QUANTITATIVE TROPOSPHERE COMPOSITION RETRIEVALS FROM SATELLITE MEASUREMENTS: INITIAL RESULTS

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ABSTRACT

The direct measurement of tropospheric trace gases from space is difficult and so it is necessary to combine models and data to fully exploit the potential of observations. To obtain quantitative estimates of the tropospheric abundance the contamination from clouds and the stratosphere have to be removed accurately, and the multiple-scattering of photons in the troposphere has to be correctly interpreted. The aim of this project is to introduce a new method for calculating tropospheric column amounts of trace gases that reduces the large error that is already associated with this kind of calculation. We will initially focus on estimating the concentration of NO₂, BrO, SO₂ and HCHO using data from the Global Ozone Monitoring Experiment (GOME). There are two steps to this process.

The first step is to remove the stratospheric component of the column. Most studies approximate this component by either assuming a profile from model output or assuming that over remote areas of the world there is no pollution so that the gas will be present only in the stratosphere (for example NO₂). We use an existing chemical data assimilation scheme within the SLIMCAT/TOMCAT three dimensional (3D) chemistry transport model (CTM) to produce an accurate simulation of the stratosphere, which can then be removed from the measurements.

The second step is to transfer the measured slant column densities to vertical column densities. Here we will use a multiple scattering radiative transfer model (GOMETRAN) along with information about the aerosol, cloud cover and surface reflectance from the Along Track Scanning Radiometer 2 (ASTR-2) to simulate the tropospheric vertical column amount.

1 STAGE I: CALCULATING STRATOSPHERIC COLUMNS

SLIMCAT/TOMCAT is an off-line chemistry transport model [1]. It contains a detailed stratospheric chemistry scheme including gas-phase and heterogeneous chemistry. It uses horizontal winds from the ECMWF ERA-40 reanalysis and vertical motion from diagnosed heating rates. Although the run used here contains no explicit tropospheric chemistry there is still interaction between the two atmospheric regions to enable more realistic results.

The SLIMCAT/TOMCAT model was used to perform multiannual runs with and without data assimilation from 1992 onwards. The assimilation scheme uses the sequential sub-optimal Kalman filter scheme [3]. In SLIMCAT we assimilate four species; O₃, H₂O, HCl and CH₄ measured by the Halogen Occultation Experiment (HALOE) [2].
Figure 1: Shows the zonal mean distributions from SLIMCAT on the 30th of January 1992 with and without assimilation. These gases are both long lived tracers, CH$_4$ which is directly assimilated into the model and N$_2$O which is updated in the model via its preserved correlation with CH$_4$. We see, in figure 1, an increase in gradients in the subtropics. This is a documented phenomenon and is due to the errors in the tropical dynamics in the meteorological assimilation models that are used to generate the winds that are then used to force the model [4]. As the two gases are correlated we see similar changes in both cases.

The validation of the data assimilation scheme has been carried out for non diurnal varying species.

Figure 2: Comparison of ATMOS 1 profiles for N$_2$O and CH$_4$, at 47° S and 115°E, for 23rd March 1992
Figure 2 shows the validation of CH$_4$ and N$_2$O for the 23rd of March 1992 with and without assimilation using data from the ATMOS 1 mission. The figure shows that the assimilation scheme improves upon the non assimilation scheme (green). NO$_2$ is directly related to N$_2$O through its relation to NOy. This means that improvements to the model N$_2$O will directly impact and improve the modelled stratospheric NO$_2$.

In order to use these calculated stratospheric amounts we need to sample SLIMCAT at the correct local time of the GOME overpass (passing over the equator at 10.30). It is important to do this due to the strongly diurnal nature of the species we will be studying. Figure 3 shows the SLIMCAT vertical column of NO$_2$ when sampled like GOME. This is a purely stratospheric column.

![SLIMCAT total column](image)

**Figure 3:** The total column of NO$_2$ (cm$^2$) calculated from SLIMCAT (sampled like GOME) for the 1st of July 1997

Figure 3 also shows that NO$_2$ varies with longitude, i.e. not zonally symmetric. This would not be easy to simulate in an assumed stratospheric distribution and therefore suggests that this type of modelling greatly improves the stratospheric calculation.

2 STAGE II: SIMULATING THE TROPOSPHERE

In order to correctly simulate the radiation in the troposphere a multiple scattering radiative transfer model is used (GOMETRAN). GOMETRAN was designed for retrievals from GOME and calculates the simulated spectrum of radiance as would be seen by the satellite and the weighting functions, which are derivatives of the radiance with respect to the parameters being retrieved.

2.1 Sensitivity of the Radiative Transfer Scheme

To permit quantitative measurements we must understand how the results of the radiative transfer equation vary with small changes in the input values. This will allow us to quantify the errors created within GOMETRAN with respect to errors in the input parameters. For example, if the surface albedo has an error of 5%, what size error will that relate to in the calculated column.

This was investigated by creating 12 geotemporal scenarios. These are at four different locations at four different times of the year.
The 12 scenarios have different trace gas, temperature and pressure profiles along with different view angles. GOMETRAN was run for each scenario with changes in the other parameters:

- Surface Albedo
- Number of Atmospheric Layers
- Number of Streams
- Number of Legendre Moments (Phase Function)
- Aerosol Type and Profile Shape
- Cloud Fraction, Height, Albedo, Type and Optical Depth
- Trace Gas, Temperature and Pressure Profiles

Results of these tests so far carried out and are presented below.

### 2.1.1 Surface Albedo Results

Surface albedo varies with wavelength. In this study there are two ways we can derive the albedo. Firstly we can use data from the ATSR-2 instrument, however this is only available in one wavelength range. The error on the albedo here will be relatively high. The second way to calculate the albedo is by retrieving the albedo directly from GOME. In this case it will be in the same wavelength ranges as the slant columns being retrieved. This method will have a relatively low error. Figure 4 shows a plot of surface albedo versus error in the tropospheric column amount when comparing a range of surface albedo's with one of 0.01.

![Figure 4: The sensitivity of GOMETRAN to changes in the surface albedo compared to a surface albedo of 0.01 (scenario July 5° N)](image)

Figure 4 shows that for low surface albedo, when most radiation is absorbed, the error in the column grows large with a small change in the albedo. This is the opposite for high surface albedo. For example an error of ±0.1 at 0.3 gives an error of ±5%, whereas an error in the surface albedo of ±0.1 at 0.01 gives an error of around 60%. This could potentially cause problems in areas where there are large surface albedo changes within small areas. For example at the poles where there is ice (albedo ~ 0.9) and sea (albedo ~ 0.02).
2.1.2 Further Results

Tests have been run on other parameters and are summarised here:

- Number of Layers. The model needs around 610 layers, over 60km, with a spacing of at least 50 - 100m in the first 20km then around 1km for the next 40km.
- Number of Streams and Legendre Moments. NO₂ needs at least 25 streams and therefore 55 Legendre moments. Other gases need 20 streams and therefore 45 Legendre moments.
- Further tests on GOMETRAN are in progress and show that cases with high aerosol asymmetry cause problems in resolving the phase function. A new function has been placed in the model to simplify the solution (delta M scaling [5]). This removes some of the forward scattering and replaces it with unscattered radiation.

2.2 Running the full Radiative Transfer Scheme

In order to improve the radiative transfer calculation, for every GOME pixel, we use data from ATSR-2 on aerosol and cloud. The ATSR-2 data has been mapped onto the GOME scene as part of a NERC/RAL funded project to produce a data set called GRAPE. With the inclusion of the results from section 2.1 the running of the full code will result in fully quantitative retrievals.

3 CONCLUSIONS

This project aims to introduce a new quantitative scheme for retrieving tropospheric trace gases from satellite instruments such as GOME. It incorporates modelling results and data from a number of different sources to allow realistic information to be entered into the process.

When calculating the stratospheric column the SLIMCAT 3D CTM with a data assimilation scheme is used to improve upon the model results. The validation of this with ATMOS mission data shows that the model predicts values to within a reasonable amount of error which can be easily quantified.

The radiative transfer model, GOMETRAN, has been tested to find that the surface albedo needs to be retrieved alongside slant column data from GOME. This will produce the smallest error. Also, the testing shows that the model does not perform well at high asymmetry and a new function, delta-M scaling, has been introduced into the model.

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6 REFERENCES


