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Data assimilation: a powerful tool for atmospheric chemistry

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Issues such as ozone depletion, acid rain and photochemical smog are all of considerable environmental importance. These issues are studied using the dual approach of observations and numerical modelling. In making balanced assessments of these issues it is vital to make the best use of *all* the information available to us, both theoretical and observational. This is a non-trivial task. The technique of '*data assimilation*' is a powerful tool that allows us to address this issue. It is revolutionizing the way we can study atmospheric chemistry. Data assimilation allows us to simultaneously make good use of however many observations are available to us, our theoretical understanding, and any *a priori* information we have, within a mathematical framework. It even allows us to infer information about chemical constituents that are *not* observed. As we move into the new millennium it is a technique that is set to grow rapidly in importance.

Keywords: chemical data assimilation; 4D-Var; ozone depletion; atmospheric chemistry

1. Introduction

The global distribution of ozone and other atmospheric constituents has changed considerably over the last decade, with record low global ozone levels in the last few years and downward trends over much of the globe (WMO 1992, 1994). The analysis of the changes in the ozone fields, and of atmospheric chemistry in general, is severely hampered by a lack of consistent datasets, and, especially, from the lack of insight in to the role of chemistry and dynamics on the behaviour of ozone. Chemical data assimilation allows us to dramatically improve this state of affairs.

In order to be able to characterize and to predict the evolution of the Earth's atmosphere one has to know its present state. Since the 1960s, one of the most important sources of data on our atmosphere has been from satellites. The ability of satellites to observe the Earth ranges from nearly complete global coverage every day, to continuous observations of a particular part of the Earth. Satellites offer the only practical method of obtaining valuable data over much of the world, especially the oceans and remote land areas. The Earth Observing System (EOS) and ENVISAT, both of which will be launched in the next decade, will provide the most complete set of Earth observations ever taken.

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The intelligent use of these data on a wide variety of chemical constituents, measurements that have cost many millions of dollars/pounds to make, is a non-trivial task as the observations are not collocated in time or space. Satellites make measurements of atmospheric constituents by a range of methods, and at a range of times and locations. The measurements are not made on a regular spatial grid or at the same times of day. Since the analysis of satellite measurement is so complex, the measurements have not been used to their full potential.

2. Context

The analysis of chemical trace species has received little attention in comparison with the analysis of meteorological variables. Current methods tend to treat species independently, ignoring the complex balances that exist between species. Moreover, the large diurnal variations in the concentrations of many species are either accounted for in very simple ways, or avoided by analysing concentrations at fixed local time. This is a great shame as the shape of a species' diurnal cycle, and the relative partitioning between species, contains a lot of valuable information that is completely wasted if we do not use a technique that can exploit this information. Naturally, such information can only be exploited if it includes a theoretical understanding of the chemical system.

3. The objective

The specific objective of chemical data assimilation is to produce a self-consistent picture of the chemical state of our atmosphere as synoptic analyses. A synoptic analysis is a term that means a general view of the atmosphere at a given moment in time over a broad area. When we say self-consistent, we mean consistent both with the observations available to us and their associated errors *and* our theoretical understanding of the chemical system as encapsulated by a deterministic model. This involves information on a large number of chemical species at many different locations. Needless to say, such an exercise is valuable as it highlights areas of improvement in both the observations and our theoretical understanding.

However, obtaining such analysis is not the goal, but, rather, a means to reach our goal. Our goal being a comprehensive understanding of the processes involved and their environmental implications. With the advent of chemical data assimilation we can, for the first time, produce such comprehensive, self-consistent, synoptic, chemical analysis of atmospheric constituents and then start to *use* them to examine, in unprecedented detail, the chemical mechanisms involved in such important environmental issues as ozone depletion.

Figure 1. (*a*) shows an example trajectory of an air parcel in the upper stratosphere. The diamonds along the trajectory indicate the locations for which Upper Atmosphere Research Satellite (UARS) observations were available. (*b*) The ATMOS instrument in the payload bay of the Shuttle Atlantis. This picture comes from the ATMOS ATLAS 1 Mission Operations web page. (*c*)–(*h*) The background colours correspond to the analysis produced by data assimilation of NO, NO_2, N_2O_5, $HONO_2$, $ClONO_2$ and HCl of the ATMOS data for 29 March 1992. The colour scale indicates the concentration as a volume mixing ratio (VMR). The horizontal axis in each case is the local solar time (LST), and the vertical axis is height expressed as a potential temperature (K). The instrument only makes measurements at sunrise and sunset, as indicated by the colour-filled triangles, whereas we can see that the technique of data assimilation has allowed us to fill in values throughout the day.

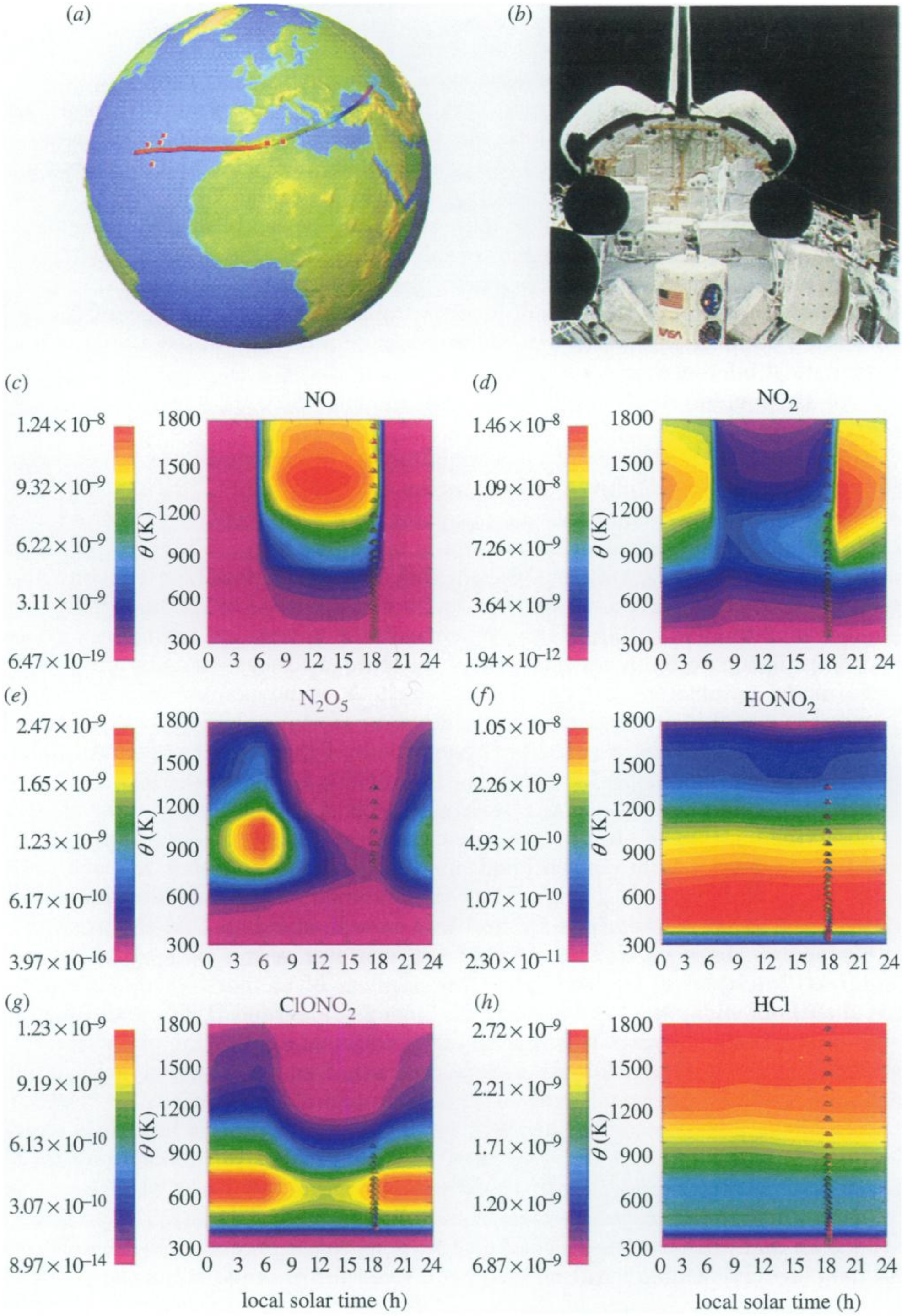

Figure 1. For description see opposite.

4. What is data assimilation?

Data assimilation is a mathematical technique that allows us to use all the information available to us within a time window. This includes observational data, any *a priori* information we may have, and, very importantly, a deterministic model that describes our system and encapsulates our theoretical understanding. Data assimilation is a Bayesian method, so called after the English theologian and mathematician Thomas Bayes (born 1702, in London, died 1761, in Tunbridge Wells, Kent). Thomas Bayes was the first to use probability inductively and he established a mathematical basis for probability inference (a means of calculating, from the frequency with which an event has occurred in prior trials, the probability that it will occur in future trials). Bayes's theorem, or the laws of inverse probability, allow one to combine prior information about a parameter with information contained in observations to guide the statistical inference process. The key benefit is that the Bayesian approach permits the use of objective data *and* subjective opinion where the subjectivity is made explicit.

So the mathematical basis of data assimilation is estimation theory or inverse problem theory, a good introduction is given by Cohn (1997). Estimation theory provides a natural mathematical foundation for data assimilation; it is an organized set of mathematical techniques for obtaining useful information about the physical world on the basis of observations (Menke 1984). In a conventional problem one would use a set of known, *a priori*, parameters to predict the state of the physical system. Usually called the 'forward' problem. Whereas in the 'inverse' or estimation problem we attempt to use available observations of the state of the system to estimate poorly known model parameters and/or the state itself. A prime example of where such a technique is useful is in the study of satellite data of atmospheric constituents.

The UARS has provided a rich set of observations of trace-species concentrations in the middle atmosphere (see, for example, Reber 1993; Reber *et al.* 1993). The asynoptic nature of the observations, the precessing orbit of the satellite, and the ability of the observing instruments to produce collocated observations of several species highlight the shortcomings of conventional analysis techniques. Fisher & Lary (1995) discuss the technique of Lagrangian four-dimensional variational data assimilation (4D-VAR) in detail, an analysis method that is well suited to UARS observations, and asynoptic observations in general, from whatever source. The 4D-VAR technique was developed in the context of the analysis of meteorological variables by several authors, including Lewis (1985), Ledimet & Talagrand (1986) and Talagrand & Courtier (1987). The method may be regarded as an application of the theory of optimal control (Lions 1971). 4D-VAR is currently being considered by a number of centres for the operational analysis of meteorological observations for numerical weather prediction. The method has been applied to the analysis of humidity; treated as a passive tracer (Andersson *et al.* 1992). A comprehensive bibliography for the subject is given by Courtier *et al.* (1993) and an overview by Cohn (1997).

The reason that the technique of data assimilation is so effective is that it seeks to produce an analysis that fits a set of observations taken over a 'time window' (not just the observations made at one instant in time, for example along the air-parcel trajectory indicated in figure 1*a*), subject to the strong constraint that the evolution of the analysed quantities is governed by a deterministic model describing the given observations.

By imposing the equations of the model as strong constraints, the analysis problem is reduced to that of determining initial values for the model such that the subsequent evolution minimizes a measure of the fit to the observations. The analysis method is, therefore, able to elegantly exploit information which is not available to conventional analysis techniques, such as the shape of the diurnal cycles of atmospheric constituents. Thus, using our knowledge of the processes involved intelligently has allowed us to extract much more information from the observations.

As a result, asynoptic satellite observations made at whatever location or time within a 'time window' can be used to produce a set of self-consistent synoptic analyses of the observed species. In addition, synoptic analyses can be inferred for species included within the model, even though they are not actually observed. In other words, an intelligent use of our knowledge of the processes involved extends the information contained within the observations to infer observations not made. This can itself be used as a stringent test of the technique.

Figure 1 shows an example of this using the technique of data assimilation on data from the space-shuttle born Atmospheric Trace Molecule Spectroscopy (ATMOS) instrument. The colour scale indicates what concentration each colour corresponds to. The horizontal axis in each case is the local solar time (LST), and the vertical axis is height expressed as a potential temperature (K). The instrument only makes measurements at sunrise and sunset, as indicated by the colour-filled triangles. We can see that the technique of data assimilation has allowed us to fill in values for throughout the day. We also notice in figure 1*e* how much fewer observations of N_2O_5 there are than NO_2 (figure 1*d*), yet the 4D-VAR technique has still done a good job of filling in the 'missing' N_2O_5 points as it has information on N_2O_5 from observations of other species that react with N_2O_5 or form it, and from the numerical model that contains our theoretical understanding of the chemistry. It is quite remarkable the way in which we can make good use of such sparse observational data. As King Solomon wisely observed three millennia ago in the middle of the 10th century BC: 'wisdom is profitable to direct' (Ecclesiastes 10:10).

(*a*) *Chemical applications of* 4D-VAR

The technique of data assimilation was first applied to atmospheric chemistry by Fisher & Lary (1995). They used the data-assimilation technique to produce synoptic analysis of chemical species from asynoptic satellite data. In addition, the method allows many useful insights to be gained that cannot be obtained by any of the other techniques currently available. The work of Fisher & Lary (1995) has now been considerably extended to have a more detailed chemical scheme. Since this, other 4D-VAR chemical studies have been performed, such as those of Elbern *et al.* (1997), Levelt *et al.* (1996, 1998), Riishojgaard (1996), Lyster *et al.* (1997), LeMarshall *et al.* (1997), Eskes *et al.* (1998) and Khattatov *et al.* (1999). A look at the future of data assimilation and its uses is given in the editorial by Kalnay *et al.* (1997).

Khattatov *et al.* (1999) investigated the validity of the linear approximation of the stratospheric photochemical box model. They presented the time evolution of the linearization and error covariance matrices and investigated some of their properties. It was found that the rank of the linearization matrix decreases rapidly. This indicated that the concentration of only a few of the species need to be measured to predict the state of the stratospheric chemical system in question. Based on the ideas

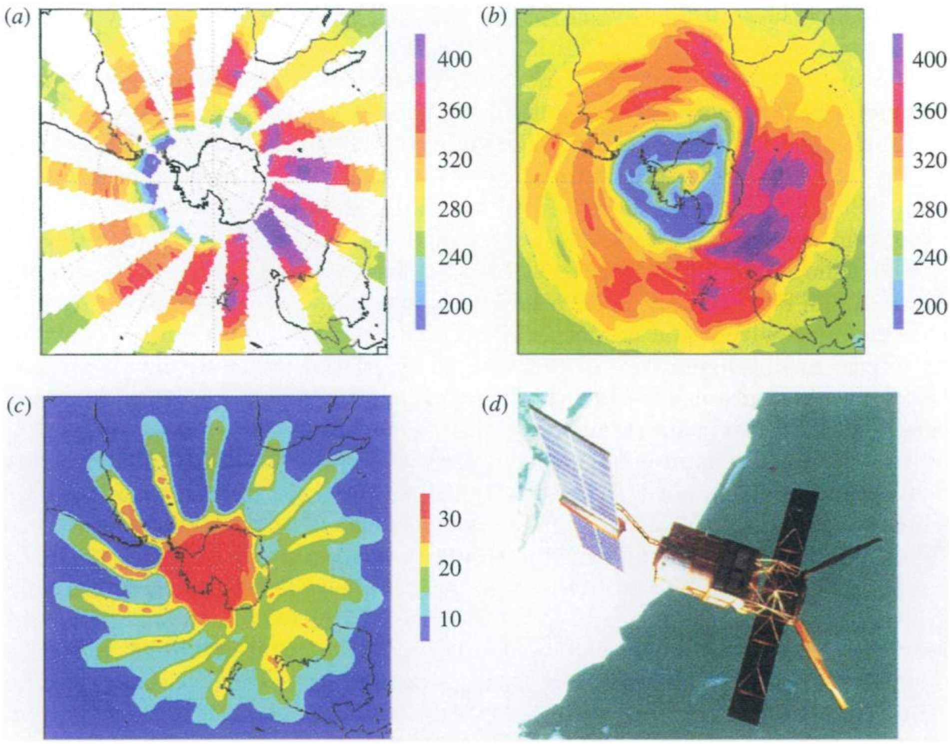

Figure 2. The ozone hole of September 11, 1996. The Southern Hemisphere GOME total ozone data used as input for the assimilation are shown in (*a*); and (*b*) is the assimilated field at 12.00 GMT. The corresponding uncertainty distribution is shown in (*c*). All scales are in Dobson units. (*d*) is an artist's impression of the ESA Remote Sensing Satellite (ERS) in orbit (from the ESA Web site). (*a*)–(*c*) are from Eskes *et al.* (1998).

of Fisher & Lary (1995), they employed a trajectory model and the photochemical box model for assimilation and mapping of the UARS measurements of chemical species. They produced an assimilation using both the variational technique and the Kalman filter.

(i) *A regional acid-deposition model*

Elbern *et al.* (1997) used a comprehensive tropospheric gas-phase chemical mechanism in the regional acid-deposition model (RADM2) that comprises 157 gas-phase reactions with 63 model species. They showed that 4D-VAR can produce realistic analyses of unmeasured species, but chemically coupled to those observed with a time-series of only one or very few species. However, the numerous volatile organic compounds cannot be analysed without *a priori* knowledge.

Figure 3. On the left-hand side, the rate of propagation of the NO_x, HO_x, ClO_x and BrO_x ozone loss catalytic cycles, based on 4D-VAR chemical analysis of ATLAS-1/ATMOS data in an equivalent latitude band centred on 40° S, for 29 March 1992 is shown. On the right-hand side, the corresponding chain length (how many times the catalytic cycle is executed before the radical is removed) for the NO_x, HO_x, ClO_x, and BrO_x cycles is shown.

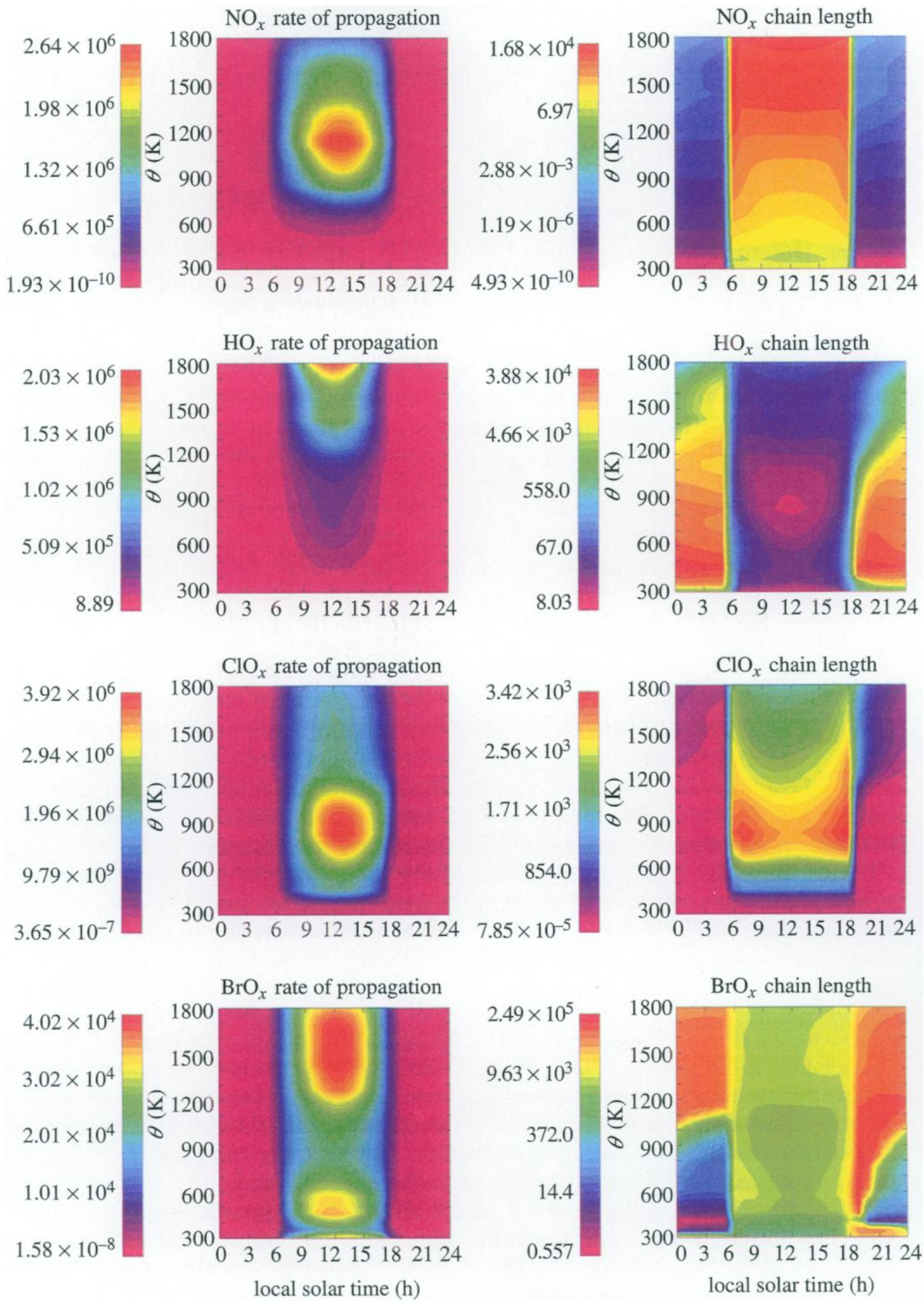

Figure 3. For description see opposite.

(ii) *Meteorological agencies*

Meteorological agencies such as the European Centre for Medium Range Weather Forecasting (ECMWF), the United Kingdom Meteorological Office (UKMO), Météo France, and the NASA Data Assimilation Office (DAO) are also examining assimilating observations of ozone to aid in improving their weather forecasts. They have found that ozone data appear to be able to provide valuable information on wind fields. The most information is gained in areas where the ozone horizontal gradients are strong.

At the Dutch Meteorological Institute (KNMI), 4D-VAR has been used for the assimilation of total ozone data from the Global Ozone Monitoring Experiment (GOME). GOME is a satellite instrument on board the European satellite ERS-2. Piters *et al.* (1996), Levelt *et al.* (1996) and Eskes *et al.* (1998) have found that combining GOME-retrieved ozone with a global tracer advection model by means of data assimilation extracts a maximum amount of information from the satellite data (figure 2). It results in almost global ozone fields at any moment in time, taking advantage of measurements from previous days. The variational assimilation approach makes very efficient use of the data because of its ability to incorporate both past and future measurements into the analysis, thereby reducing the estimated error. Apart from delivering total ozone fields, they were able to make estimates of the space- and time-dependent accuracy of the total ozone fields, which makes the results useful for data-validation studies. Near real time assimilated ozone are provided via the KNMI Web site http://www.knmi.nl/onderzk/atmosam/GOME.

(iii) *A three-dimensional global model*

Levelt *et al.* (1998) implemented a method for assimilating observations of ozone into the three-dimensional global stratospheric chemistry transport model ROSE. The model contains an extensive photochemical scheme that includes heterogeneous chemistry and uses temperature and wind fields from the UKMO stratospheric analysis. Ozone measurements obtained by the microwave limb sounder (MLS) on board the UARS were assimilated in the model using the sequential statistical interpolation approach. The stratospheric total ozone fields computed from the analysis were compared with the TOVS total ozone measurements and it is shown that they agree within the uncertainty of the data.

(iv) *The Lagrangian approach*

If the analysis of dynamical and chemical variables is performed separately, a considerable reduction in computational cost results. This is because it is necessary neither to model the entire three-dimensional domain, nor to include a dynamical model in the iterative analysis procedure. However, by taking a 'Lagrangian' approach, and making a separation between the chemical and dynamical analyses, we are prevented from utilizing some useful information. Specifically, observations of chemical species contain information on the wind and temperature distributions, which is ignored when one takes the Lagrangian approach. Nonetheless, a complete Lagrangian treatment of the chemistry within the data-assimilation framework is such a major advance over previous methods of analysis that much useful information can be made, as we have already seen from the results shown in figure 1.

When using the Lagrangian approach we use a photochemical 'box' model. That is, a model that simulates the evolution of chemical trace species for a number of independent air parcels whose trajectories are assumed to be known *a priori*. Usually, we calculate them from the relevant meteorological analyses.

Figure 1 shows an example trajectory of an air parcel in the upper stratosphere in order to illustrate how the technique of 4D-VAR data assimilation works. In this example, the trajectory is coloured by the temperature of the air parcel, a key variable as far as the chemistry is concerned. The diamonds along the trajectory indicate the locations for which UARS observations were available. We note that the observations are not at regular intervals along the trajectory, and also that they are rather sparse. As already mentioned, this is a major obstacle for previous analysis techniques. However, 4D-VAR allows us to take such asynoptic observations and produce a synoptic analysis.

(v) *A case study*

Now that we have looked at the principles of 4D-VAR data assimilation, let us look at a case study using data from the ATMOS experiment. ATMOS is an infrared Fourier transform interferometer that has, on four occasions, flown in the payload bay of the space shuttle and measures the concentrations of gases present in the atmosphere at altitudes of between 10 and 150 km. As the shuttle's orbit carries it into and out of the Earth's shadow, the ATMOS instrument views the Sun as it sets or rises through the atmosphere. The spectrometer measures changes in the infrared component of sunlight as the Sun's rays pass through the atmosphere. Trace gases absorb very specific wavelengths, which allows us to determine what gases are present, in what concentrations and at what altitudes. ATMOS has flown several times. More information on ATMOS can be found on the Web site at http://remus.jpl.nasa.gov/.

The STS-45/ATLAS 1 mission was launched on 24 March 1992 from the Kennedy Space Center. During eight days of operation, the ATMOS instrument made a total of 98 observations, spanning a substantial portion of the globe. The 53 measurements taken at orbital sunrise covered the mid-latitude and equatorial regions of the Earth from 30° S to 30° N. The 41 sunset observations were made at 25° S to 55° S. ATMOS was only able to monitor the atmosphere down to a height of about 20 km, due to a recent eruption of Mount Pinatubo, which clouded the region below that with dust and aerosols.

Over this period, the equivalent PV latitude for which the vertical profiles covered the largest range of altitudes, and for which the largest number of species was observed was centred on *ca.* 40° S. For this equivalent PV latitude a detailed chemical analysis was performed on the basis of our chemical data assimilation. To perform this analysis a chemical scheme containing a total of 59 species, and 357 reactions, was used.

Figure 1 shows some of the results from these analyses. The vertical axis in each case is the potential temperature, θ, in K, the horizontal axis is the local solar time in hours. Overlaid on the analyses are the observations made by ATMOS. As the ATMOS instrument uses solar occultation, the observations are only available at sunrise or sunset. It can be seen that the 4D-VAR was capable of simultaneously reproducing many of the observations made by ATMOS. For example, if we look at the NO and NO_2 distributions in figure 1*c*, *d* we can see that the 4D-VAR analysis has captured the concentrations of NO and NO_2 at sunset over the entire altitude range remarkably well (i.e. the colours of the filled triangles correspond to the background colours). We see a similar agreement for $ClONO_2$. This indicates that both the observations and model are of a reasonably good quality.

Likewise, the overall features of the $HONO_2$ and HCl distributions shown in figure $1f, h$, respectively, are reproduced by 4D-VAR analysis. However, when one looks more closely at the $HONO_2$ distribution in the lower stratosphere in figure $1f$, we notice that the optimal model simulation, i.e. our 4D-VAR analysis, overestimates the amount of $HONO_2$ present. This is a well-known deficiency of models, which tend to overestimate the amount of $HONO_2$ in the atmosphere (Chatfield 1994). It also shows how 4D-VAR can highlights deficiencies in our theoretical understanding.

Now that we have been able to produce a self-consistent analysis, we can use it to examine, in detail, the ozone loss mechanisms. Ozone loss takes place via various catalytic cycles. The importance of atmospheric catalytic cycles was first recognized by Bates & Nicolet (1950). Since then, it has become well established that the concentration of stratospheric ozone is controlled by the balance between its production and its destruction, and that the destruction of ozone is mainly due to catalytic cycles involving nitrogen, hydrogen, chlorine and bromine species.

The effectiveness of catalytic cycles in destroying ozone is controlled by two factors: the chain length of the catalytic cycles; and the abundance of the radical that is the chain centre. The chain length is the number of times the catalytic cyclic is executed before the reactive radical involved is destroyed. To date, the chain length of catalytic ozone destruction cycles has received relatively little attention, with emphasis being placed almost exclusively on the abundance of the chain centres involved. Lary (1997) presents a systematic study of the effectiveness of ozone destruction cycles in the stratosphere. On the left-hand side of figure 3 the rate of propagation of the NO_x, HO_x, ClO_x and BrO_x ozone loss catalytic cycles, based on the 4D-VAR chemical analysis of ATLAS-1/ATMOS data in an equivalent latitude band centred on 40° S, for 29 March 1992 is shown. On the right-hand side the corresponding chain length (how many times the catalytic cycle is executed before the radical is removed) for the NO_x, HO_x, ClO_x and BrO_x cycles is shown.

Although the full chemical analysis of these data is beyond the scope of this article, it is very exciting to see that we are now able to extract such a level of detail from atmospheric observations.

5. Looking forward

The EOS and the ENVISAT, both of which will be launched in the next decade, will provide the most complete set of Earth observations ever taken. The 'state of the art' technique of data assimilation is the only way we currently have of making the best use of these data. In the next millennium, the importance of data assimilation is set to rise rapidly in the making of our scientific assessments, and the political decision-making that results from this. This is likely to be true on every level, from the global scale right down to the mesoscale, where we study the pollution in urban areas. Data assimilation will be most useful in improving our understanding of tropospheric chemistry, which is much more complex than that of the stratosphere. It is very likely that we will see chemical data assimilation being used to infer key parameters that are hard to measure directly, such as chemical emission rates.

Meteorological agencies are already starting to use observations of ozone in improving their weather forecasts. This is likely to be extended to other species as more satellite data become available and the power of computers increases.

In conclusion, we can say, with certainty, that the technique of data assimilation is opening up a whole new world of opportunities for us to study the chemistry of our atmosphere and to protect our environment. We will most certainly look back and see its advent as a milestone in atmospheric chemical research. A fact that has been recognized and supported by the Royal Society in a most timely way.

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Anyone interested in performing data assimilation is welcome to contact David Lary at the email addresses given at the front of this paper.

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AUTHOR PROFILE

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Born in Omdurman, Sudan, David John Lary is 34 and studied at King's College, London, where he graduated with first class joint honours in physics and chemistry with a Sambrooke Exhibition Prize for Natural Science in 1987. In 1991 he obtained his PhD in Modelling of Atmospheric Photochemistry at Churchill College, Cambridge, while at the Centre for Atmospheric Science. David was awarded a Royal Society University Research Fellowship at Cambridge to further his research in Atmospheric Chemical Data Assimilation, which was described as pioneering by the Editor of the *Quarterly Journal of the Royal Meteorological Society.* In 1998 he was also awarded an Alon Fellowship by the government of Israel, the highest honour for a young scientist in Israel. He is a senior lecturer in the Department of Geophysics and Planetary Sciences at Tel Aviv University. This is a joint appointment with Cambridge. He has published over 30 papers in the past 10 years. David has just married Tatiana, whose first language is Russian, but who is also fluent in English and Hebrew. Scientific interests include mathematical methods to make the best use of theoretical and observational information on the atmosphere; recreations include Bible study, music and photography.