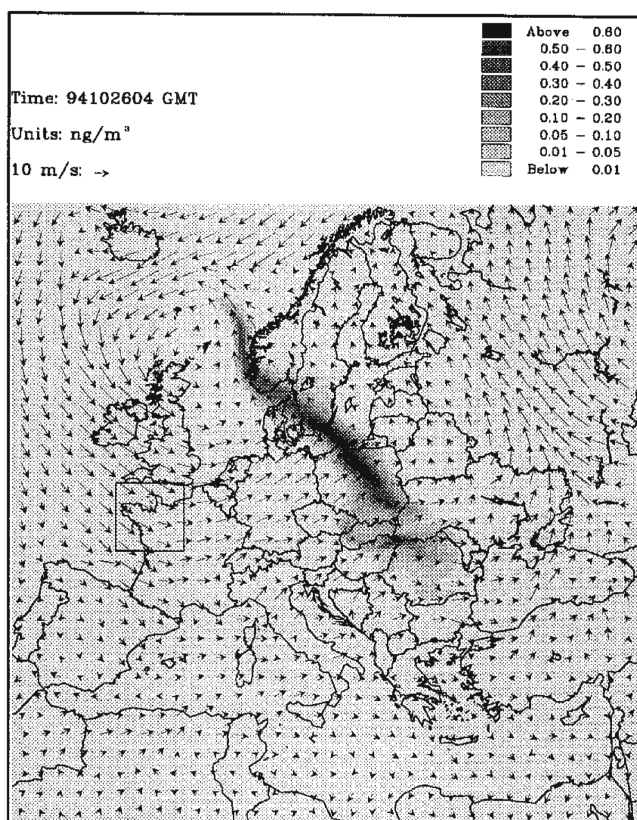
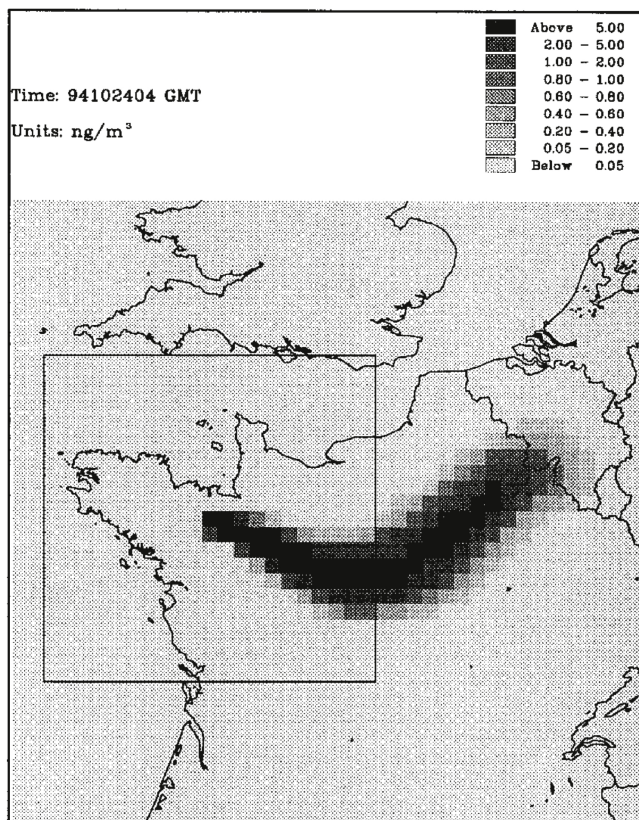


Figure 2: Model simulations of ETEx-1 (upper figures) and ETEx-2 (lower figures). The left figures are 12 h after start of the releases, and the right figures are 60 h after start of the releases. The duration of the release are, in both experiments, 12 h. The square in the figures indicates the area where the Lagrangian model is operating. Source: *Brandt et al. (2000)*

Most case studies have looked either at very large-scale dispersion (typically of radionuclides) or the short-range dispersion of heated gas from a tall stack. We present a case study which is physically similar to the CSG problem, a relatively slow release of a non-heated pollutant near the ground. The case study is the European Tracer Experiment (ETEX)

In October and November of 1994, two releases of a perfluorocarbon tracer were made from a site in western France as part of the European Tracer Experiment (ETEX). The aims of the experiment were to evaluate real time forecasting of the tracer concentration field as well as providing data for future (subsequent) model analyses of the transport patterns. The sampling network was comprised of 168 stations across Europe which sampled every 3 hours (*Mosca et al.*, 1998).

Many types of dispersion models have been validated against this dataset (*Nodop*, 1997). One such model is the Danish Rimpuff and Eulerian Accidental release Model (DREAM) (*Brandt*, 1998; *Brandt et al.*, 1998). The DREAM model is a combination of a Lagrangian short-scale puff model and an Eulerian long-range transport model. The Lagrangian component of the model is used near the source to calculate the initial transport and dispersion of the emission/release. Beyond this, the Eulerian model takes over to calculate the long-range transport and dispersion in the rest of the model domain.

Figure 2 shows the simulation of the spread of the release for ETEX-1 (upper figures) and ETEX-2 (lower figures) as taken from *Brandt et al.* (2000), with the left figures showing spread 12hrs after release and the right figures 60 hours after the release. The marked rectangle shows the area where the Lagrangian model was operating. For both releases, the prevailing weather was characterised by low pressure systems with centres north of Scotland giving rise to westerlies at the release site. However, there were high wind speeds during ETEX-2 which resulted in fast transport of the plume over Europe.

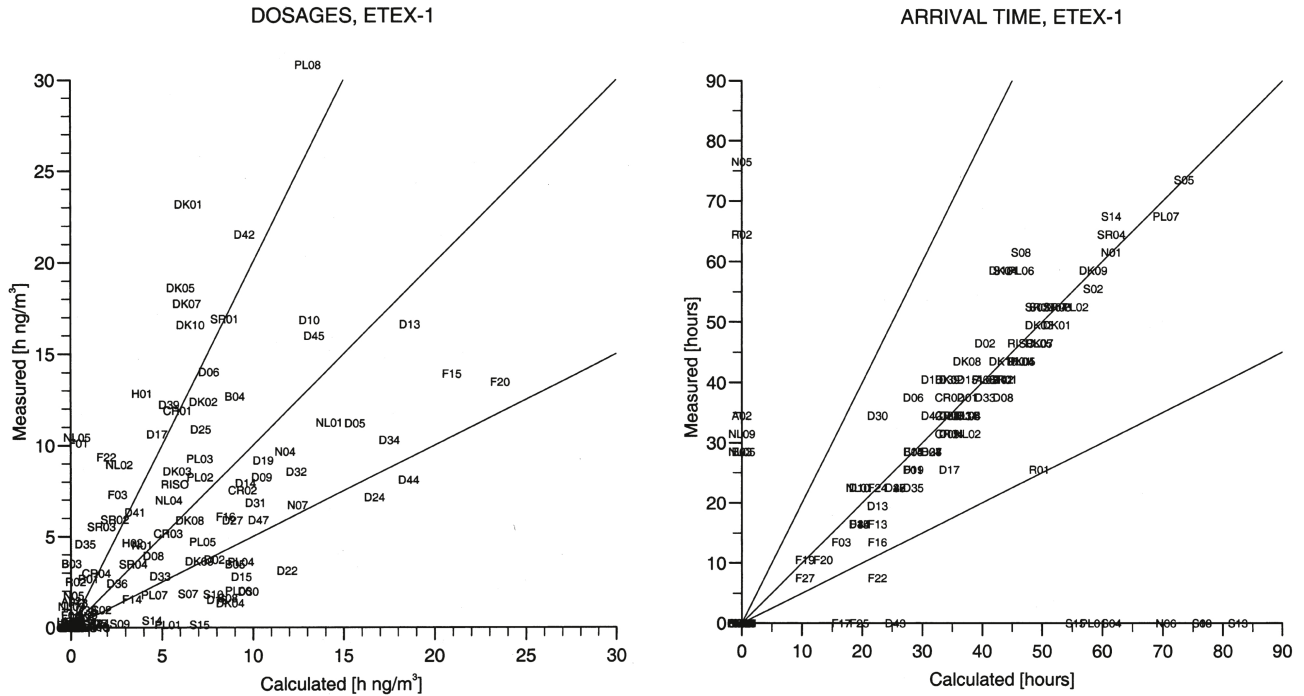


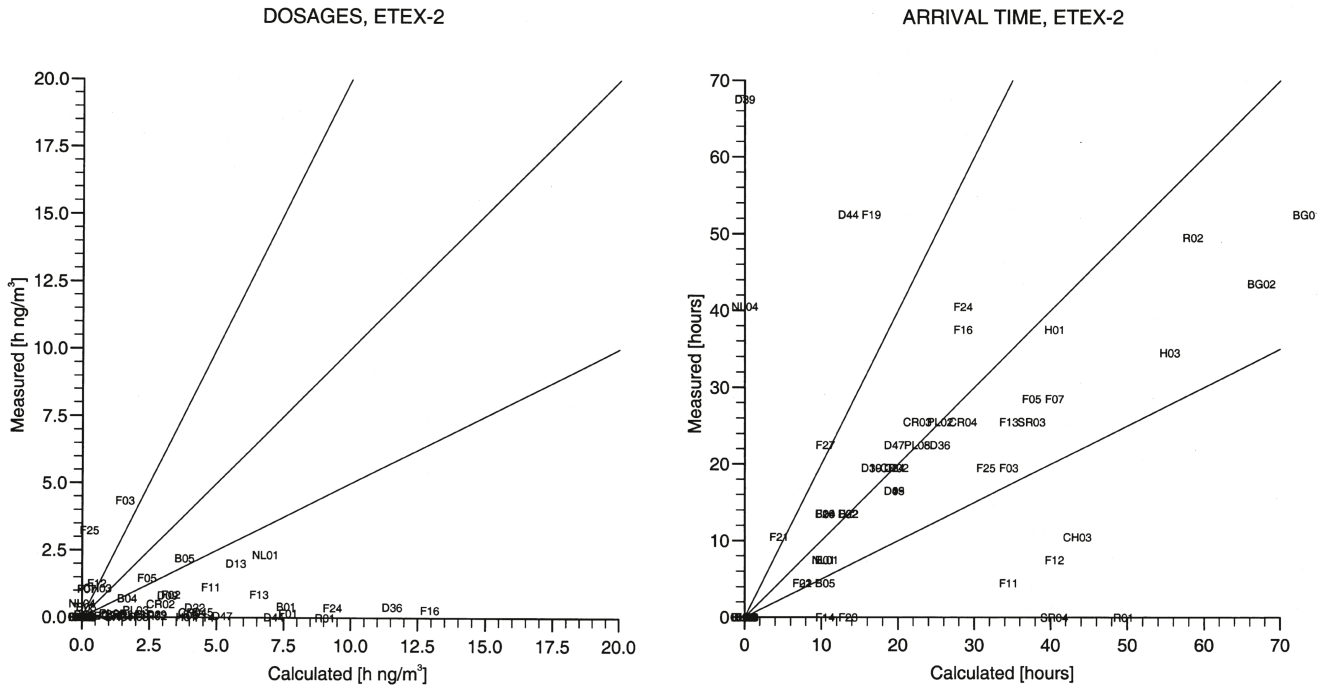
Figure 3: Comparisons of model calculations with measurements for ETEX-1. Left figures are dosages (or integrated concentrations), and right figures are the arrival times at the measurement stations. Source: *Brandt et al.* (2000)

Figures 3 and 4 show comparisons of model calculations with measurements given as scatter-plots of total dosages (or integrated concentrations) and arrival times at the different measurements sites.

In the case of ETEX-1, the accuracy of the calculated model dosages are, at worst, within a factor of 2-3, and for the arrival times within 2-3 hours. This puts DREAM within the best performing models according to the conclusions from the ETEX experiment. It is worth bearing in mind that the uncertainty on measured concentrations from ETEX-1 was estimated to be within $\pm 50\%$ which might explain some of the model-measurement discrepancies.

Worse agreement was observed for ETEX-2 where DREAM (like all other models run for ETEX-2) was found to overestimate the dosages to a larger extent than during ETEX-1. Although no explanation for this has been found *Nodop* (1997), possible reasons include bad simulation of the meteorology during the experiment and lack of wet deposition of the tracer during the heavy rainfall observed during ETEX-2. This is why it is very important that in the design of any modelling experiment, careful attention should be given to the prevailing weather conditions which will, in turn, guide the inclusion or omission of parameterizations in the model.

The releases in the ETEX experiments are similar to a point source or accidental release in a CSG operation. Thus the performance of the models is a good guide to how well they will work for CSG cases. In general they work well close to the source and their performance further afield is largely determined by the quality of the meteorological fields they use.



9 Application to CSG Modelling

There are two major applications for atmospheric modelling of CSG emissions: the risk of exposure to pollutants and the estimation of fugitive emissions. As we have seen these require different tools and are at different stages of development. Here we summarize the next steps for each problem in the CSG context.

9.1 Risk Analysis

Adopting the usual framework of $\text{Risk} = \text{Likelihood} \times \text{Consequences}$ we can break the risk analysis for pollutant exposure into three steps:

1. the likelihood of emission of a given pollutant
2. the exposure of people or the environment to that pollutant
3. the consequences of such an exposure.

Atmospheric modelling addresses only step 2 of this chain. It establishes the relationship between the emission rate for a pollutant and the exposure. This information must be integrated with the other two steps, especially detailed information on emissions. We recommend this as a follow-up study.

9.2 Steps for Estimating Emissions

Estimating emissions from a CSG field using atmospheric measurements remains a research problem. The success of the method at many other scales combined with the difficulty of an engineering-based estimate does suggest it is both necessary and likely to succeed. The concrete steps to setting up such a calculation are listed below including references to the sections where they are explained.

1. Choose a domain for the study (4.1, 5.4).
2. Prepare an inventory of possible emissions (5.3).
3. Carry out a network design study (5.3) to select optimal locations and types for atmospheric measurements (5.8).
4. Build and install network. Note the staffing requirements in (5.8).
5. Install system for inverse estimation of emissions (5.2). At time of writing there is no turn-key system for emissions at these scales so this needs collaboration with a research group. Such a system could, however, be built for deployment over many CSG fields.
6. Combine measurements and inverse system to estimate emissions.
7. Carefully investigate the residuals (differences between predicted and observed concentrations). These can indicate either problems with the CTM (e.g. 3.3) or that our emissions inventory is missing important sources (5.3).

10 Recommendations

The following gather the key recommendations from earlier sections

1. Agencies involved in assessment should have access to a high-quality, general purpose CTM. The best candidate at time of writing is the WRF-STILT combined Eulerian-Lagrangian model (3.7).
2. Use of any forward model should be preceded by a detailed screening program to identify major characteristics of meteorology of the area (6.1).
3. Any assessment team should have access to trained modellers at the relevant scales (4.2.3).
4. The best possible inventory of all important emissions should be obtained but it should also be realised that this will be incomplete and contested so assessments must include higher and lower case scenarios (5.3, 9.1).
5. Inverse methods at this scale are still a developing research area. Thus they should be tested in one gas field to see whether they can detect the integrated emissions (5.9). Ground truth for this test should be provided by a detailed survey
6. Given the success of airborne measurements in the U.S. We should also test the ability of atmospheric measurements from aircraft upwind and downwind of the gas-field to estimate emissions (5.8).
7. There should be a standing capability for surveying gas-fields in detail (5.7).
8. A follow-up study should combine information on likely emission rates, atmospheric transport and health impacts to generate a risk assessment (9.1).

References

- Aubinet, M., T. Vesala, and D. Papale (2012), *Eddy Covariance: A Practical Guide to Measurement and Data Analysis*, Springer Atmospheric Sciences, Springer.
- Baklanov, A.: Chemical weather forecasting: a new concept of integrated modelling, *Adv. Sci. Res.*, *4*, 23-27, doi:10.5194/asr-4-23-2010
- Bolin, B., and C. D. Keeling (1963), Large-scale atmospheric mixing as deduced from the seasonal and meridional variations of carbon dioxide, *J. Geophys. Res.*, *68*, 3899–3920.
- Brandt, J., Christensen, J. H., Frohn, L. M., and Zlatev, Z.: Numerical modelling of transport, dispersion, and deposition - validation against ETEX-1, ETEX-2 and Chernobyl, *Environmental Modelling and Software*, *15*, 6-7, 521-531.
- Brandt, J.: Modelling transport, dispersion, and deposition of passive tracers from accidental releases. Ph.D. thesis, National Environmental Research Institute, Denmark.
- Brandt, J., Bastrup-Birk, A., Christensen, J.H. , Mikkelsen, T., Thykier-Nielsen, S., and Zlatev, Z.: Testing the importance of accurate meteorological input fields and parameterizations in atmospheric transport modelling using DREAM - validation against ETEX-1, *Atmospheric Environment*, *32*, 24, 4167-4186.

- Carmichael G. R., Peters L. K., and Saylor R. D.: The STEM-II regional scale acid deposition and photochemical oxidant model -I: An overview of model development and applications, *Atmospheric Environment*, 25A, 2077-2090.
- Chevallier, F., N. Viovy, M. Reichstein, and P. Ciais (2006), On the assignment of prior errors in bayesian inversions of CO₂ surface fluxes, *Geophys. Res. Let.*, 33, L13,802, doi:10.1029/2006GL026496.
- Chevallier, F., et al. (2010), CO₂ surface fluxes at grid point scale estimated from a global 21-year reanalysis1 of atmospheric measurements, *J. Geophys. res.*, 115, submitted.
- Christoudias, T., and Lelieveld, J.: Modelling the global atmospheric transport and deposition of radionuclides from the Fukushima Dai-ichi nuclear accident, *Atmospheric Chemistry and Physics*, 13, 1425-1438, 2013
- Cox, W.M., Tikvart, J.A.: A statistical procedure for determining the best performing air quality simulation model, *Atmospheric Environment*, 24A, 2387-2395, 1990.
- Draxler, R.R. and Rolph, G.D., 2013. HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) Model access via NOAA ARL READY Website (<http://www.arl.noaa.gov/HYSPLIT.php>), NOAA Air Resources Laboratory, College Park, MD.
- Emmons, L. K., Walters, S., Hess, P. G., Lamarque, J. F., Pfister, G. G., Fillmore, D., Granier, C., Guenther, A., Kinnison, D., Laepple, T., Orlando, J., Tie, X., Tyndall, G., Wiedinmyer, C., Baughcum, S. L., and Kloster, S.: Description and evaluation of the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4), *Geosci. Model Dev.*, 3, 43-67.
- Enting, I. G. (2002), *Inverse Problems in Atmospheric Constituent Transport*, 392 pp., Cambridge University Press.
- Evans, S. N., and P. Stark (2002), Inverse problems as statistics, *Inverse problems*, 18, R55–R97.
- Fan, S., M. Gloor, J. Mahlman, S. Pacala, J. Sarmiento, T. Takahashi, and P. Tans (1998), A large terrestrial carbon sink in North America implied by atmospheric and oceanic CO₂ data and models, *Science*, 282, 442–446.
- Fast, J.D., Gustafson Jr., W. I., Easter, R. C., Zaveri, R. A., Barnard, J. C., Chapman, E. G., Grell, G. A., and Peckham, S. E.: Evolution of ozone, particulates, and aerosol direct radiative forcing in the vicinity of Houston using a fully coupled meteorology-chemistry-aerosol model, *J. Geophys. Res.*, 111, 2006.
- Fung, I., John, J., Lerner, J., Matthews, E., Prather, M., Steele, L. P., and Fraser, P. J.: Three-Dimensional Model Synthesis of the Global Methane Cycle, *J. Geophys. Res.*, 96, 13033-13065.
- Geels, C., et al. (2006), Comparing atmospheric transport models for future regional inversions over europe. part 1 : Mapping the CO₂ atmospheric signals, *Atmos. Chem. Phys. discuss.*, 6, 3709–3756.
- Grell, G. A., S. E. Peckham, R. Schmitz, S. A. McKeen, G. Frost, W. C. Skamarock and B. Eder, 2005: Fully coupled online chemistry within the WRF model, *Atmos. Environ.*, 39, 6957-6975.

- Grell, G. A., Dudhia, J., Stauffer, D. R.: A description of the fifth-generation Penn State/NCAR Mesoscale Model (MM5), NCAR/TN-398+STR, NCAR Technical note, Mesoscale and Microscale Meteorology Division, National Center for Atmospheric Research, Boulder, CO.
- Gurney, K. R., et al. (2002), Towards robust regional estimates of CO₂ sources and sinks using atmospheric transport models, *Nature*, *415*, 626–630.
- Hammer, S., et al.: Feasibility study of using a "travelling" CO₂ and CH₄ instrument to validate continuous in situ measurement stations, *Atmos. Meas. Tech.*, *6*, 1201–1216.
- Hollingsworth, A., Engelen, R. J., Textor, C., et al. Toward a monitoring and forecasting system for atmospheric composition: The GEMS project, *Bulletin of the American Meteorological Society* *89*, 8, 1147–.
- Kaminski, T., M. Heimann, and R. Giering (1999a), A coarse grid three-dimensional global inverse model of the atmospheric transport, 1, Adjoint model and Jacobian matrix, *J. Geophys. Res.*, *104*(D15), 18,535–18,553.
- Kaminski, T., M. Heimann, and R. Giering (1999b), A coarse grid three-dimensional global inverse model of the atmospheric transport, 2, Inversion of the transport of CO₂ in the 1980s, *J. Geophys. Res.*, *104*(D15), 18,555–18,582.
- Hurley, P.: The Air Pollution Model (TAPM) Version 3. Part 1: Technical Description, CSIRO Atmospheric Research Technical Paper No. 71.
- Hurley, P.J., Manins, P.C., Lee, S., Boyle, R., Ng, Y, and Dewundege, P.: 'Year-long, high-resolution, urban air shed modelling: Verification of TAPM predictions of smog and particles in Melbourne, Australia', *Atmos. Environ.*, *37*, 1899–1910.
- Karion, A., et al. (2013), Methane emissions estimate from airborne measurements over a western united states natural gas field, *Geophysical Research Letters*, pp. n/a–n/a, doi:10.1002/grl.50811.
- Kolb, H., Seibert, P., Zwatz-Meise, V., Mahringer, G.: Comparison of trajectories calculated during and after the Chernobyl accident. In: Evaluation of atmospheric models applied to the release from Chernobyl. *sterreichische Beitrage zu Meteorologie und Geophysik*, *1*, 61–69.
- Lauvaux, T., et al. (2008), Mesoscale inversion: First results from the CERES campaign with synthetic data, *Atmos. Chem. Phys.*, *8*, 3459–3471.
- Lauvaux, T., et al. (2009), Bridging the gap between atmospheric concentrations and local ecosystem measurements, *Geophys. Res. Lett.*, *36*, L19,809, doi:10.1029/2009GL039574.
- Lauvaux, T., et al. (2012), Constraining the CO₂ budget of the corn belt: exploring uncertainties from the assumptions in a mesoscale inverse system, *Atmospheric Chemistry and Physics*, *12*(1), 337–354, doi:10.5194/acp-12-337-2012.
- Law, A.M., Simulation model's level of detail determines effectiveness, *Industrial Engineering*, *23*, 16–18 (1991).
- Law, R. M., et al. (1996), Variations in modelled atmospheric transport of carbon dioxide and the consequences for CO₂ inversions, *Global Biogeochem. Cycles*, *10*, 783–796.

- Law, R. M., et al. (2008), TransCom model simulations of hourly atmospheric CO₂: experimental overview and diurnal cycle results for 2002, *Global Biogeochem. Cycles*, *22*, GB3009, doi:10.1029/2007GB003050.
- Loh, Z., R. Leuning, S. Zegelin, D. Etheridge, M. Bai, T. Naylor, and D. Griffith (2009), Testing lagrangian atmospheric dispersion modelling to monitor CO₂ and CH₄ leakage from geosequestration, *Atmospheric Environment*, *43*, 2602–2611, doi:10.1016/j.atmosenv.2009.01.053.
- Lorimer G, 1986.: The AUSPLUME Gaussian-plume Dispersion Model. EPA/86-02, Environment Protection Authority: Melbourne.
- Michalak, A. M., L. Bruhwiler, and P. P. Tans (2004), A geostatistical approach to surface flux estimation of atmospheric trace gases, *J. Geophys. Res.*, *109*, D14,109, doi: 10.1029/2003JD004422.
- Mosca, S., Graziani, G., Klug, W., Bessasio, R. and Bianconi, R.: ATMES-II - Evaluation of long-range dispersion models using first ETEX release data, EUR 17756/EN, Luxemborg: Office for Official Publications of the European Commission
- K. Nodop (Ed.): Proceedings of the ETEX Symposium on Long-range Atmospheric Transport, Model Verification and Emergency Response, 13-16 May, Vienna, Office for Official Publications of the European Communities, Luxembourg (1997)
- Petron, G., et al. (2012), Hydrocarbon emissions characterization in the colorado front range: A pilot study, *Journal of Geophysical Research: Atmospheres*, *117*(D4), n/a–n/a, doi:10.1029/2011JD016360.
- Peylin P., Bousquet P., Quere C. L., Sitch S., Friedlingstein P., McKinley G., Gruber N., Rayner P., and Ciais P.: Multiple constraints on regional CO₂ flux variations over land and oceans, *Global Biogeochemical Cycles*, *19*,1, GB1011.
- Pfister, G. G., Parrish, D. D., Worden, H., Emmons, L. K., Edwards, D. P., Wiedinmyer, C., Diskin, G. S., Huey, G., Oltmans, S. J., Thouret, V., Weinheimer, A., and Wisthaler, A.: Characterizing summertime chemical boundary conditions for airmasses entering the US West Coast, *Atmos. Chem. Phys.*, *11*, 1769–1790.
- Pillai, D., C. Gerbig, R. Kretschmer, V. Beck, U. Karstens, B. Neininger, and M. Heimann (2012), Comparing lagrangian and eulerian models for co₂ transport a step towards bayesian inverse modeling using wrf/stilt-vprm, *Atmospheric Chemistry and Physics*, *12*(19), 8979–8991, doi: 10.5194/acp-12-8979-2012.
- Pleim, J. E., Chang. J. S. and Zhang, K.: A nested grid meso-scale atmospheric chemistry model, *Journal of Geophysical Research*, *96D*, 3065-3084.
- Raupach, M. R., P. J. Rayner, D. J. Barrett, R. DeFries, M. Heimann, D. S. Ojima, S. Quegan, and C. Schmullius (2005), Model-data synthesis in terrestrial carbon observation: Methods, data requirements and data uncertainty specifications, *Glob. Change Biol.*, *11*, 378–397.
- Rayner, P. J., M. R. Raupach, M. Paget, P. Peylin, and E. Koffi (2010), A new global gridded dataset of CO₂ emissions from fossil fuel combustion: 1: Methodology and evaluation, *J. Geophys. Res.*, *115*, D19,306, doi:10.1029/2009JD013439.

- Rayner, P. J., I. G. Enting, and C. M. Trudinger (1996), Optimizing the CO₂ observing network for constraining sources and sinks, *Tellus*, 48B, 433–444.
- Rayner P. J., Law, R. M., Allison, C. E., Francey, R. J., Trudinger, C. M., and Pickett-Heaps, C.: Interannual variability of the global carbon cycle (1992-2005) inferred by inversion of atmospheric CO₂ and ¹³CO₂ measurements, *Global Biogeochem. Cy.*, 22, GB3008, doi:10.1029/2007GB003068
- Reynolds S. D., Roth P. M. and Seinfeld J. H.: Modeling of photochemical air pollution -I. Formulation of the model, *Atmospheric Environment*, 7, 1033-1061.
- Reynolds S. D., Liu M. K., Hecht T. A., Roth P. M. and Seinfeld J. H.: Modeling of photochemical air pollution - III. Evaluation of the model. *Atmospheric Environment*, 8, 563-596.
- Roselle, S. J., Pierce, T. E. and Schere K. L.: The sensitivity of regional ozone modeling to biogenic hydrocarbons. *Journal of Geophysical Research*, 96, 7371-7394.
- Rödenbeck, C., C. Gerbig, K. Trusilova, and M. Heimann (2009), A two-step scheme for high-resolution regional atmospheric trace gas inversions based on independent models, *Atmos. Chem. Phys.*, 9, 5331–5342.
- Schuh, A. E., et al. (2013), Evaluating atmospheric CO₂ inversions at multiple scales over a highly-inventoried agricultural landscape, *Glob. Change Biol.*, 19, doi:10.1111/gcb.12141, (in press).
- Scire, J., Strimaitis, D., Yamartino, R., 1999. A User's Guide for the CALPUFF Dispersion Model (Version 5.0), Earth Tech, Concord, Mass.
- Seigneur, C., Pun, B., Pai, P., Louis, J. F., Solomon, P., Emery, C., Morris, R., Zahniser, M., Worsnop, D., Koutrakis, P., White, W., Tombach, I.: Guidance for the performance evaluation of three-dimensional air quality modelling systems for particulate matter and visibility, *Journal of Air and Waste Management Association*, 50, 588-599, 2000.
- Sutton, O. G.: The problem of diffusion in the lower atmosphere, *Q. J. Roy. Meteorol. Soc.*, 73, 257-281.
- Tarantola, A. (2004), *Inverse Problem Theory and Methods for Model Parameter Estimation*, SIAM, (ISBN 0-89871-572-5).
- Tait D., Santos I., Maher D., Cyronak T., and Davis R.: Enrichment of Radon and Carbon Dioxide in the Open Atmosphere of an Australian Coal Seam Gas Field, *Environ Sci Technol.*, 47, 3099-3104, 2013.
- Tans, P. P., Fung, I. Y., and Takahashi, T.: Observational constraints on the global atmospheric CO₂ budget, *Science*, 247, 1431-1438.
- Utembe, S., Rayner, P., Genkova, I., Clark, A., Richardson, A., Lunney, C., Atkins, P., O'Brien, D., Jones, N., and Griffith, D.: Using WRF-Chem to estimate point source emissions from a power plant in Port Augusta, *in preparation*.
- van Dop H., Addis R., Fraser G., Girardi F., Graziani G., Inoue Y., Kelly N., Klug W., Kulmala A., Nodop K., Pretel J.: ETEX A European tracer experiment observations, dispersion modelling and emergency response, *Atmospheric Environment*, 32, 4089-4094, 1998.

- Turnbull, J. C., et al. (2011), Assessment of fossil fuel carbon dioxide and other anthropogenic trace gas emissions from airborne measurements over sacramento, california in spring 2009, *Atmospheric Chemistry and Physics*, 11(2), 705–721, doi:10.5194/acp-11-705-2011.
- Walmsley, J. L. and Wesely, M. L.: Modification of coded parameterizations of surface resistances to gaseous dry deposition, *Atmospheric Environment*, 30, 1996, 1181-1188.
- Vogel, H., Frstner, J., Vogel, B., Hanisch, T., Mhr, B., Schttler, U., and Schad, T.: Simulation of the dispersion of the Eyjafjallajökull plume over Europe with COSMO-ART in the operational mode, *Atmos. Chem. Phys. Discuss.*, 13, 13439-13463.
- Whelpdale, D. M.: Monitoring and assessment in Canada. In Pierce, R.C., Whelpdale, D.M., and Sheffer, M.G. (Eds.) Proceedings Symposium on Monitoring and Assessment of Airborne Pollutants, pp. 25-52. NRCC No. 20642. National Research Council of Canada, Ottawa.
- Willmott, C.: Some comments on the evaluation of model performance. *Bulletin of the American Meteorological Society*, 63, 1309-1313, 1982.
- Wu, L., M. Bocquet, F. Chevallier, T. Lauvaux, and K. Davis (2013), Hyperparameter estimation for uncertainty quantification in mesoscale carbon dioxide inversions, *Tellus*, 65b, accepted.

Glossary of Terms

Air parcel a quantity of air which is transported intact in the atmosphere.

Advection transport by the wind

Boundary conditions The values of atmospheric variables at the edges of the domain

Domain area over which the model is run

Jacobian the sensitivity of an observation to a change in a source at a point

Numerical diffusion a computational artefact where strong features are smeared out in the model. It is particularly serious when we wish to calculate peak exposure.

Online the CTM is run as part of the meteorological model.

Offline the CTM is run separately, reading meteorological data from a previous run of the meteorological model.

Parameterization Description of atmospheric processes which are too small to be handled directly in the model.

Partial differential equation An equation describing how some quantity changes in both space and time

PDF Probability Density Function, a function that described the relative likelihood for a random variable to take on a given value

Prior the value we assume for the source before running an inverse model

Source-receptor relationship the amount by which the concentration changes at a (receptor) point for a change in the source at a (source) point. It can be used both to calculate exposure from a given source and also to quantify a source using observations in an inverse model.