ASSIMILATION OF ATMOSPHERIC COMPOSITIONS MEASUREMENTS FOR FUTURE ATMOSPHERIC CHEMISTRY MISSIONS

Executive Summary

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Assimilation of ATMOSPHERIC COMPOSITION MEASURMENTS for Future ATMOSPHERIC CHEMISTRY MISSIONS

EXECUTIVE SUMMARY

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Contents

1 Introduction

In 2000 ESTEC issued ITT AO/1-3608/99/NL/DC, which was entitled "Assimilation of Atmospheric Composition Measurements for Future Atmospheric Chemistry Missions". In July 2000 our consortium, was awarded the contract to carry out this study. The structure of our consortium comprised:

The Centre for Atmospheric Sciences, University of Cambridge

The School of the Environment, University of Leeds

The Atmospheric Science Division of the Space Science Department at RAL

Serco Europe Limited, who were prime contractor.

2 Objectives

The principal objective of this study was to develop data assimilation schemes for the purpose of validating measurements of minor species from new satellite instruments.

This objective was approached through three strands:

Strand 1: Aspects of conventional validation

Working in flow tracking coordinates in order too improve the focus of statistical measures (see subsection 3.2)

Compiling a Trace Gas Database using flow tracking coordinates (see subsection5)

Understanding better the limitations of existing observation systems used for validation (see subsection 5)

Understanding better the limitations of spatial and temporal coincidence (see subsection 7)

Strand 2: Assimilation of long-lived species into a 3D CTM

Investigating the potential of using a Chemical Transport Model for validation purposes (see subsection 11)

Strand 3: Assimilation into a detailed chemical assimilation model

Descriptions of assimilation machinery (see subsection 3.2)

By assessing diurnal cycles of certain species, and verify chemical self-consistency of observations and analyses (see subsection 6)

Assessing a Reference Atmosphere generated from quality controlled observations (see subsection 8)

Demonstrating an innovative "Bias Measurement Assistant" tool (see subsection 10)

3 Data Assimilation Machinery

3.1 Introduction

The major part of the work of this study used a set of assimilation machinery, which incorporated the AutoChem chemical model. This machinery was considered the most appropriate for the main validation activities. This approach is introduced and described in Section 3.2.

An alternative approach, which was very much less demanding upon computing resources, was also investigated. This approach used a set of assimilation machinery incorporating a 3D CTM. This approach is introduced and described in Section 3.3.

3.2 Assimilation Machinery using AutoChem

Lagrangian 4D data assimilation
For this machinery a Lagrangian approach is adopted: an ensemble of photochemical "box" models have been used. The model simulates the evolution of chemical trace species for a number of independent air parcels whose trajectories are assumed to be known *a priori*. The dynamics and chemistry are separated.

Sequential approach: Kalman-Bucy filter
In the sequential approach each observation is processed separately and the analysis length is the time between two consecutive observations. Model simulations at time t, the end of the analysis interval, are considered to be the a priori background estimate, $\mathbf{x}_\mathrm{b}$. The optimal analysis equation is used to obtain the best estimate of $\mathbf{x}$ at time t from the corresponding model value and observation. The result is used as the initial condition for model integration during the next analysis period, from t until the time of the next observation.

4D Var analysis method
In the variational approach all observation are processed simultaneously. This effectively avoids discontinuities in the resulting analyses. The machinery for the 4D Variational analysis used in this study is fully described in the main Final Report document. Although 4D Var can provide the most satisfactory analyses, it requires greater computing resources.

The AutoChem chemical model
The AutoChem model has been extensively validated. It is explicit and uses the adaptive-timestep, error monitoring, time integration scheme designed for stiff systems of equations (*Press et al.*, 1992).

The version of the model used here contains a total of 60 species. 55 species are time integrated, namely. The model contains a total of 420 reactions, 278 bimolecular reactions, 32 trimolecular reactions, 60 photolysis reactions, 4 cosmic ray processes, 46 heterogeneous reactions.

Flow tracking coordinates
Under adiabatic conditions air parcels move along isentropic surfaces. These are surfaces of constant potential temperature, θ. When considering tracer fields, θ is found to be a suitable vertical coordinate, since it acknowledges the likely vertical motion of air parcels.

Potential Vorticity (PV) plays a central rôle in large scale dynamics, where it behaves as an approximate material tracer. It is useful to normalise PV into the form of PV equivalent latitude (ϕ_e), and use it as a horizontal spatial coordinate instead of latitude and longitude. This reduces the tracer field from three dimensions to two.

In this study, we have taken these now well established ideas and used them as a framework for our chemical data assimilation.

Analysis Grid dimensions
The Reference Atmosphere is discussed in Section 8. The Reference Atmosphere is constructed on an Analysis Grid has the following characteristics:

(a) 21 potential temperature levels spaced equally in $\log(\theta)$ between 400 K and 2000 K

(b) 32 equivalent PV latitudes spaced evenly between -90° and 90°

(c) Local solar time every one hour (00, 01, etc)

Pseudo Observations
Pseudo Observations are generated by combining a set of real observations, which are centred on a specific location. Generating a Pseudo Observation is a necessary part of the assimilation procedure. This is because both the observation, which is to be assimilated, and resulting analyses need to be dealing with the same location. An additional advantage of using pseudo observations is that the combining of a number of real observations provides improved "signal-to-noise".

3.3 Assimilation Machinery using 3D CTM

Tools used to study the use of the CTM
For this part of the study, we combined a data assimilation scheme using a sub-optimmal kalman filter with the existing SLIMCAT 3D chemical transport model. SLIMCAT has already been used extensively for studies of stratospheric chemistry over multi-annual timescale.

By assimilating the observations into an established 3D model, our aim was not to produce daily, global fields of assimilated species with as little model content as possible, but to use the observations to continually 'nudge' the 3D model towards reality.

The SLIMCAT 3D CTM
Horizontal winds and temperatures are specified using meteorological analyses. Vertical advection is calculated from heating rates using the MIDRAD radiation scheme and chemical tracers are advected by conservation of second-order moments. The model has the most important species in the O_x, NO_y, Cl_y, Br_y, HO_x families along with a CH_4 oxidation scheme and long-lived tracers. The model has a detailed gas-phase stratospheric chemistry scheme as well as a treatment of heterogeneous chemistry of liquid and solid aerosols.

3.4 Assessing Quality of Assimilation

Once the analyses had been performed their quality was quantified by generating a set of statistics that compare the observations used in making the analyses with the analyses themselves.

The following measures were used:

(A-F) The analysis increment (A-F)is a good measure of model bias.

(A-O) The analysis-observation (A-O) is probably the best measure of analyses bias.

(O-F) The observation increment (O-F) is probably the best measure of forecast skill.

Scatter plots of observations against analyses were used to visually highlight any biases present.

Quantile-quantile plots of observation vs. analysis are usful for determining whether two samples come from the same distribution.

3.5 Points for Consideration

Use of tracer correlations
For the entire UARS period we have observations of CH_4 available from HALOE. Unfortunately, we do not have observations of N_2O, NO_y, ClO_y, or BrO_y. Therefore, where

no N_2O observations are available we can use inter-specie correlations to estimate N_2O (there will obviously be a larger uncertainty associated with these estimates than actual observations). More importantly, we can also use inter-specie correlations to estimate the total amount of reactive nitrogen, chlorine, and bromine (NO_y, ClO_y, and BrO_y).

Conservation of mass during assimilation
Data assimilation can easily cause a serious violation of conservation of mass, if total mass of reactive nitrogen, chlorine, and bromine, are not included as control variables. We have carefully addressed this conservation issue and it is therefore not likely to hinder the use of data assimilation for validation.

Observation groups violating conservation of mass
There is a conservation issue, which becomes apparent, when certain combinations of observation values are logically impossible. In this study this was illustrated in the case of version 5 MLS HNO_3, where the observation values erroneously suggest that there is more nitrogen in HNO_3 than the total amount of nitrogen available in the atmosphere.

Inconsistency between datasets: bias, error values
Inconsistencies between datasets due to bias, or unsuitable error quotation are also a problem. They are harder to deal with, and are most likely to be present in short lived species.

4 Approach to Validation used in Study

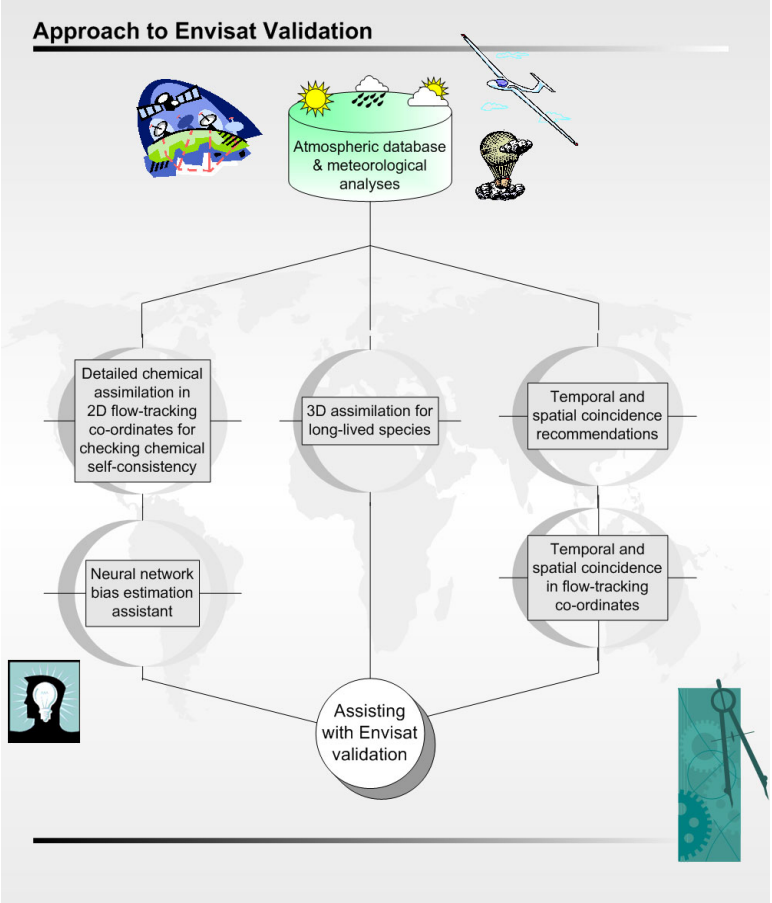

Figure 1: Approach to Validation used in Study.

The study investigated the use of data assimilation to assist in satellite validation of atmospheric constituent measurements. It contains several elements and suggestions which have not previously been used in satellite validation. Figure 1 shows the three basic strands that were used in the investigation:

Temporal and Spatial Coincidence
This first strand *(right)* investigates aspects of the conventional validation. Exactly what coincidence is defined, and the use of flow tracking co-ordinates is recommended. Probability distribution functions are shown to help detect biases. In addition, it is pointed out that if data assimilation is used the requirements for coincidence are more relaxed.

Assimilation of long-lived species in 3D CTM

This strand investigates the strategy of using 3D sub-optimal Kalman filter to assimilate species without a significant diurnal cycle into a 3D CTM. This technique requires very much reduced computing resources, in comparison to the "Detailed Chemical Assimilation", which formed the third strand.

Detailed chemical assimilation
This third strand is suited for both long and short lived species. It is very useful in checking chemical self-consistency between observations. It used a full Kalman filter cast in 2D flow-tracking co-ordinates and the comprehensive AutoChemchemical model. Using this as a major tool, investigations were made into an innovative technique using a neural network as a bias estimation tool.

5 Trace Gas Statistics

Introduction

In meteorological data assimilation a key step is data quality control. Quality control can only be reliably performed if we have good statistics available on the quantities we are measuring. This is already available within meteorology for NWP, but is not as yet available for trace gases. Therefore a major component of this work was to construct reliable observation statistics for as many trace gases as possible. This was done using a variety existing, validated, instruments (space borne, air borne, and ground based sensors).

Statistics in Flow Tracking Co-ordinates

A major component of the variability of trace gases is due to the atmospheric motions. It is therefore useful to construct our trace gas statistics in a co-ordinate system that 'moves' with the large scale flow pattern. We have done this by taking all the observations we have available to us by instrument and binning them in equivalent PV latitude (ϕ_e), potential temperature (θ). For each bin we then construct probability distribution functions (PDF) of the concentrations in each ϕ_e-θ bin as a function of month/season. In addition we calculate the mean, median, standard deviation, average deviation, skew and kurtosis of the PDF for each ϕ_e-θ bin as a function of month/season. All of this is then made accessible from an interactive web site so that the detailed information can be readily disseminated.

The value of such statistics can be immediately seen by examining Figure 2 which overlays the observed PDFs from different instruments observing the *same* region. Several other useful points become apparent from these statistics: are very useful for quality control: (a) The sonde histograms show rogue points at very low ozone concentrations for several of the bins, (b) the CLAES observations generally do not agree with the other instruments, (c) there is a small bias between MLS and SBUV2. These are just the types of issues that we need to identify and then correct within any quality control/validation system.

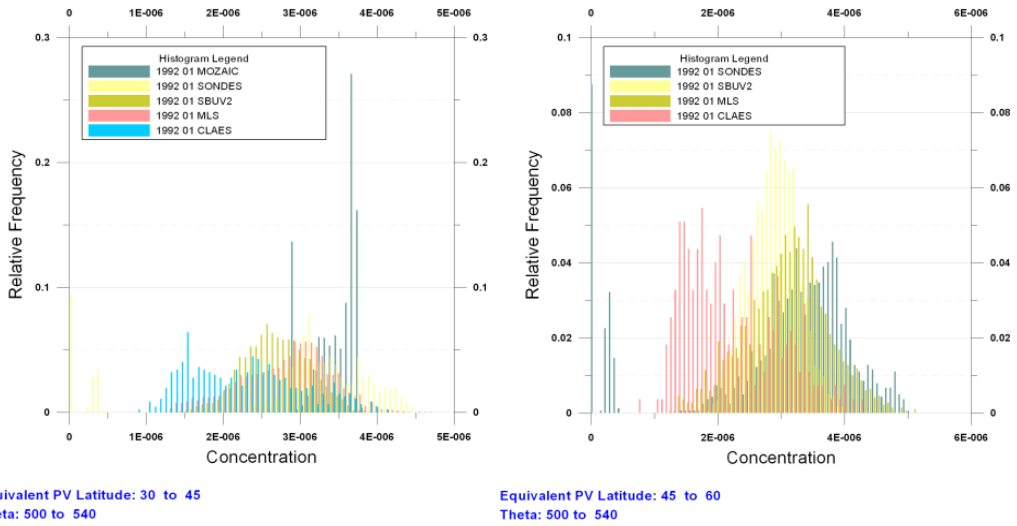

Figure 2: Ozone statistics from several sensors

A comparison of ozone statistics from several sensors in flow tracking ϕ_e-θ co-ordinates from MLS for January 1992. The region shown is between ϕ_e: $30^\circ \mapsto 60^\circ$ and θ:460 K $\mapsto$ 580 K. Each bin is 15° wide and 40 K high.

6 Diurnal Cycles & Chemical Self-Consistency

For more than two decades vertical profile measurements of trace gases have been made by solar occultation, using instruments such as SAGE, HALOE, POAM, ATMOS, and ILAS. Their observations are self-calibrating and are therefore useful for the analysis of temporal trends.

However, by definition the use of solar occultation limits the measurement opportunities to satellite sunrise and sunset. It is therefore useful to use chemical data assimilation cast in flow tracking co-ordinates to reconstruct full diurnal cycles and to check the chemical self-consistency of the solar occultation measurements.

This illustrates how the use of flow-tracking co-ordinates allows a more complete global view to be obtained from a rather sparse latitudinal coverage (in this case of solar occultation measurements).

For this part of the study, data from Atlas 1 was used to generate a vertical profiles of observations, in flow tracking co-ordinates at an equivalent PV latitude, Φ_e, of 38°S. This equivalent PV latitude was chosen as it was where ATMOS observed the atmosphere's composition over the largest range of altitudes. We assimilated ATMOS Atlas-1 observations of 14 species, namely: O_3, NO, NO_2, N_2O_5, HNO_3, HO_2NO_2, HCN, $ClONO_2$, HCl, H_2O, CO, CO_2, CH_4, and N_2O.

In this way chemical data assimilation was able to take solar occultation measurements of 14 species at sunrise and sunset and produce full diurnal cycles of analyses, which gave volume mixing ratios for 55 species, together with an associated uncertainty. The associated uncertainty values included a treatment of the observation instrumental error, representativeness, and also the photochemical theory uncertainty.

The results confirmed that there was good self-consistency between the ATMOS Atlas-1 observations of O_3, NO, NO_2, N_2O_5, HNO_3, HO_2NO_2, HCN, $ClONO_2$, HCl, H_2O, CO, CO_2, CH_4, and N_2O, and between the ATMOS observations and photochemical theory.

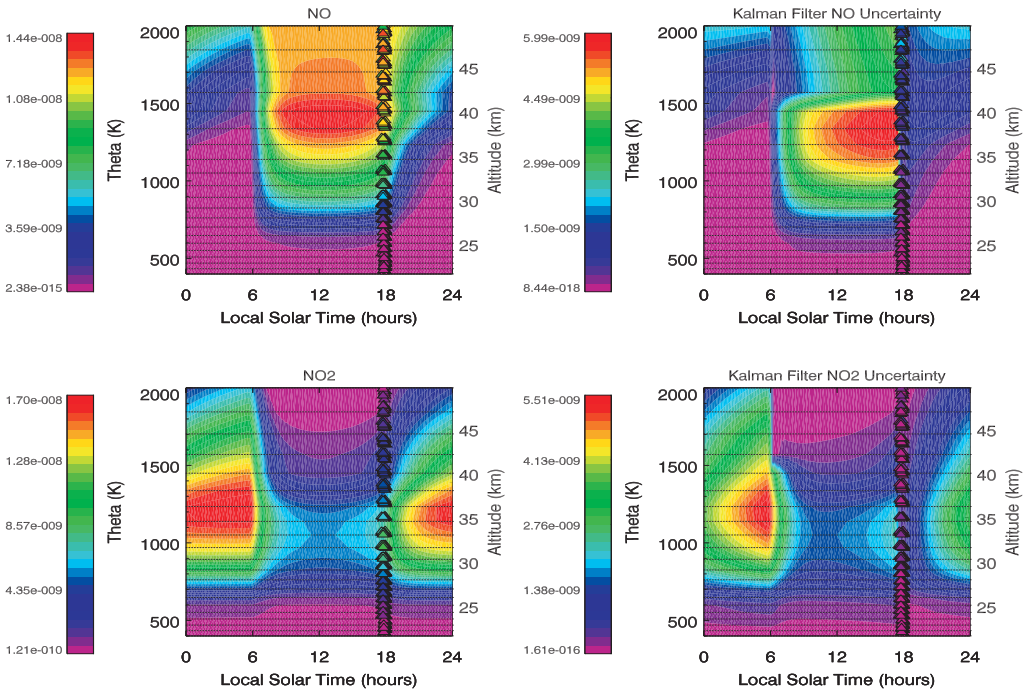

Figure 3: Diurnal cycles of NO, and NO_2

Diurnal cycles of NO, and NO_2 vertical profiles for the 30 March 1992 assimilation of ATMOS Atlas-1 data.
Left Plots: Analyses overlaid with ALL AVAILABLE observations (color filled triangles).
Right plots: Analyses uncertainties overlaid with corresponding observation uncertainties.

7 Assessing Coincidence

7.1 Introduction

Conventionally, validation is carried out by a series of measurements made by the instrument being validated, and corresponding coincident measurements made by another instrument of know quality. The problems of obtaining coincidence bring up the question as to "How coincident is coincident?" When considering flow coordinates, the answer to this question has two parts, one is spatial and the other temporal. The final report addresses both issues in detail.

7.2 Spatial Coincidence

Approach to assessment
To understand the spatial coincidence issue a specific example, that of the validation of the Tropospheric Emissions Spectrometer (TES) may be considered. As part of the validation of TES there is to be 'regular' sonde launches, typically at noon and on Wednesdays. It seems that it will be rare for a sonde launch to be even within 150 km of an overpass in the tropics. The question is: "Is this coincidence, coincident enough for validation?" The answer to this question is certainly situation dependent.

This question was addressed by using a high resolution data set from the CRISTA missions, which were carried aboard the Space Shuttle. This CRISTA datasets were considered particulary suitable since the prime science objective of CRISTA was to study small scale dynamical structures seen in the global trace gas distributions.

When we ask is 150 km of an overpass in the tropics sufficient, we are really asking, do we expect a large variability in the trace gas concentration over this scale?

We have chosen to answer this by transforming the CRISTA data to flow tracking coordinates. In the vertical there were 21 θ levels spaced equally in $\log(\theta)$ between 350 K and 2000 K, and in latitude there were 90 ϕ_e grid boxes from -90° to 90°. These were 2° wide, which corresponds to $\approx$ 222 km (the spatial scale under investigations).

The ozone measurements were each binned into the appropriate grid box. The largest median ozone concentration and largest percentage variability for each box were then determined. The percentage variability within each 220 km grid box served as an indicator as to whether or not 150 km could be regarded as satisfactorily coincident.

Results and discussion
Figure 4 (a) and (b) show the largest median ozone concentration for the two missions as a volume mixing ratio (v.m.r.). Figure 4. Figure 4 (e) and (f) show the largest percentage variability within a (ϕ_e, θ) grid box for the two missions.

Close to the tropopause there is a large vertical gradient in O_3. So even though the (ϕ_e, θ) grid boxes are just 30 K in this region there is still considerable variability in ozone over a grid box. In the region of strongest vertical gradient this can easily be as much as 45%. However, above this region, and away from polar regions the ozone variability over a (ϕ_e, θ) grid box is typically 10% or less. In the vortex edge (surf zone) region there is larger variability, typically around 20%. Where rapid ozone loss is occurring within the vortex the can also be large variability (bottom left of August 1997) plots.

So in answer to the question "Is 150 km of an overpass in the tropics sufficient?", we can say that above the region of sharp ozone gradient it almost certainly is since the variability expected is around the 10% level, or less. The region of strong vertical ozone gradients is more problematic, and the key factor for coincidence will be accurate height registration, rather than horizontal variability.

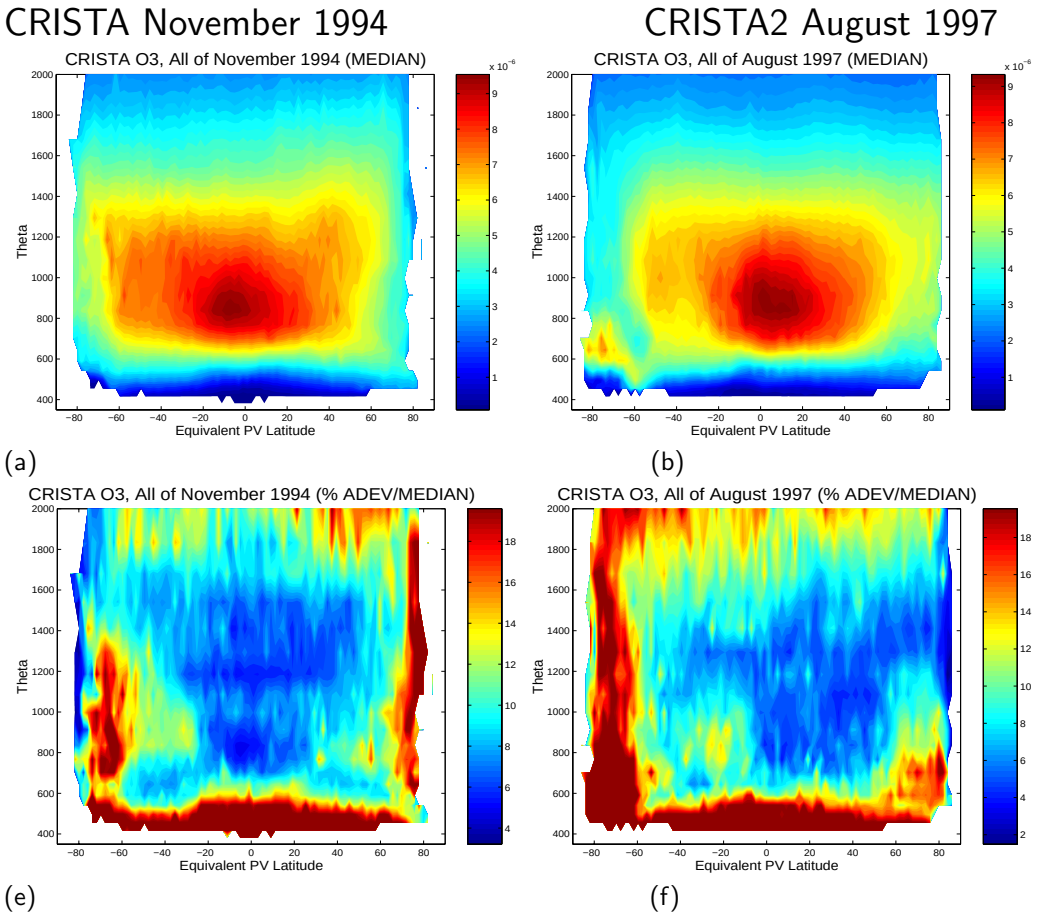

Figure 4: Ozone data from the CRISTA and CRISTA2 missions

Ozone data from the CRISTA and CRISTA2 missions during November 1994 and August 1997 transformed to ϕ_e, θ flow tracking co-ordinates. The left hand panel is for November 1994 and the right hand panel is for August 1997. (a) and (b) show the largest median ozone concentration for the mission as a volume mixing ratio (v.m.r.). (e) and (f) show the largest percentage variability within a (ϕ_e, θ) grid box for the mission.

7.3 Temporal Coincidence

The issue of temporal coincidence, which is also described Chapter 7 of the Final Report, was investigated in a similar manner, using data from ATMOS. ATMOS was another shuttle born instrument which simultaneously observed more than 14 species (many more than CRISTA).

Season and location will affect the answer to the question how coincident is coincident? To ensure that the answer is as precise as possible a specific example has been taken to describe a suitable methodology.

Due to space limitations here we just present a summary table. The Final report contains details of all species.

Species	**Ideal Validation Times** (in local solar time)	**Tolerance** (in local solar time)	**Unsuitable Validation Times** (in local solar time)	**Chances of Good Validation**
O_3	0:00-24:00	6 hours +	None	High
NO	9:00-15:00	within 1-2 hours (within 2° SZA)	2 hours either side of sunrise and sunset	Good away from terminator
NO_2	before 5:00 9:00-14:00	within 1-2 hours (within 2° SZA)	1 hr before sunrise, 3 hrs after sunrise, 3 hrs before sunset, 6 hrs after sunset	Good away from terminator
NO_3	9:00-15:00 0:00-4:00 < 25 km	within 1-2 hours within 1 hour (within 2° SZA)	During the dark except below 25 km	Poor without assimilation
N_2O_5	0:00-24:00	within 3 hours	None	High
HONO	0:00-5:00 7:00-24:00 > 27 km	within 3 hours	1 hour after sunrise 10:00-13:00 below 27 km	High
HNO_3	0:00-24:00	6 hours +	None	High
HO_2NO_2	0:00-24:00	6 hours +	None	High
$ClONO_2$	0:00-24:00	3 hours +	None	High
OClO	The 2 hours before sunrise	1 hour (within 2° SZA)	2 hours before sunset and 6 hours after sunset	Good
ClO	0:00-24:00	1 hour (within 2° SZA)	4 hours after sunrise in the lower most stratosphere	Good
Cl_2O_2	0:00-24:00	1 hour (within 2° SZA)	None	High
HCl	0:00-24:00	6 hours +	None	High
HOCl	0:00-24:00	1 hours	None	High
CH_3COCl	0:00-24:00	6 hours +	None	High
BrO	10:00-14:00	1 hour (within 2° SZA)	for a few hours after sunrise and before/after sunset, see notes in §??	Good
HOBr	3:00-6:00 close to local noon	1 hours (within 2° SZA)	several hours after sunrise and sunset	Good
BrCl	11:00-12:00	1 hour (within 2° SZA)	4 to 6 hours after sunrise 14:00-24:00	Good
HBr	0:00-24:00	4 hours	None	High
$BrONO_2$	5:00 16:00-17:00	1 hours (within 2° SZA)	8:00-10:00, 18:00-19:00 Please see notes in §??	Good
OH	13:00-15:00	1 hour (within 2° SZA)	5:00-11:00, 18:00-24:00 Please see notes in §??	Good
HO_2	14:00-17:00 12:00-24:00 > 27 km	1 hour (within 2° SZA)	Please see notes in §??	Good
HCHO	0:00-24:00	6 hours +	None	High
CO	0:00-24:00	6 hours +	None	High

Table 1: Temporal coincidence limits

Summary of coincidence in local solar time required for validation in the chosen example of 30 March 1992 for an equivalent PV latitude, ϕ_e, of 38°S. Where relevant a suggested tolerance in the solar zenith angle (SZA) is included.

8 Construction of Reference Atmosphere

Detailed chemical assimilation
A major part of the study was devoted to constructing a Reference Atmosphere to represent a typical day in mid January 1992.

The primary purpose of this was to tune the necessary assimilation techniques and provide a standard atmosphere having realistic distributions of all the important chemical species. This Reference Atmosphere provided a standard both for further work both within the study and for work in the future.

Assimilated Species
Many sensitivity tests of assimilation were performed using various combinations of data sets from the four chemical instruments aboard UARS. As a result of these experiments a number of problems concerning biases were uncovered. Also there were a number of inconsistencies within and between data sets, such as misleadingly small estimated error values. Overall, it was found that the HALOE data was normally of better quality.

The final selection of observational products, which were assimilated to produce the Reference Atmosphere, are shown in the table below.

Instrument	Dataset	Observations
HALOE	version 19	O_3 HCl CH_4, H_2O
CLAES	version 9	HNO_3 N_2O
ISAMS	version 10	CO

Use of tracer correlations
Although many important species could not be assimilated due to lack of reliable datasets, through the AutoChem chemical model inter-specie correlations were used to estimate a range of constitutents. There was of course be a larger uncertainty associated with these estimates than with direct observations.

Diagnostics for Assessment of Assimilation
Once the assimilation had been carried out and the analyses were performed, it was necessary to quantify the quality of the Reference Atmosphere. This is done by generating a range of diagnostic statistics. These diagnostics both assess the internal quality of the assimilation, and compare the observations used in making the analyses with the analyses themselves.

One part of these diagnostics comprised a range of plots of various parameters on PV Latitude and the Potential Temperature (Theta) axes: Analyses, bias, analyses uncertainty, uncertainty of representivity, and the residuals (O-F) and (A-F). Thus these plots give a view of the spatial distribution of variability of these parameters.

The other part of the diagnostics, which gave a view of the overall global of variability of the various parameters, comprised a series of scatter plots and distribution plots for each species. The various plots comprised: scatter plot of pseudo observations against analyses, quantile/quantile plots of observations against analyses, and distribution plots of biases, various uncertainties, and the residuals.

Summary
Table 2 summarises the findings from the scatter plots. It is seen that for all species (A-F) and (O-F) have a median value of less than 1%. These skill scores show that the analyses and observations are in good agreement. This is in part because several iterations were used to produce the Reference Atmosphere.

These objective skill scores show that the Reference Atmosphere for January 1992 is of good quality.

	Distribution Plot Percentiles		
Observations and Diagnostics	25 %	**50 %**	75 %
O_3 % Obs. Unc.	0.577	**0.963**	2.21
O_3 % Analyses Unc.	5.49	**6.28**	7.47
O_3 % Pseudo Obs. Unc.	0.662	**1.29**	10.8
O_3 % Pseudo Obs. Rep. Unc.	8.15	**10.1**	13.3
O_3 % (A-O) Analyses Bias	0.0116	**6.58x10^{-6}**	0.00303
O_3 % (A-F) Model Bias	0.189	**0.0231**	0.0113
O_3 % (O-F) Forecast Skill	0.203	**0.0271**	0.0115
HNO_3 % Obs. Unc.	5.19	**6.47**	11.5
HNO_3 % Analyses Unc.	8.87	**10.4**	12.2
HNO_3 % Pseudo Obs. Unc.	5.46	**7.11**	12.2
HNO_3 % Pseudo Obs. Rep. Unc.	18	**23.8**	31.5
HNO_3 % (A-O) Analyses Bias	0.274	**0.00636**	0.533
HNO_3 % (A-F) Model Bias	-1.42	**0.128**	0.452
HNO_3 % (O-F) Forecast Skill	-1.61	**0.143**	1.45
HCl % Obs. Unc.	4.18	**6.58**	11.8
HCl % Analyses Unc.	3.17	**4.47**	7.91
HCl % Pseudo Obs. Unc.	4.47	**6.71**	12.8
HCl % Pseudo Obs. Rep. Unc.	3.75	**6**	14.8
HCl % (A-O) Analyses Bias	0.385	**1.67x10^{-5}**	0.192
HCl % (A-F) Model Bias	0.471	**0.0502**	0.0705
HCl % (O-F) Forecast Skill	0.937	**0.00625**	0.291
CO % Obs. Unc.	10	**10**	10
CO % Analyses Unc.	8.33	**10.6**	14.3
CO % Pseudo Obs. Unc.	9.77	**10.2**	11.3
CO % Pseudo Obs. Rep. Unc.	16.2	**24.8**	43.4
CO % (A-O) Analyses Bias	0.0691	**0.0127**	-9.12x10^{-4}
CO % (A-F) Model Bias	0.608	**0.171**	0.0214
CO % (O-F) Forecast Skill	0.721	**0.192**	0.0286
CH4 % Obs. Unc.	0.234	**0.397**	0.694
CH4 % Analyses Unc.	4.35	**6.38**	7.96
CH4 % Pseudo Obs. Unc.	0.161	**0.329**	0.531
CH4 % Pseudo Obs. Rep. Unc.	5.77	**10.4**	15
CH4 % (A-O) Analyses Bias	3.43x10^{-6}	**2.67x10^{-5}**	2.77x10^{-4}
CH4 % (A-F) Model Bias	3.16x10^{-4}	**0.00137**	0.00684
CH4 % (O-F) Forecast Skill	3.28x10^{-4}	**0.00142**	0.00725
N2O % Obs. Unc.	10.1	**10.3**	11.1
N2O % Analyses Unc.	7.74	**9.92**	12.5
N2O % Pseudo Obs. Unc.	9.27	**10.2**	11.4
N2O % Pseudo Obs. Rep. Unc.	14.3	**22.1**	33.8
N2O % (A-O) Analyses Bias	2.73x10^{-5}	**2.28x10^{-4}**	0.00213
N2O % (A-F) Model Bias	7.86x10^{-4}	**0.00433**	0.0229
N2O % (O-F) Forecast Skill	8.49x10^{-4}	**0.00475**	0.0254
H2O % Obs. Unc.	2.55	**4.9**	9.73
H2O % Analyses Unc.	3.15	**4.33**	5.38
H2O % Pseudo Obs. Unc.	2.82	**5.67**	11.2
H2O % Pseudo Obs. Rep. Unc.	3.72	**5.73**	7.91
H2O % (A-O) Analyses Bias	-3.34x10^{-5}	**-4.93x10^{-6}**	3.76x10^{-7}
H2O % (A-F) Model Bias	0.00208	**-5.21x10^{-4}**	-2.47x10^{-6}
H2O % (O-F) Forecast Skill	0.0021	**-5.48x10^{-4}**	-2.22x10^{-6}

Table 2: Summary Diagnostics of Analyses and Observation Quality

Summary of diagnostics of analyses and observation quality for all the observed species. Showing the 25%, 50%, and 75% percentiles for the distribution plots of (i) analyses uncertainty, (ii) observation uncertainty, (iii) pseudo observation uncertainty, (iv) pseudo observation representativeness uncertainty, (v) bias, (vi) (O-F) and (vii) (A-F) distributions.

9 Simulated GOMOS and MIPAS Observations

Overview
Key to validation of instruments is the identification and measurement of any bias in the instrument's observations.

In order to understand the influence of biased observations on the assimilation tools and the Reference Atmosphere, and to develop suitable bias measurement tools, it was necessary to have a range of realistic simulated GOMOS and MIPAS observations. These observations could then be assimilated in a series of tests with a range of known biases.

Parameters used as basis of simulation
The simulated measurements for GOMOS and MIPAS were generated using a series of datasets giving the following input parameters:
a) The "Reference Atmosphere for January 1992"
b) GOMOS observation locations nominally for 12 January 2002
(with associated star class as supplied by ESTEC)
c) GOMOS measurement precision, or expected standard deviation d) MIPAS nominal-mode observation geometry and associated ESDs e) UKMO UARS assimilation meteorological analyses for Jan 1992

These parameters were used as a basis to provide realistic simulated observations with realistic estimated uncertainties. Figure 5 show an example set of locations of simulated observations.

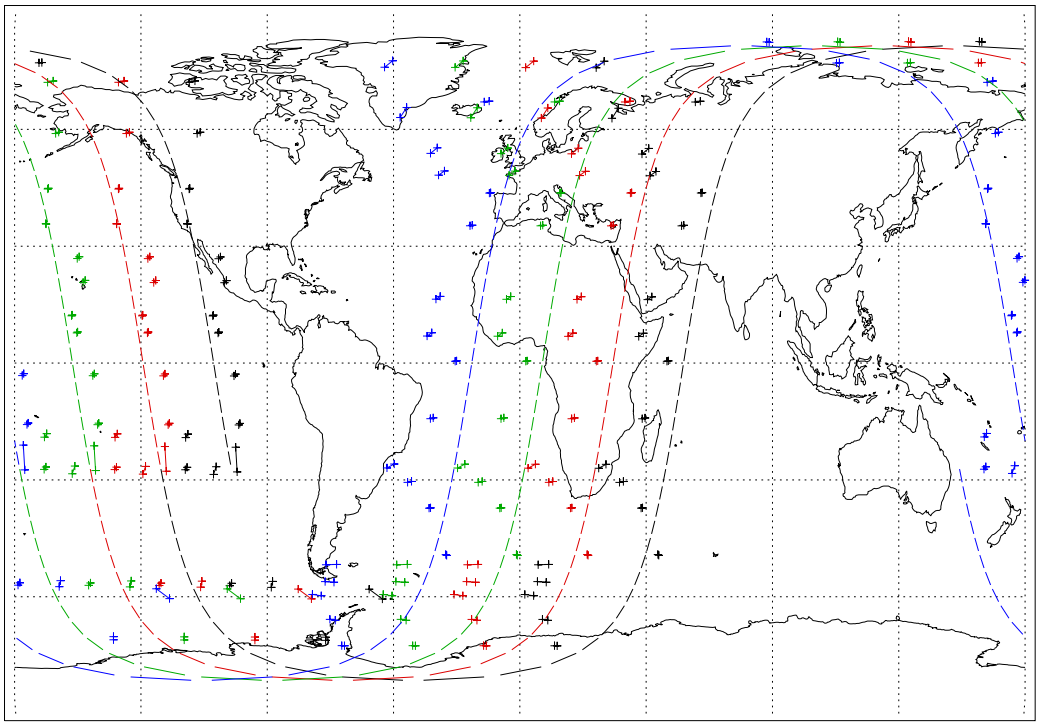

Figure 5: Measurement tangent point locations for GOMOS

Measurement tangent point locations for GOMOS (lines with "+" symbols) and MIPAS. Extent of each line indicates measurement extent in horizontal from top to bottom of limb scan. First four simulated orbits are plotted (ascending node crossings 05:16:22 to 10:18:10 for 12 January).

10 Bias Estimation Tool: the NN Assistant

Requirement for an innovative tool

The two most critical data quality issues are the correct specification of uncertainties and the presence of biases. Of the two, detecting biases is probably the most difficult. We have already seen in Section 5 that using concentration PDFs of several instruments can help in this. However, assimilation can probably help as well.

Biased observations will cause a set of multi-specie observations to be inconsistent. We have already seen in Section 6 that Assimilation can be used to examine the self-consistency of a dataset. The measures we use to quantify the quality/skill of the assimilation analyses are:

Observation Increment	(O-F)	(Measure of forecast skill)
Analysis Increment	(A-F)	(Measure of model bias)
Analysis-Observations	(A-O)	(Measure of analyses bias)

So, if we conducted a set of experiments where we added *known* biases (that span all the likely range of possible biases) to synthetic GOMOS and MIPAS observations and calculated these skill scores we would know how our assimilation system responds to bias. Inverting this gives a bias detection assistant/tool.

It has been clear for a long time that human brain analyses data and information quite differently from parametric statistical methods, and that it processes the flood of data and learns from them along quite different lines. Neural networks (NN) started as an attempt to build mathematical models that worked in the same way that brains do. In a nutshell, the NN is a nonparametric statistical model for extracting nonlinear relations in the data. In NN, instead of matching the architecture of the model to a problem, a model is used that can describe almost anything, and careful training of the model is used to constrain it to describe the data.

As a demonstration of the possibilities of this technique, a simple tool known as the "Bias Estimation Neural Network Assistant" was developed. In the final report we describe the neural network structure and the training algorithm that this toll employed for predicting biases in our analyses. The results obtained from this demonstrator were very encouraging.

The future

The application of the Bias Estimation Neural Network Assistant to ENVISAT is clearly a matter for a future study. The results so far are encouraging, and it is likely to give some help at estimating biases. With neural networks a key issue is always training, has the training data set adequately spanned the system?

The field of neural networks has a history of some five decades but has found solid application only in the past fifteen years, and the field is still developing rapidly. Thus, it is distinctly different from the fields of control systems or optimization where the terminology, basic mathematics, and design procedures have been firmly established and applied for many years. We do not view the Neural Network assistant as an application of established procedures. Rather, we hope that it will be a useful tool for validation. Time is required to find what works and what doesn't.

11 Data Assimilation using the 3D CTM

Introduction
The CTM was run with a horizontal resolution of 7.5° × 7.5° (T15 Gaussian grid) and 18 isentropic levels from 330 K to 3000 K (approximately 10 to 55 km). A set of 24-hourly United Kingdom Meteorological Office (UKMO) analyses were used to force the model.

A basic model run with no assimilation was initialised in October 1991 from a 2-D model and then integrated until December 31, 1991. This output was then used to initialise a series of four assimilation runs, which are summarised below:

CON A control run without assimilation.

HAL As **CON** but including assimilation of HALOE O_3, CH_4, H_2O, and HCl.

HALC As **HAL** but including optimised constraints on non-assimilated species

HALX As **HAL** but including over-strict constraints on non-assimilated species

Initial problems
Our first attempts to assimilate HALOE HCl directly into the 3D CTM resulted in the modelled total inorganic chlorine (Cl_y) rapidly becoming unrealistic. The modelled values of Cl_y exceeded 4 ppbv, when we know from atmospheric chlorofluorocarbon (CFC) abundances that there is a limit of Cl_y of about 3.6 ppbv for 1990's conditions. Indeed, it is relatively straightforward for an atmospheric model to predict Cl_y based on known CFC abundances. In our 3D CTM (which is used for long trend simulations, for example) the incorrect Cl_y would be very undesirable. Therefore, we need to impose a limit on the model Cl_y (and other inorganic chlorine species) when we assimilate HCl (see below for details).

Another relationship between certain atmospheric species concerns the correlations that are observed between pairs of long-lived tracers. Measurements of one long-lived tracer can be used to derive other long-lived tracers.

Specification of constraints
Within the philosophy of data assimilation into a 3D CTM in this Chapter (i.e. to constrain multiannual simulations of an established 3D model), it is desirable both to assimilate observations and to maintain the correlations with non-assimilated long-lived tracers. One method would be to use a fit to the CH_4 v N_2O correlation based on atmospheric observations to infer a pseudo-observed N_2O. This would, in effect, be mixing the HALOE data with other (e.g. ER-2) observations. An alternative approach would be to use the model-predicted correlations to derive a new N_2O value for each assimilated CH_4 point. We have chosen this latter approach so that tracer correlations remain determined by the model chemistry (i.e. so that other observations can still be used to test the model's N_2O distribution).

12 Recommendations for Validation Campaigns

R1: Choose Optimal Regions for Initial Validation
Although all regions need to be validated, the tropics are probably the most suitable region for initial validation. This is because, for many gases, there are large spatial variations in concentration in the polar regions, and in the mid-latitude upper troposphere there are tropopause folds and extensive laminations.

R2: Select Observation Latitude and Local Solar Time to maximise Coincidence
From a temporal (local solar time) point of view, how coincident is coincident in both time and space is a key question, see Section 7.3.

Table 1 summarises the recommendations for temporal coincidence for validation campaigns. It is highly desirable to use the solar zenith angle *as well as* local solar time when defining coincidence.

R3: Use Flow-tracking Coordinates to maximise Coincidence
As well as the traditional approach to validation using spatial coincidence it is desirable to use flow tracking co-ordinates (equivalent PV latitude (ϕ_e), potential temperature (θ)).

This is especially true when dealing with the often limited global coverage of solar occultation instruments, since flow tracking coordinates will lead to many more possible 'coincidences' for validation. In the case of short lived species such as NO, NO_2, NO_3, ClO, OClO, BrO, OH, and HO_2, it is highly recommended to also use solar zenith angle as well as local solar time.

R4: Assess Vertical Resolution of Observations
An assessment should be made of the actual (as opposed to the theoretical) vertical resolution of GOMOS and MIPAS, i.e. the vertical scale on which ENVISAT can resolve concentration profiles. Is the vertical resolution of one species measurement transferable to other species?

R5: Assess Altitude Registration of Observations
An assessment should be made of the altitude registration of GOMOS and MIPAS.

R6: Assess realism of Observed Diurnal Cycles
Are the diurnal cycles observed by GOMOS and MIPAS realistic? For example, we know that some some species are so long lived that they do not have a diurnal cycle, is this what is observed by GOMOS and MIPAS? This could highlight possible difficulties with the retrieval algorithms.

R7: Take advantage Photochemical Theory to validate NO_3
NO_3 would benefit from the use of photochemical theory via steady state assumptions or assimilation, as described in detail in the Final Report.

R8: Take advantage of Multivariate Modelling
Data Assimilation is a very valuable tool for instrument validation since it is a multi-variate analysis technique which can be used to analyse multi-variate observations *and* include our theoretical knowledge of the system. It can be used to great effect as full diurnal cycles can be reconstructed from even solar occultation measurements.

R9: Long-term Retrospective Validation
Long-term retrospective validation is of considerable value. Not all validation (such as an evaluation of drifts) can be done in the first phase immediately after the launch. An ENVISAT re-analysis programme should be considered when a significant data time series has been collected, this should be done with high quality measurements such as are available from the Network for the Detection of Stratospheric Change (NDSC). The value for such a programme can be seen in the case of UARS where periodic enhancements are made to the data quality. If such a reanalysis is carried out too early their will be insufficient data, probably at least a year is required.

13 Conclusions

13.1 Choice of Data Assimilation Schemes

This study used a range of chemical assimilation schemes in different models, namely:

1. 4D-Var
2. Full Kalman filter
3. 3D Sub-optimal Kalman filter in a CTM

The first two were used in a chemical box model (in flow tracking coordinates) and the third was used in a full chemistry CTM.

The main advantage of 4D-Var is that no time discontinuities are present in the analyses. The analyses is produced by starting from an a priori of the chemical concentrations (called the background) which is then improved by the application of 4D-Var. This then gives a new updated set of initial conditions which is used to produce a new forecast, known as the analysis. However, no time evolving estimate is provided of the uncertainty associated with the analyses.

The main advantage of the Kalman filter is that the full species error co-variance matrix is time integrated. This provides a full time evolving estimate of the uncertainty associated with the analyses. However, in the way the Kalman filter is usually used this can also give rise to time discontinuities when observations are processed and the analysis state adjusted accordingly.

So we have modified the use of the Kalman filter. As with the 4D-Var case we start from an a priori of the chemical concentrations (called the background) which is then improved by the iterative application of the Kalman filter (usually two iterations). This then gives a new updated set of initial conditions which is used to produce a new forecast, known as the analysis. This then gives an analysis with no time discontinuities and the full time evolving estimate of the uncertainty associated with the analysis.

Due to the cost of both the 4D-Var scheme and Full Kalman filter, they can only be used in a full chemistry 3D model for very short simulations (possibly a few weeks). For the longer simulations that many 3D models are used for the only practical option of those considered here is the sub-optimal Kalman filter. As this is a sequential assimilation scheme (i.e. the model runs forward in time once only) there are limitations on the species which can be usefully assimilated. Essentially one could say that the photochemical lifetime of the species need to be sufficiently long that the 'memory' of the assimilation persists until the next observation time. Even with the sparse coverage of HALOE, the long-lived species like CH_4 and H_2O satisfy this requirement. In contrast, sparse observations of a short-lived species like NO_2 would not be useful in such a 3D model. Consequently we used 4D-Var and the full Kalman filter in 2D flow-tracking co-ordinates.

13.2 Quality of Analyses

The quality of the analyses produced by data assimilation depends on: The quality of the observations, the quality of the observation uncertainties (this have often found to be in error), the quality of the photochemical model, the quality of the meteorological analyses, the quality of the assimilation algorithm.

In the ATMOS case study we have shown that chemical data assimilation has been able to take solar occultation measurements of 14 species at sunrise and sunset from ATMOS and produce full diurnal cycles for 55 species together with an associated uncertainty. The associated uncertainty includes a treatment of the observation, representativeness, and photochemical theory uncertainty. As one would hope from such a quality instrument as ATMOS we confirm that there is good self-consistency between the ATMOS Atlas-1

observations of O_3, NO, NO_2, N_2O_5, HNO_3, HO_2NO_2, HCN, $ClONO_2$, HCl, H_2O, CO, CO_2, CH_4, and N_2O, and between ATMOS and photochemical theory. If there had been inconsistency the assimilation would have highlighted these and quantified the magnitude.

The assimilation of the ATMOS data set without any problem demonstrates the validity of the chemical model, the assimilation machinery, and that the quality of the meteorological analyses are not a primary issue.

A high quality data set, such as ATMOS, combined with data assimilation gives excellent results. Giving both full diurnal cycles from partially observed diurnally cycles, and good quality estimates together with an associated uncertainty of unobserved species. Assimilation can clearly demonstrate the presence or absence of data self-consistency.

The fact that the same can ***not*** be said for assimilating data from UARS shows that data quality and the reliability of uncertainty estimates is most certainly a major issue. For example three different instruments were observing NO_2 during January 1992, HALOE, CLAES, and ISAMS, and they each give quite different NO_2 distributions. MLS version 5 HNO_3 in many locations apparently reports more nitrogen in HNO_3 than is available in the atmosphere! Self consistency and bias are clearly big issues for UARS, and may well be for ENVISAT and EOS AURA. That is why part of this project has been a lateral thinking attempt at detecting biases using neural networks.

A key 'zeroth order' issue is clearly conservation of mass. The MLS version 5 HNO_3 example just mentioned graphically illustrates this. If this important issue is not given careful attention the analyses produced could be virtually worthless. From some of the chemical assimilation studies I have seen this is a very important point to make, which can not be made strongly enough.

A key part of the data inconsistency issue is specification of uncertainties. The assimilation takes in *two* pieces of information from each observation, the actual concentration and its associated uncertainty. The uncertainty is used by the assimilation as the 'weight' (if you like the credence) that should be given to this observation. If the specified uncertainty does not reflect the actual uncertainty serious problems can result. For example, if an observation has in reality a very large uncertainty, yet a small uncertainty has been specified the assimilation will try and move the chemical state of the atmosphere to agree with this spurious observation, sometimes with catastrophic results. This was most obvious for HALOE NO and NO_2, typically at sunrise. At some locations the NO or NO_2 observations, with apparently unsuitable uncertainties, would be assimilated and lead to a serious negative impact on the quality of the HCl. To make matters worse, this does not happen uniformly everywhere. It would typically be observed at 10-15% of grid points, noticeably at the summer pole.

It may be that diurnal corrections as were applied to ATMOS are required. Whatever the reason, incorrectly specified uncertainties are bad news. One way to partly deal with this in future would be to make bias a control variable, as it appears that in several cases the observations with unsuitable uncertainties are also biased.

13.3 Biases

The issue of biases is a non-trivial task. As just mentioned, in future studies this could be dealt with by making bias a control variable. In this study we have chosen a different approach, to use machine learning neural networks to 'learn' how our assimilation system responds when biases of known magnitudes are present. We used an extensive database of observations to make sure that the likely range (and even unlikely range) of biases for each species were covered. Once the neural networks have been trained they can then be used to estimate the likely magnitude of an unknown bias which may be present.

The application of the Bias Estimation Neural Network Assistant to ENVISAT is clearly a matter for a future study. The results so far are encouraging, and it is likely to give some help at estimating biases. With neural networks a key issue is always training, has the training data set adequately spanned the system?

The field of neural networks has a history of some five decades but has found solid application only in the past fifteen years, and the field is still developing rapidly. Thus, it is distinctly different from the fields of control systems or optimization where the terminology, basic mathematics, and design procedures have been firmly established and applied for many years. We do not view the Neural Network assistant as an application of established procedures. Rather, we hope that it will be a useful tool for validation. Time is required to find what works and what doesn't.

13.4 Pure Data Assimilation or with constraints?

A rather philosophical question in the subject of data assimilation is whether the pure observations should be assimilated whatever they may be, so that the assimilation scheme and the formal errors of the observations and model are the only factors which determine the analysed field, or whether other constraints should be applied to prevent assimilated fields violating some basic atmospheric laws. As discussed in Chapter 12 in developing the 3-D CTM assimilation tool in this project we decided to use the observations of a single long-lived tracer (e.g. HALOE CH_4) to constrain essentially all of the other long-lived tracers in the 3-D model. There are many questions about whether to apply such a correction and the answers probably depend on the reason for data assimilation and the desired end product.

First, it is worth reiterating that in the real atmosphere (i.e. stratosphere for our purposes) it is now well known that long-lived chemical tracers (i.e. species whose chemical lifetime is longer than the timescale for transport/mixing) reveal compact correlations. This has nothing to do with them being coupled chemically - e.g. CH_4 and N_2O show this compact correlation. Beyond this there are also more constrained relationships between chemical families (independent of chemical lifetimes of individual members). An obvious example here is that total chlorine in an air parcel is conserved and so as organic Cl (e.g CFCs) is destroyed inorganic Cl (ClO_y=HCl + ClO + etc) is produced. Generally, atmospheric models do a good job of predicting the compact correlations between e.g. N_2O and CH_4 and all models (that conserve mass!) will maintain the constraint between CFCs and ClO_y.

If observations of a limited number of species were assimilated into a 3D model (e.g. HALOE CH_4 and HCl) without any such constraint the modelled fields would rapidly lose self-consistency. First, the model would fail to maintain the compact correlations and second the model would start to create or destroy chlorine. With no limit on the model Cl, unrealistic values would result which would feed back onto O_3 and therefore any other species. While the model HCl and CH_4 might be realistic in the vicinity of observations, the other model fields (e.g. ClO, $ClONO_2$, O_3 etc) could be nonsense. This would occur because slight errors in the model chemistry would mean that assimilating HCl would act as a source or sink of chlorine. It is not clear how long the model would take to become unrealistic (depends on the model /data differences) but it would likely be rapid (a few days - weeks). For 3D models which need to simulate the atmosphere over decadel timescales this is clearly not useful. For some short period studies (e.g. a few days) it may be possible to run a model without constraints.

It is also worth noting that the 4D-Var and Kalman filter technique explicitly provides the full 'coupling' between species that the sequential scheme used in the 3-D model does not. This coupling allows us to check the chemical self-consistency of the observations. In addition, it allows the inference of unobserved species together with an associated uncertainty from the subset of observed species.

13.5 Assimilation of Chemical Species in Dynamical Models

In all the models used in this study the atmospheric circulation was fixed and taken from meteorological analyses (e.g. ECMWF, UKMO). These wind and temperature products are themselves the product of assimilation of meteorological variables into a dynamical model. Here the focus has been on chemical data assimilation as a tool for improving

our understanding of atmospheric chemistry. However, differences between observed and modelled long-lived chemical species could also be the result of poor transport. This is quite likely a cause for the discrepancy of the 3D CTM CH_4 field in Figure 11.8 of the final report.

A very useful extension of chemical data assimilation (which many groups are now working on) is the assimilation of long-lived chemical tracers into the dynamical models which are used to produce these meteorological analyses. Continuity of a chemical tracer would impose a constraint on the modelled winds. This may be particularly useful in the context of Figure 11.8 as the tropics is a region where the coupling between winds and temperature gradients through the thermal wind balance is weaker and so, for example, assimilated temperatures are less of a constraint on the winds. In this region assimilation of good chemical tracers may be very useful.

Finally, it is worth noting that if a long-lived tracer was assimilated in a full chemistry dynamical model to modify the circulation, all of the modelled tracers would experience this new, correct circulation. Their distributions would improve and remain self-consistent; there would be no need for the tracer-tracer correlation modifications which we imposed in the off-line CTM simulations in Chapter 11 of the final report.

13.6 How Coincident is Coincident?

With the recent launch of Envisat and the upcoming launch of EOS-AURA the issue of satellite validation is of current importance. During the validation of satellite instruments observing atmospheric composition one question is always faced, how coincident is coincident? Both the spatial and temporal part of this question have been addressed and quantified.

As well as the traditional approach to validation using spatial coincidence (i.e. latitude, longitude, pressure) it is desirable to use flow tracking co-ordinates (equivalent PV latitude (ϕ_e), potential temperature (θ)). This is especially true when dealing with the often limited global coverage of solar occultation instruments. The use of flow-tracking co-ordinates allows a more complete global view to be obtained from a given latitudinal coverage. This will lead to many more possible 'coincidences' for validation.

13.7 Summary

We have shown that chemical data assimilation can demonstrate the presence or absence of a data sets self-consistency. As far as assimilation is concerned the main problems faced with data are incorrect specification of uncertainties and the presence of bias. We have constructed a tool which uses neural networks to 'learn' the response of our assimilation system to biases.

The issue of coincidence is always important during validation. We have quantified spatial and temporal coincidence required for good validation and shown that using assimilation in flow tracking coordinates greatly assists in obtaining more 'coincidences' for validation.