N95-11105

LOCAL FLUCTUATIONS OF OZONE FROM 16 KM TO 45 KM DEDUCED FROM *IN SITU* VERTICAL OZONE PROFILE

G. MOREAU, C. ROBERT

Laboratoire de Physique et Chimie de l'Environnement, CNRS
45071 Orléans, France

ABSTRACT

A vertical ozone profile obtained by an *in situ* ozone sonde from 16 km to 45 km, has allowed to observe local ozone concentration variations. These variations can be observed, thanks to a fast measurement system based on a UV absorption KrF excimer laser beam in a multipass cell. Ozone standard deviation versus altitude calculated from the mean is derived. Ozone variations or fluctuations are correlated with the different dynamic zones of the stratosphere.

1. INTRODUCTION

Most common ozone sondes, used up to now, have allowed to obtain fairly reliable ozone concentrations, at least in the lower and middle stratosphere. Today, these launched sondes are electrochemical [Komhyr *et al*, 1980], chemiluminescent [Hilsenrath and Kirschner, 1980; Aimedieu, 1981], and UV absorptive [Proffitt and McLaughlin, 1983]. However, all these probes have disadvantages, mainly due to ozone decomposition on the walls of the pumps and pipes. Though several modern materials are available, which decompose ozone only at low rates, the reversible transfer of ozone from walls to the gaseous phase filters observable ozone variations throughout the measurements. This can be partially eliminated by a fast air flow.

In addition, temperature and pressure inside the instrument have to be measured carefully, since ozone measurements are made only at low pressures. Some additional uncertainty on ozone concentrations results by using *in situ* ozone sondes, when passing through aerosol layers. In mid-1991, the Pinatubo volcano sent up great amounts of aerosols into the atmosphere. The reliability of ozone profiles obtained in the lower stratosphere have been questioned since then.

The multipass ozonometer has been designed to reduce wall effects, thanks to a multipass cell in which air enters freely by natural convection. The RMS differential wind speed between balloon and gondola is over 1m/s. Absorption takes place on a long optical path - exactly L = 245.27 m - thanks to 74 reflections on two concave mirrors 3.256 m away from each other. The number of reflections, and consequently the absorption path, is defined and adjusted before launching. It can be easily varied up to 120, the maximum absorption path length can therfore reach 400 m. The chosen number of reflections is a compromise between detector S/N ratio, unavoidable mirror absorption, and ozone absorption at 40 km (we aim at measuring ozone with 2 % accuracy at an altitude of 40 km).

The light source is an excimer laser emitting 40 ns duration pulses, every 4 seconds, at a wavelength of 248.5 nm. Transmission measurements are performed at the same rate, and ozone concentration is deduced using the Beer-Lambert law. Total transmission, without absorption, is measured when flushing out air by inert gas.

With such a device, one measurement lasts less than 100 ns, i.e., the pulse transit time inside the multipass cell. Rather fast ozone variations can therefore be observed during a vertical ascent or descent, or along a floating altitude. Since ozone absorption is dominant over attenuation by aerosols, especially in the lower stratosphere, this instrument is quite suitable for ozone when aerosols are present, whereas aerosols increase uncertainty in other ozone sondes by chemical effects.

2. THE EXPERIMENT

The multipass ozone sonde has been launched 6 times since the first flight report [Moreau and Robert, 1989], from French sites located at Aire Sur L'Adour (N 43.42, W 00.15) and Gap (N 44.27, E 06.01).

Improvements have been made since then to achieve the expected instrument performances.

Laser beam adjustments, and generally speaking, optics, are maintained during flights, even at the low stratospheric temperatures, thanks to the self adusting Herriott two-mirror cell [Moreau and Robert, 1985. Two fast vacuum photodiodes collect the two reference and absorbed beams. Small nonlinearities exist in detection electronics, but they mutually cancel each other out when signals from the two detectors are equal. In all cases, corrections are made, deduced from known optical densities.

There is no measurable deviation from the Beer-Lambert law, because of the low energy laser pulses: 10^{11} photons are sufficient (or 80 nJ). Considering absorption and losses on mirrors, only 10^{9} photons are absorbed at 40 km, which is quite negligible compared to the 10^{15} ozone molecules "seen" by the laser beam.

One of the main difficulties with this probe lies in the mirrors. The dielectric mirror reflectance varies normaly with temperature, but water trapped inside the dielectric coatings enhances this effect. The solution for reducing the influence of temperature consists in removing water from the coatings in a moderate vacuum, just before the flights, and in maintaining the mirrors at a constant temperature by thermal regulation during the flights.

The smallest measurable absorptions are not less than 10^{-3}, meaning 4 10^{9} molecules/cm^3 ozone concentration. However, accuracy is in the order of 8 10^{9} molecules/cm^3, i.e. , 1.5 % at an altitude of 40 km (uncertainty on σ, the ozone absorption cross section at 248.5 nm not included). Since absolute transmission without absorption is determined at low pressures and high altitudes, which enable fast and complete air flushings, extrapolation of absolute transmission to lower altitudes makes ozone measurements less accurate. An absolute value of 2 10^{10} molecules/cm^3, or 0.4 %, seems reasonable at around 25 km. Ozone absorption cross sections at a wavelength of 248.5 nm are from Molina [Molina and Molina, 1986]. The cross section has a value of σ = 1.080 10^{-17} cm^2 at 298 K, and increases to σ = 1.093 10^{-17} cm^2 at 226 K.

3. RESULTS

Results obtained during ascent near Gap (France), 2 July 1991, at the end of the night are reported. The altitude measurement range extends from 16.5 km to 45 km. Exact altitude is known from radar measurements to better than 50 m. Altitude and pressure fit very well with data found in tables.

Figure 1 shows the vertical profile resulting from this flight. Each point represents one measurement. It can be seen that the profile contains a certain number of "accidents" up to 28 km. Above 28 km, all points form a quite smooth line. At lower altitudes (from 16.5 km to 19.5 km), measurements are highly dispersed. A sudden change occurs however, at 19.5 km, at the bottom of an ozone "hole", where the dispersion curve becomes narrower. From 19.5 km to 28 km, dispersion tends to a minimum. At ceiling altitude, numerous points are within a distribution, with a calculated standard deviation of σ = 1.2 10^{10} molecules /cm^3. During the ascent, the gondola may be in the wake of the balloon if horizontal winds are low. In this case, there is destruction of local ozone continuity, if it exists, but the overall ozone distribution can be maintained. This happened up to 19.5 km, but not above, because wind was observed to increase abruptly at 19.5 km.

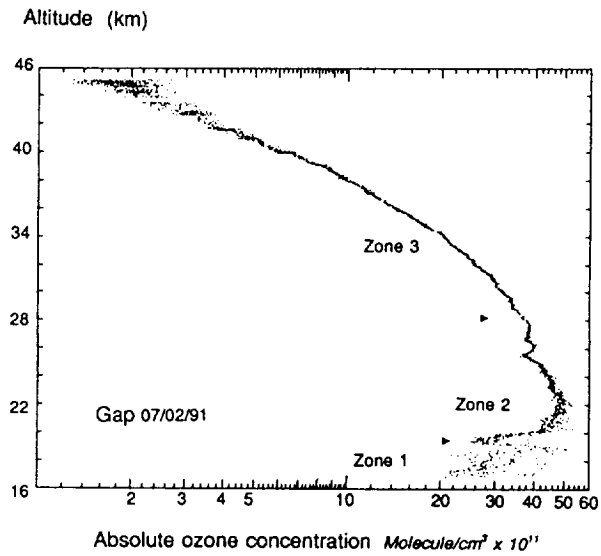

Fig. 1: The ozone profile obtained 7/02/1991 near Gap (France) is obtained, at the end of the night, from 16.5 km to 45 km, during ascent. Each point represents one ozone measurement. The stratosphere is divided into 3 zones, depending on the values of local ozone fluctuations.

The ozone standard deviation profile from a mean ozone profile is derived. The mean profile is the running mean calculated along 200 m increments, with a sinus function weight. The same weight function is applied for calculating the standard deviation curve. Figure 2 depicts the change in ozone standard deviation with altitude.

4. DISCUSSION

Figure 2 confirms quantitatively the description of the ozone profile. Three zones appear clearly:

1) From 16.5 km to 19.5 km, relative variations are in the order of 20 %. In this domain, ozone concentrations are known to change over very short periods, as has been observed by lidars [Claude and Wege]. Stratified layers with different concentrations come from different

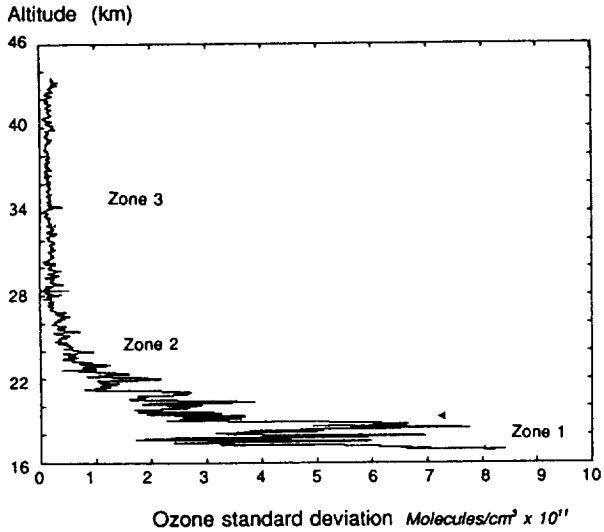

Fig. 2: Profile of ozone fluctuations, the standard deviations from a running mean. The 3 zones are clearly visible.

places. Vertical fluctuations are the counterpart of temporal fluctuations. When present, the aerosols have an influence on temperature and ozone. On the day of launch, aerosols were present. They were coming from Pinatubo volcano, which erupted on the beginning of june 1991. Pinatubo aerosols had been observed below 20 km at Garmisch-Partenkirchen located also at mid-latitude, one day before the flight [EOS, 1991] .

2) At 19.5 km, relative variations suddenly fall to about 6 %. From 19.5 km to 28 km, they decrease from 6 % down to 0.3 %. This zone is less dynamically active than the first one. Latitudinal mass transfers are also not as fast. Pinatubo aerosols arrived later in this zone, in September 1991.

3) Above 28 km, the atmosphere is calmer, due to the constancy of absolute local variations, about 2-3 times the detection limit. The standard deviation profile however presents small irregularities. One can be seen just above 34 km. Approaching floating altitude, in the wake of the balloon, points are apparently dispersed, but standard deviation from a straight line drawn along the profile, has the same value as below (above 28 km).

CONCLUSION

Thanks to its low detection limit and sensitivity, the multipass ozonometer can give new information on stratospheric ozone fluctuations during ascent or descent. Fluctuations are quantified by calculating standard deviation calculation from the mean profile. Normal ozone profile, in addition to standard deviation profile, exhibits the dynamic behaviour of the stratosphere.

This instrument has been selected as a correlative instrument for UARS.

Acknowledgements: We thank the Centre National d'Etudes Spatiales, which financed the gondola and provided launching facilities. It is a pleasure to thank the technical staff: G. Maille, G. Pesnel, J. Lebihan, and P.Badets.

REFERENCES

AIMEDIEU P., Measurement of the vertical ozone distribution by means of an *in situ* gas-phase chemiluminescence ozonometer during the intercomparison ozone campaign, Gap, France, June 1981, Planet. Space Sci., 31, 1981.

CLAUDE C., K. WEGE, The hohenpeissenberg lidar: first results and intercomparison with Brewer-Mast soundings, Proc. Quad. Ozone Symp., Göttingen FRG, 4-13 August 1988; A. Deepak Hampton Va USA, 739-742, 1989.

EOS Trans. Am. Geophys. Union, Extent of Pinatubo cloud probed, 72 (33), August 13 1991.

HILSENRATH E., P.T. KIRSCHNER, Recent assessment of the performance and accuracy of a chemiluminescent rocket sonde for upper atmospheric ozone measurement, Rev. Sci. Instrum., 51, 1381, 1980.

KOMHYR W.D., S.J. OLTMANS, A.N. CHOPRA, P.R. FRANCHOIS, Performance characteristics of high altitude ECC ozonesondes, Proc. Quad. Ozone Symp.; D. Reidel, Dordrecht, 499-503, 1985.

MOREAU G., C. ROBERT, P. RIGAUD, A new Multipass Ozonometer, Proc. Quad. Ozone Symp., Göttingen FRG, 4-13 August 1988; D. Reidel, Dordrecht 755-758, 1989.

MOLINA L.T., M.J. MOLINA, Absolute cross sections of ozone in the 185- to 350-nm wavelength range, J. Geophys. Res., 91(D13), 14501-14508.

MOREAU G., C. ROBERT, Etude des variations d'un faisceau lumineux dans une cellule à passages multiples, J. Optics, 16(4), 177-183, 1985.

PROFFITT M.H. , R.J. McLAUGHLIN, Fast-response dual-beam UV-absorption ozone photometer suitable for use on stratospheric balloons, Rev. Sci. Instrum., 54, 1719-1728, 1983.