N95-11099

VERTICAL DISTRIBUTION OF CH₄ and N₂O OVER THE TROPICAL SITE HYDERABAD

Shyam Lal and B.H. Subbaraya Physical Research Laboratory Ahmedabad 380 009, India

P. Fabian^{*} and R. Borchers Max Planck Institute for Aeronomy D-3411, Katlenburg-Lindau-3, Germany

ABSTRACT

Vertical distribution profiles of N₂O and CH₄ have been measured from Hyderabad, India using balloon borne cryogenic air sampler. The samples have been analysed using gas chromatographic techniques. Results for two balloon flights made in 1987 and 1990 show effects of tropical characteristics like higher tropopause and upwelling motion due to Hadley circulation. These profiles also exhibit perturbations around 25 km height, which are likely to be due to dynamical effects. A comparison with the SAMS data show that the SAMS values for both these gases are higher by a factor of about 1.5 to 2 around 30 km height.

1. INTRODUCTION

Nitrous oxide and methane are the two important trace gases which have biogenic sources. Both these gases play important role in atmospheric chemistry and radiation processes, particularly relating to ozone and global warming. Methane also reacts with OH radical which is the controlling radical for entry into the stratosphere for many trace gases. These gases have also been identified to play significant role in global warming through the absorption of the outgoing earth's infrared radiation (Ramanathan et al., 1985; IPCC, 1990). It has been found that methane and nitrous oxide contribute about 21% to the total change in radiative forcing from 1980 to 1990 (IPCC, 1990). Abundances of these gases have been increasing in the earth's atmosphere. While, nitrous oxide is found to be increasing at a rate of about 0.2-0.3% per year (Weiss, 1981; Rasmussen and Khalil, 1986; Robinson, 1988; Prinn et al., 1990; Sheel et al., 1990; WMO, 1991), methane is increasing at a rate of about 0.8-1% per year (Blake and Rowland, 1988; WMO, 1992). Tropical regions contribute about 50% of the total global sources

*Presently at : Lehrstuhl fur Bioklimatologie und Immissionsforschung, der Universitat Munchen Hohenbachernstr.22, of both these gases (WMO, 1985). Tropical forests and oceans are the major sources of N_2O . The North-West Indian ocean (15-25°N) represents one of the most significant marine source of N_2O (Law and Owens, 1990). Major sources of methane are natural wetlands, rice paddies and enteric fermentation. These too have large spread in the measurements (IPCC, 1990).

The vertical distribution data, which are important for modelling the chemistry of the troposphere and stratosphere are also limited particularly in the tropical region. There have been a series of measurements from the midlatitude region using balloonborne sampling technique (Ehhalt, 1980; Fabian et al., 1981; Schmidt et al., 1984; Lal et al., 1989a). However, only few measurements of these gases have been made in the tropical regions (Ehhalt and Toennissen, 1979; Lal et al., 1989b; Borchers et al., 1989).

A programme has been undertaken jointly by Physical Research Laboratory (PRL), Ahmedabad, India and Max Planck Institute for Aeronomie (MPAE), Lindau, Germany to study the vertical distribution of trace gases from Hyderabad (17.5°N). This paper presents the results of two balloon flights conducted from this site on March 27, 1987 and April 9, 1990.

2. EXPERIMENTAL DETAILS

Three balloon flights carrying a composite payload consisting of a cryogenic air sampler of MPAE, Lindau and a suntracking multichannel photometer of PRL, Ahmedabad were launched from the tropical site, Hyderabad (17.5°N). The details of the balloon flights are given in Table The neon cooled cryogenic air sampler 1. collected 15 samples during each flight in the height range of 10 to 35 km. The descent of the balloon was controlled using an apex valve and some of the samples were collected during this part of the trajectory also. The first flight in 1985 was a test flight and adequate amount of samples could not be collected during this flight. Air samples of about 10 litre at STP were collected with sampling times ranging from 1

W-8050 Freising Weihenstephan, Germany

Launch Date	Launch Time Hrs. (IST)	Cut-off Time Hrs. (IST)	Ceiling Altitude (km.)	Experiments
27.03.1985	0608	1150	35.3	Cryogenic air sampler and multichannel photometer
26.03.1987	0545	1508	34.5	-do-
09.04.1990	0536	1350	33.7	-do-

Table 1 : Details of balloon flights from Hyderabad, India for trace gas studies

minute at 11 km to 60 minutes at 34 km during the last two flights. Further, details of the cryogenic air sampler and sampling procedure are given in Fabian (1981).

The collected air samples have been analysed at the MPAE, Lindau using gas chromatographic and coupled mass-spectrometric techniques for N_.O and CH_. alongwith many other trace gases. Nitrous oxide was analysed using a gas chromatograph coupled with a quadrupole massspectrometer while methane was analysed using a gas chromatograph with a Flame Ionization Detector (FID). The analyses were done against a laboratory standard which was calibrated using known mixtures of high purity gases. The repeatability of the analyses is better than 2%. However, the errors in calculating absolute mixing ratios which arise mainly due to calibration mixtures are of the order of 5%.

3. RESULTS

The profiles of CH₄ and N₂O obtained from balloon flights in 1987 and 1990 are shown in Figures 1 and 2 respectively. Both the gases show almost uniform mixing ratio in the altitude region below 20 km. Above this height the mixing ratios start decreasing. This feature is concurrent with the high tropical tropopause. Also, there is no appreciable difference in the tropospheric values of these gases for the two flights.

In the stratosphere, mixing ratio of $N_{n}O$ decreases much faster than CH_4 due to higher loss rate compared to CH_4 . The variability observed in the two sets of profiles for the two gases is identical. The 1987 profiles of CH $_{\rm d}$ and $\rm N_{2}O$ show sudden change around 25 km but this feature is not observed in the profiles of 1990. The 1987 flight shows an increasing trend in the mixing ratios of the two gases above 25 km followed a slow decrease. However, there is a sharp fall beyond 30 km but there is only a single data point. Contrary to this, the 1990 flight shows a steady decreasing trend throughout except for a small decrease in the height range of 22 to 25 km. This indicates that the region around 25 km height has some special features. Wind data for the two flight dates do not show any appreciable change around 25 km altitude eventhough the wind profiles do differ in the tropospheric region Similar abnormality in several (Fig. 3a,b).



Fig. 1. Vertical distribution of CH₄ measured over Hyderabad during 1987 and 1990 and a comparison with other measurements.

longlived trace gases has been seen in the profiles measured from Southern France (44°N) but at a lower height around 22 km (Lal et al., 1989a,b); Borchers et al., 1989). The perturbation observed in these profiles appears to be due to dynamical effects as there cannot be sudden change in the photo-dissociation rates of these gases. Spectroscopic measurements of CH₄ and N₂O during a shuttle mission from April 30 to May 6, 1985 consistently showed a 'fold' around 30 km altitude (Gunson et al., 1990).

4. COMPARISON WITH OTHER MEASUREMENTS

Figures 1 and 2 also show a comparison of the Hyderabad profiles with the average profile based on measurements made from Southern France (44°N) using similar sampling and analysis techniques. Measurements made with spaceborne techniques like Stratospheric and Mesospheric



Fig. 2. Vertical distribution of N_2O measured over Hyderabad during 1987 and 1990 and a comparison with other measurements.

Sounder (SAMS) and the Atmospheric Trace Molecule Spectroscopy (ATMOS) flown onboard Spacelab-3 (SL-3) mission given by Taylor et al. (1989) and Gunson et al. (1990) respectively, are also presented in these diagrams. Comparison with the mean mid-latitude data show very clearly the effect of upwelling in the Hyderabad profiles. The Hyderabad profiles show decrease in the mixing ratio only above 20 km while the midlatitude profile shows decrease above 15 km itself. However, above about 28 km the Hyderabad data fall within the range of the mid-latitude data. This sharp gradient above 25 km in the Hyderabad profiles appear to be due to intense solar radiation prevailing in the tropical region compared to the mid-latitude region and consequent increased photodissociation.

Average data for 20° N for the month of March based on the SAMS data for both the gases are presented in Figures 1 and 2. In case of CH₄, the SAMS values are higher than the Hyderabad values at all altitudes except three data points of the 1987 flight in the 26 to 30 km altitude. The increase is by a factor of about 1.5. Similarly, N₂O values derived from the SAMS results are higher but the difference increases with height. Around 30 km, the SAMS values are higher by a factor of about 2.

The ATMOS experiment data for CH_4 is within the average values of the mean mid-latitude profiles. However, the N₂O values are almost representing the Southern France data.

5. SUMMARY

The measurements of CH_4 and N_2O made from Hyderabad show the effect of strong upwelling motion. Over Hyderabad, the constant mixing ratio region extends upto almost 20 km height as against 15 km observed in the mid-latitude region. There is a sharper decrease in the mixing ratios of these gases above 25 km compared to the mid-latitude profiles, probably, due to higher amount of solar radiation reaching at these heights in the tropics.

The SAMS data show higher mixing ratio of these gases compared to the Hyderabad profiles while the ATMOS experiment data for both the gases for $28^{\circ}N$ almost represent the mid-latitude profiles. This aspect needs more in-situ, preferably simultaneous, observations, to delineate the biases.

ACKNOWLEDGEMENTS

These balloon flights were conducted jointly by PRL, Ahmedabad and MPAE, Lindau, Germany under the ISRO (India) and the DLR (Germany) collaborative programme. We would like to thank the members of two organizations for their help in coordinating and executing this programme. Thanks are also due to Prof.S.V. Damle, Mr.R.T. Redkar and other team members of TIFR Balloon Facility, Hyderabad and to the members of PRL, MPAE and DLR who helped in conducting these experiments. We also acknowledge the support of Mr. P. Rajaratnam of ISRO Headquarters, Bangalore for successfully managing these campaigns.

REFERENCES

- Blake, D.R. and F.S. Rowland, 1988: Continuing worldwide increase in tropospheric methane, 1978 to 1987, <u>Science</u>, <u>239</u>, 129.
- Borchers, R., P. Fabian, O.N. Singh, S. Lal and B.H. Subbaraya, 1989: The vertical distribution of source gases at tropical latitudes, In "<u>Ozone in the Atmosphere</u>", Proceedings of the Quadrennial Ozone Symposium, 1988 and Tropospheric Ozone Workshop, Eds. R.D. Bojkov and P. Fabian, A. Deepak Publishing, USA., 290.
- Ehhalt, D.H., 1980: In-situ observations, <u>Phil.</u> <u>Trans. R. Soc., Lond., A.296</u>., 175. Ehhalt, D.H. and A. Toennissen, 1980: Hydrogen
- Ehhalt, D.H. and A. Toennissen, 1980: Hydrogen and carbon compounds in the stratosphere, In "<u>Proceedings of the NATO Advanced Study</u> <u>Institute on Atmospheric Ozone : Its</u> <u>variations and Human Influences</u>, Rep. No.FAA-EE-80-20, USA., Ed. C. Aikin, 129.
- Fabian, P., 1981: Atmospheric sampling, <u>Adv.</u> <u>Space Res., 1</u>, 17.
- Fabian, P., R. Borchers, G. Flentje, W.A. Mathews, W. Seiler, H. Giehl, K. Bunse, F. Muller, U. Schmidt, A. Volz, A. Khedim and F.J. Johnen, 1981: The vertical distribution of stable trace gases at midlatitudes, J. Geophys. Res., 86, 5179.



Fig. 3a & b. Wind profiles measured during the flight periods.

- Gunson, M.R., C.B. Farmer, R.H. Norton, R. Zander, C.P. Rinsland, J.H. Shaw and B.C. Gao, 1990: Measurements of CH₄, N₂O and O₃ in the middle atmosphere by the atmospheric trace molecule spectroscopy experiment on spacelab 3, J. Geophys. Res., 95, 13867. I.P.C.C., 1990: Climate Change : "The
- I.P.C.C., 1990: Climate Change : "The Intergovernmental Panel on Climate Change -Scientific Assessment, Eds. J.T. Houghton, G.J. Jenkins and J.T. Ephraums, Cambridge Univ. Press., Cambridge, USA.
- Lal, S., R. Borchers, P. Fabian and B.C. Krueger, 1989a: The vertical distribution of CH₄, N₂O, CFC-12 and CFC-11 in the middle atmosphere at mid-latitudes, <u>J. Atmos.</u> <u>Terr. Phys.</u>, <u>51</u>, 81.
- Lal, S., B.H. Subbaraya, S. Venkataramani, P. Fabian and R. Borchers, 1989b: Minor constituents in the tropical atmosphere, Ind. J. Radio & Space Physics, 18, 171.
- Law, C.S. and N.J.P. Owens, 1990: Significant flux of atmospheric nitrous oxide from the north-west Indian ocean, <u>Nature</u>, <u>346</u>, 826.
- Prinn, R., D. Cunnold, R. Rasmussen, P. Simmonds, F. Alyes, A. Crawford, P. Fraser and R. Rosen, 1990: Atmospheric emissions and trends of nitrous oxide deduced from 10 years of ALE-GAGE data, J. Geophys Res., 95, 18369.
- Ramanathan, V., R.J. Cicerone, H.B. Singh and J.T. Kiehl (1985) : Trace gas trends and their potential role in climate change, <u>J.</u> <u>Geophys. Res.</u>, <u>90</u>, 5547.

- Rasmussen, R.A. and M.A.K. Khalil, 1986: Atmospheric trace gases : Trends and distributions over the last decade, Science, 232, 1623.
- Scheel, H., E. Brunke and W. Seiler, 1990: Trace gas measurements at the monitoring station, Cape Point, South Africa, between 1978 and 1988, J. Atmos. Chem., 11, 197.
- Schmidt, U., A. Khedim, D. Knapska, G. Kulessa and F.J. Johnen, 1984: Stratospheric trace gas distributions observed in different season, Adv. Space Res., <u>4</u>, 131.
- Taylor, F.W., A. Dudhia and C.D. Rodgers, 1987: Proposed reference models for nitrous oxide and methane in the middle atmosphere, <u>Adv. Space Res</u>., 7(9), 49.
- Weiss, R.F., 1991: The temporal and spatial distribution of tropospheric nitrous oxide, J. Geophys. Res., <u>86</u>, 7185.
- W.M.O., 1985: "Atmospheric Ozone 1985", World Meteorological Organization, Rep. No. 16, Vol. I,
- W.M.O., 1991: "<u>Scientific Assessment of Ozone</u> <u>Depletion</u>: <u>1991</u>, World Meteorological Organization, Rep. No. 25.