

REFERENCE MODEL FOR CH<sub>4</sub> AND N<sub>2</sub>O AND TRENDS

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## ABSTRACT

Data from the Stratospheric and Mesospheric Sounder on Nimbus 7 have been used as the basis for a model of the abundances of nitrous oxide and methane in the stratosphere. A version of this was produced two years ago (Taylor, Dudhia and Rodgers, /1/ - hereafter called paper 1) and in this new paper we consider some of the possible error sources in more detail, as well as long-term trends. The principal source of error in the SAMS retrievals is thought to be the use of climatological ozone profiles to invert the temperature profile data. However, we find that the effect is too small, and of the opposite sign, to explain the discrepancies between satellite and in-situ measurements, noted in paper 1. As expected, no systematic trends which exceed the estimated error in the data are found in either methane or nitrous oxide.

## INTRODUCTION

Nitrous oxide and methane are two of the important minor constituents of the atmosphere. The former is the principal source of stratospheric NO<sub>x</sub> which plays a significant role in the photochemistry of ozone, while methane is an important 'greenhouse' gas and the only in-situ source (through its photo-oxidation) of stratospheric water vapour. Accordingly, data on the mean abundance of these species is an important input to models and other studies of the middle atmosphere, currently a region of much research interest.

The only comprehensive data set on these two gases, covering most latitudes and all seasons over a period of several years, is that obtained by the Nimbus 7 Stratospheric and Mesospheric Sounder (SAMS- see Drummond et al. /2/ for a description of the instrument and Taylor /3/ for a discussion and overview of the results obtained). These data were used in paper 1 to construct a three-year average (1979 to 1981 inclusive) from which tables of mean monthly abundance versus log(pressure) and in ten degree latitude bins were constructed. Seventeen pressure levels from 20 mb. (about 25 km) to 0.1 mb (about 65 km) and thirteen latitude bins (from 50° S to 70° N) were presented.

The purpose of the present paper is primarily to re-evaluate the model in the light of work that has been done in the meantime to further validate the SAMS data and to investigate certain discrepancies with balloon data which have been uncovered. We also examine the data set for signs of trends in the abundances of both species and present tables of results for these.

## EFFECTS OF OZONE ON SAMS DATA

## (A) Sensitivity of temperature retrievals to ozone

In paper 1 we showed evidence for discrepancies between SAMS data and in-situ measurements from balloons, particularly below the 10 mb pressure level. In investigating this, we decided that, if the discrepancy was due to a systematic error in SAMS, the most likely cause was the use of climatological ozone profiles in the retrievals of temperature from SAMS 15µm carbon dioxide emission observations. A correction has to be applied to the transmission function in the temperature sounding channels because of

the overlapping opacity of the 16  $\mu\text{m}$  ozone band. The constituent abundances which are retrieved depend strongly on the temperatures, since the emission which is measured is of course a function of both. Jones and Pyle /4/ quote variations in retrieved mixing ratio of as much as 50% for both gases with a  $\pm 2\text{K}$  temperature variation at 20 mb, although this sensitivity decreases rapidly with pressure and is around 10% at higher altitudes.

In the present work, we have made use of new data on the ozone distribution derived from a combination of SBUV and LIMS measurements which provides profiles for a particular month and latitude. November 1979 was chosen for these tests since balloon data for  $\text{CH}_4$  and  $\text{N}_2\text{O}$  is available for that month (Schmidt, personal communication). The main differences between the original and the new ozone profiles for that month are

- (1) higher values in the new profiles above 0.05 mb
- (2) higher values in the peak at 10 mb., in the new low latitude profiles (but lower values at  $\pm 60^\circ$  latitude).

Barnett and Corney /5/ suggested a 30% decrease in total ozone would produce typically a  $+1\text{K}$  increase in retrieved temperature between 150-20 mb and a  $2\text{K}$  increase between 20 to 2.5 mb. To examine the effect of the temperature retrieval on the vertical structure of the ozone variations, a comparison was made between the zonal mean temperature retrievals for Day 305, 1979, using the original (climatological) ozone profile, and the retrievals obtained after perturbing this profile at various levels. The perturbation applied was a 20% increase at a selected level, decreasing above and below by 4% per 0.2 scale heights, so that the unperturbed value resumes at  $\pm 1$  scale height either side of the perturbation. The results are listed in Table 1 for perturbations applied at 8 different levels. The standard deviation refers to the variation in result across the twelve latitude bands ( $45^\circ\text{S}$ - $65^\circ\text{N}$ ).

Table 1  
Response of Retrieved Temperature (units:  $0.01\text{K}$ ) to  $\text{O}_3$  Perturbations of +20%

Level of Max. Perturbation	Response at level:							
	70mb	20mb	7mb	2mb	0.6mb	0.2mb	0.06mb	0.02mb
70mb	$-42 \pm 9$	$-11 \pm 3$	$+1 \pm 2$	$0 \pm 3$	$0 \pm 2$	$0 \pm 2$	$-9 \pm 3$	$-6 \pm 2$
20mb	$+48 \pm 17$	$-86 \pm 11$	$-4 \pm 5$	$+1 \pm 6$	$+4 \pm 4$	$-7 \pm 11$	$+27 \pm 4$	$23 \pm 4$
7mb	$-33 \pm 28$	$+35 \pm 13$	$-6 \pm 8$	$-22 \pm 4$	$+25 \pm 8$	$+103 \pm 7$	$+21 \pm 7$	$-17 \pm 2$
2mb	$-26 \pm 15$	$+9 \pm 10$	$-13 \pm 6$	$-15 \pm 6$	$+14 \pm 6$	$+33 \pm 12$	$+28 \pm 6$	$-2 \pm 4$
0.6mb	$+10 \pm 8$	$-2 \pm 2$	$-1 \pm 1$	$-1 \pm 2$	$+1 \pm 2$	$0 \pm 2$	$0 \pm 1$	$+1 \pm 1$
0.2mb	$+13 \pm 16$	$-4 \pm 7$	$-1 \pm 4$	$+3 \pm 4$	$-3 \pm 3$	$-9 \pm 10$	$0 \pm 2$	$+4 \pm 4$
0.06mb	$+13 \pm 16$	$-6 \pm 7$	$-1 \pm 4$	$+3 \pm 4$	$-3 \pm 3$	$-9 \pm 10$	$+1 \pm 2$	$+4 \pm 3$
0.02mb	$+1 \pm 1$	$0 \pm 0$	$0 \pm 0$	$0 \pm 0$	$0 \pm 0$	$0 \pm 0$	$0 \pm 0$	$0 \pm 0$

One particularly significant result is that the temperature retrievals below 10 mb are just as sensitive to the shape of the ozone profile at those levels as they are to the total column amount, so the effect of introducing the more specific ozone profiles on the temperature cannot be generalized.

#### (b) Sensitivity of constituent retrievals to temperature

The sensitivity of the retrieved constituent profiles to the shape of the temperature profile was tested in a similar manner. The amplitude of the perturbation was  $1\text{K}$  and the shape the same as for ozone. For each

perturbation, temperature and constituent retrievals were performed for the whole of November, 1979. The resulting changes in the monthly profiles are shown in tables 2 and 3.

As expected, the sign of the perturbation is negative down the diagonal elements of each table (a higher local temperature implying a lower concentration for a given radiance). The conclusion here is that, in order to account for the approximately 50% reduction in mixing ratios implied by the balloon measurements at 20 mb, the actual atmospheric temperatures would have to be 5 to 10K higher than was measured by SAMS. This is a factor of 5 greater than the estimated error in SAMS temperatures from all sources.

Table 2  
% Response of Retrieved  $N_2O$  to Temperature Perturbations of +1K

Level of Max. Perturbation	Response at level:					
	70mb	20mb	7mb	2mb	0.6mb	0.2mb
70mb	$-2.8 \pm 11.5$	$-1.2 \pm 3.1$	$+0.4 \pm 0.8$	$-0.3 \pm 0.6$	$+0.2 \pm 0.7$	$-0.7 \pm 5.8$
20mb	$+5.1 \pm 8.5$	$-12.5 \pm 5.2$	$+1.3 \pm 0.6$	$-0.8 \pm 1.2$	$+1.4 \pm 1.7$	$+5.6 \pm 5.6$
7mb	$+5.6 \pm 7.3$	$-3.0 \pm 7.6$	$-8.2 \pm 2.7$	$+2.1 \pm 1.6$	$+1.3 \pm 5.6$	$+3.5 \pm 3.9$
2mb	$+2.7 \pm 2.4$	$-0.2 \pm 1.8$	$-0.5 \pm 0.4$	$-4.0 \pm 0.8$	$+1.0 \pm 0.7$	$+1.1 \pm 1.2$
0.6mb	$-2.4 \pm 13.0$	$-1.2 \pm 3.3$	$+0.2 \pm 0.8$	$-0.3 \pm 0.5$	$-1.8 \pm 0.9$	$-1.4 \pm 6.6$
0.2mb	$+0.5 \pm 2.6$	$-0.2 \pm 1.7$	$-0.1 \pm 0.4$	$+0.0 \pm 0.2$	$-0.2 \pm 0.3$	$-0.1 \pm 1.0$

Table 3  
% Response of Retrieved  $CH_4$  to Temperature Perturbations of +1K

Level of Max. Perturbation	Response at level:					
	70mb	20mb	7mb	2mb	0.6mb	0.2mb
70mb	$-0.3 \pm 2.0$	$-0.2 \pm 1.6$	$+0.2 \pm 0.3$	$-0.1 \pm 0.2$	$+0.1 \pm 0.4$	$+0.0 \pm 1.1$
20mb	$4.3 \pm 6.3$	$-5.5 \pm 5.3$	$-0.7 \pm 0.7$	$+0.3 \pm 0.3$	$-0.1 \pm 0.2$	$+2.2 \pm 3.1$
7mb	$5.1 \pm 7.5$	$-5.1 \pm 6.2$	$-0.8 \pm 2.6$	$-0.4 \pm 2.3$	$+1.4 \pm 2.9$	$+1.2 \pm 3.7$
2mb	$3.2 \pm 1.8$	$+0.3 \pm 1.3$	$-1.1 \pm 0.3$	$-5.1 \pm 0.3$	$-0.1 \pm 0.4$	$+1.7 \pm 1.1$
0.6mb	$3.1 \pm 1.6$	$+0.8 \pm 0.9$	$-0.2 \pm 0.2$	$-1.5 \pm 0.3$	$-4.8 \pm 0.6$	$+1.6 \pm 1.3$
0.2mb	$1.0 \pm 0.6$	$+0.4 \pm 0.3$	$-0.1 \pm 0.2$	$-0.1 \pm 0.1$	$-0.8 \pm 0.2$	$-0.9 \pm 0.6$

#### (c) New retrievals of $N_2O$ and $CH_4$

The final experiment of this set was to retrieve temperatures for the whole of November 1979 using the SBUV/LIMS ozone set rather than the global/annual mean used in the original retrievals, and then to re-retrieve  $N_2O$  and  $CH_4$  for this month using the new temperature profile for each day. The resulting differences in the monthly mean are given in Table 4.

Table 4  
Effect of using specific  $O_3$  profile on November 1979 Retrievals

Constituent	% Change in vmr at level:				
	20mb	7mb	2mb	0.6mb	0.2mb
$N_2O$	$+4.3 \pm 11.3$	$+8.4 \pm 11.6$	$-1.4 \pm 3.6$	$+3.7 \pm 4.2$	$+4.0 \pm 7.6$
$CH_4$	$+2.6 \pm 3.0$	$+7.4 \pm 7.8$	$-1.6 \pm 1.1$	$+0.2 \pm 1.8$	$-2.2 \pm 2.3$

The results show that use of a specific ozone profile results in a general increase in the constituent mixing ratios. If table 4 is compared to Table 1 of paper 1, it can be seen that the effect is too small, and of the opposite sign, to that required to explain the discrepancies between SAMS and in-situ measurements for that month.

#### SEARCH FOR TRENDS IN CH<sub>4</sub> AND N<sub>2</sub>O ABUNDANCES

Both nitrous oxide and methane are increasing in the troposphere and it is interesting to consider whether this is reflected in the stratospheric abundances at various levels. The use of satellite data to look for trends must be approached with caution, however, since the measurements techniques are novel and subject to errors, while the expected trends are quite small. The data of Rowland /6/, for example, shows methane trends of +1.25% per annum between 1979 and 1985 at the surface, which we might expect to see repeated in the stratosphere if it has been going on long enough. With 5 years of data to examine, a total change of around 6% would be expected; small compared to the estimated uncertainty in the data (20% or more, see paper 1) but perhaps just possible to detect since most of the errors in the data are systematic. In fact, the results (see tables below) are inconclusive.

In each case we have looked at three year means, using the same data set as in paper 1, and also five year means, using the entire SAMS data set. The latter are obviously better in some ways in looking for trends, except that the data from SAMS was of poorer quality towards the beginning and end of its lifetime. In the former case, the instrument was still being characterized and was used in various exploratory modes; in the latter, there were problems with the instrument, leading to intermittent data taking, and with the atmosphere, which was atypical in behaviour due to the eruption of el Chichon. In fact, similar results were obtained from both sets. Table 1 shows the temperatures and their standard deviations. A warming of around 0.15 degrees maybe present near the 2mb level. Tables 2 and 3 show the percentage changes in the minor constituents; again, some levels exhibit changes which appear marginally statistically significant but the evidence is unconvincing. The main conclusion to be drawn from this study is that a longer data set of more precise data is needed to identify trends.

Table 5. Temperature trends and standard deviations

<u>Pressure level</u> <u>(mb)</u>	<u>°K change/yr</u> <u>(3 year set)</u>	<u>°K change/yr</u> <u>(5 year set)</u>
20	0.09±0.12	0.31±0.14
7	-0.06±0.13	-0.10±0.08
2	0.14±0.07	0.17±0.04
0.6	-0.10±0.16	-0.25±0.11
0.2	0.15±0.21	0.01±0.14

Table 6. Methane trends and standard deviations

<u>Pressure level</u> <u>(mb)</u>	<u>% change/yr</u> <u>(3 year set)</u>	<u>% change/yr</u> <u>(5 year set)</u>
20	10.97 ± 5.21	5.50 ± 5.53
7	5.53 ± 4.08	6.04 ± 2.08
2	-12.12 ± 4.27	-5.55 ± 5.99
0.6	-5.47 ± 6.67	0.51 ± 10.24
0.2	4.38 ± 5.79	4.81 ± 5.08

Table 7. Nitrous oxide trends and standard deviations

<u>Pressure level</u> <u>(mb)</u>	<u>% change/yr</u> <u>(3 year set)</u>	<u>% change/yr</u> <u>(5 year set)</u>
20	-21.52 $\pm$ 15.15	-7.34 $\pm$ 8.63
7	7.37 $\pm$ 8.94	5.19 $\pm$ 4.94
2	-14.31 $\pm$ 11.6	0.38 $\pm$ 10.61
0.6	-6.38 $\pm$ 24.64	-2.25 $\pm$ 14.27
0.2	11.89 $\pm$ 25.27	-17.04 $\pm$ 12.21

## CONCLUSIONS

The results of this work are summarized as follows:

1. The model presented in paper 1 with monthly values for the vertical and latitudinal distribution of methane and nitrous oxide is the best which can be produced with present data.
2. There do seem to be real discrepancies between satellite and balloon data at lower levels in the middle atmosphere, but we have been unable to explain these by limitations in the data reduction methods used for SAMS.
3. There are no systematic trends in the middle atmosphere abundances of the two species studied which can be detected reliably with the data available; this is consistent with expectations based on other data.
4. Further progress awaits new instruments like those forming the scientific payload of the Upper Atmosphere Research Satellite /7/. This will include an improved version of SAMS, called ISAMS, which will measure methane and nitrous oxide with much greater sensitivity and precision.

## REFERENCES

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