(Reference II.10)

STRATOZ, AN EXPERIMENT OF RADIOMETRIC SURVEY OF THE STRATOSPHERE

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The last few years have been characterized by increased interest in the problem of the actual content in nitrogenic compounds of the stratosphere (ref. 1, 2).

Since Johnston and Crutzen pointed out that low concentrations in N_2O, NO, NO_2 (due to stratospheric flight and extensive use of fertilizers) could reduce the ozone concentration by means of a catalytic reaction process, numeric simulations of the photochimical equilibriums were attempted and stratospheric measurement were required.

STRATOZ, a joint experiment by E.E.R.M., O.N.E.R.A. and I.A.S.B. (1, 2, 3) consists in a survey of stratospheric O_3, HNO_3, NO, NO_2, HCl, $CFCl_3$, CF_2Cl_2 and CO on board a Caravelle aircraft which was cruising from Groenland to South America. Characteristics of the mission are given in figure 1.

Three instrument were flown on the Caravelle (4):

- a grille spectrometer } built by O.N.E.R.A.
- a NO_2 visible radiometer } built by O.N.E.R.A.
- a IR radiometer .... built by I.A.S.B.

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(4) The aircraft was from Centre d'Essais en Vol (C.E.V.) F - 91220 BRETIGNY-SUR-ORGE. Organization of the mission was supplied by C.E.V. and E.E.R.M.

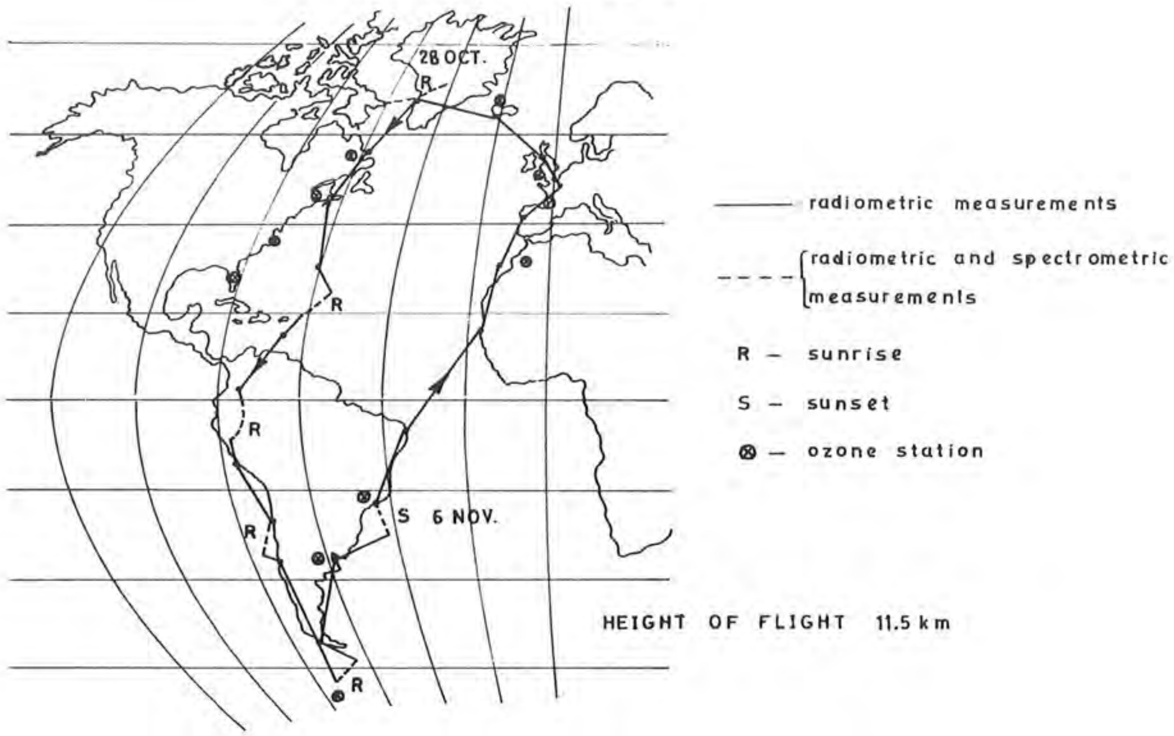

fig. 1 : Flight route for STRATOZ experiment

Grille spectrometer

Like in precedant experiences done by Girard et al., Ackerman et al. (1973 to 1978) (ref. 3), the grille spectrometer (ref. 4) is associated with a sun-pointer as shown in figure 2.

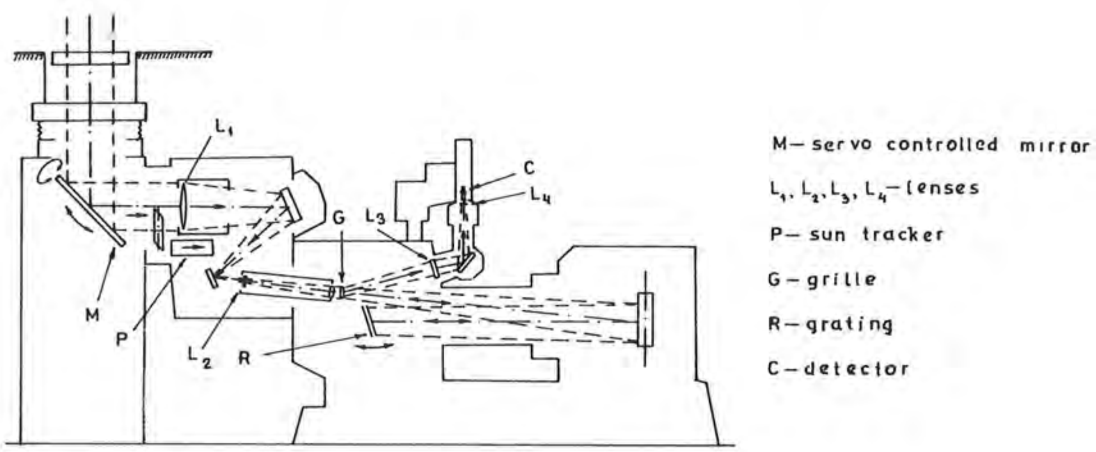

fig. 2 : optical scheme of grille spectrometer

Using the sun as a source, absorption measurements are made in the I.R. range between 3 and 10 microns. The long absorption path obtained at sunset or sunrise makes it possible to detect and to measure chemical species at very low mixing ratios (below 10^{-9} in volume). Because of the high resolution (0,1 cm^{-1}), great luminosity and full automatization of the spectrometer, narrow spectral ranges containing a few single lines are scanned rapidly : each spectral range of the measurement programm shown in table 1 is repeated on an average every 4 minutes, which is roughly the time interval corresponding to a variation of 1 degree in zenith angle of the sun.

studied constituant	spectral range (cm^{-1})	other constituants having week lines in the same range
HNO_3	1324 - 1328	CH_4, H_2O
CO	2135 - 2142	N_2O, O_3
NO_2	1603 - 1607	H_2O
HCl	2941 - 2949	CH_4
$CFCl_3$	1080 - 1085	H_2O, O_3
NO	1913 - 1919	CO_2
O_3	1723 - 1731	H_2O
CFCl2	1158 - 1162	O_3, CH_4

Table 1 : measurement program of Grille-spectrometer

Precision of the calculated vertical profiles of concentrations is poor above the height of flight. Good results are obtained for the lower altitudes. In particular, a great precision is obtained for the concentrations at altitude of flight by comparing absorption at 91° and 89° zenithal distances. The main paramater derived from theese measurements is the vertical column density.(ref. 5).

Complete results are not yet available. Figures 3 and 4 show latidudinal variations of total HNO_3 and NO_2 obtained for the "STRATOZ" Mission and comparison is possible with results of mission "Latitude Survey" (october, 1976) (ref. 10).

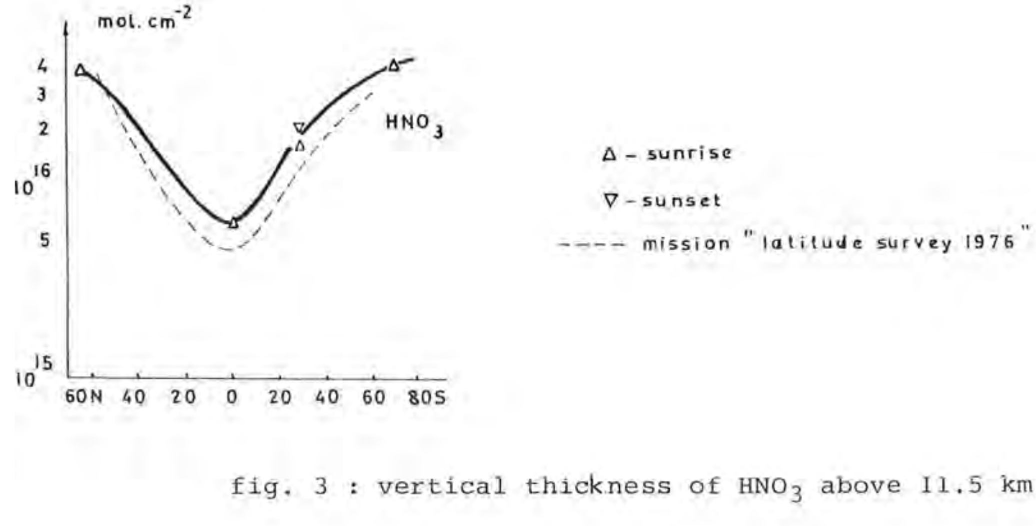

fig. 3 : vertical thickness of HNO_3 above 11.5 km

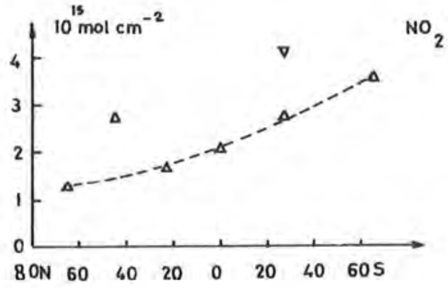

fig. 4 : vertical thichness of NO_2 above 11.5 km

Δ – sunrise } STRATOZ experiment
∇ – sunset }
------- mission "latitude survey 1976"

NO_2 - radiometer

The principle (ref. 6) of the radiometer is also an absorption measurement, but the source is the blue light of the sky scattered by molecules and aerosols at great altitudes. (fig. 5)

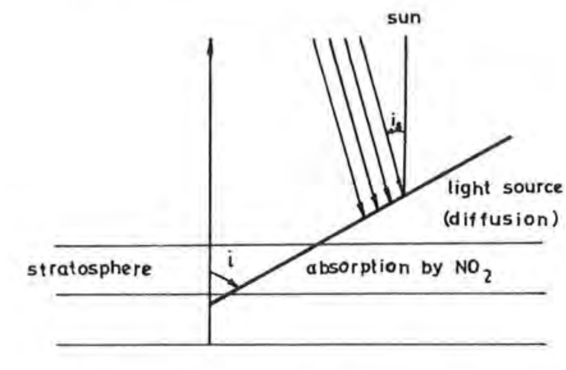

fig. 5 : principle of NO_2-radiometer

A filter selects a wide spectral area (between 0,4 and 0,5 microns) corresponding to the visible absorption band of NO_2. As the apparatus is non dispersive, detection of NO_2 is due to the deviation from the exponential extinction law which appears on a wide spectral range. A device with four cells (figure 6) filled with NO_2 yields a deviation which is proportionnal to the atmospheric NO_2 thickness.

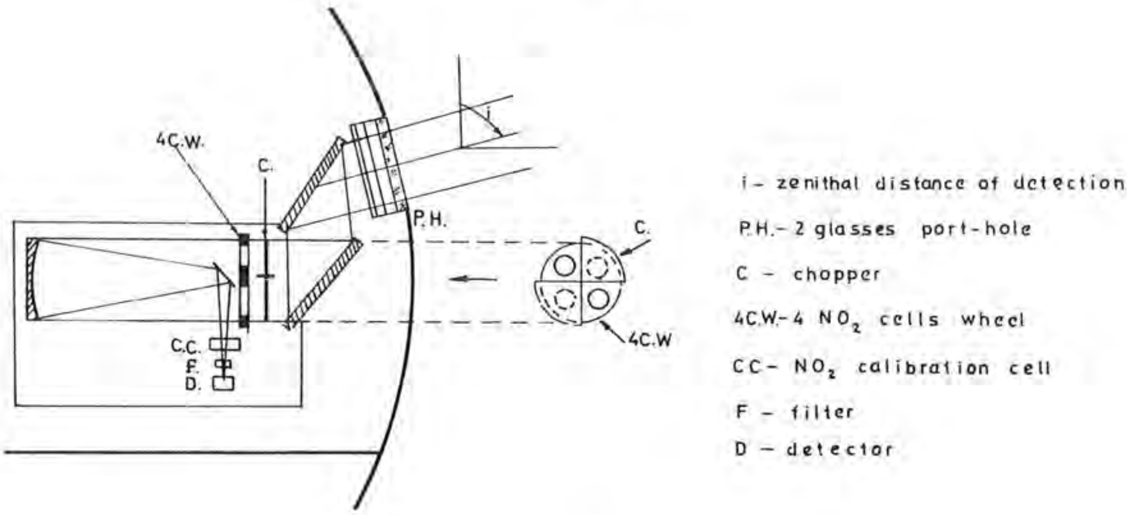

fig. 6 : schematic diagram of NO_2-radiometer

Actually, atmospheric vertical thickness E_A cannot be calculated from measured thichness E_M with the relation

$$E_A = E_M \times \cos i$$

because scattering and absorption are not separate in space. From an other point of view, it is not possible to consider that the sky blue luminance is constant with varying i, zenithal distance of sight and i_s, zenithal distance of the sun. For these reasons, it was necessary to compute, for each measurement, i.e for each (i, i_s) an equivalent of 1/cos i called number of atmospheres. The number of atmospheres N is defined as follows :

$$\frac{1}{N} = \frac{\text{vertical thickness for a given standard } NO_2 \text{ profile}}{\text{apparent thickness (A) for the same given profile}}$$

$$e^{-A} = \frac{\text{calculated sky luminance in direction of sight with a } NO_2 \text{ containing atm}}{\text{calculated sky luminance in direction of sight with a } NO_2 \text{ free atmosph.}}$$

The signal variation for a variation of 1 number of atmosphere gives the NO_2 thickness between 11.5 km and top of atmosphere. Then results of calibrations done with known thickness cells (calibration cells) leads to the value in ppm.m or molecules. cm^{-2}.

Up to this date, only the 6 flights with sun tracking have been analysed because they have large number of atmosphere variations. Figure 7 shows the result of this work.

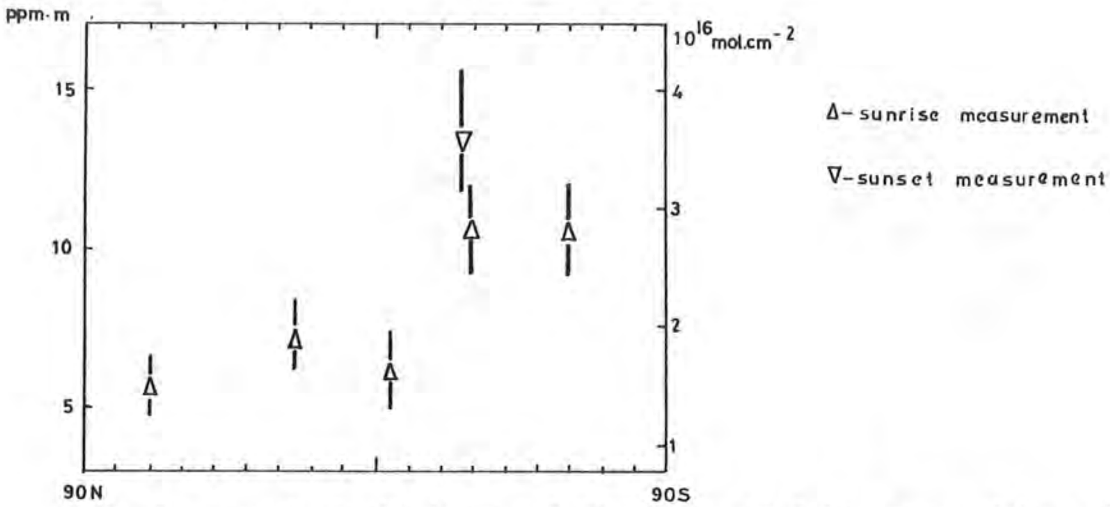

fig. 7 : vertical thickness of NO_2 above 11.5 km measured by radiometer

I.R. Radiometer (ref. 9)

Characteristics of the I.R. Radiometer are shown schematically in figure 8. With an accurate regulating of chopper temperature and frequent calibrations on emission of liquid nitrogen (before and after each flight), it is possible to get spectra of thermal emission of the stratosphere. Spectra obtained with the same kind of device during a balloon flight (1973) are shown in figure 9.

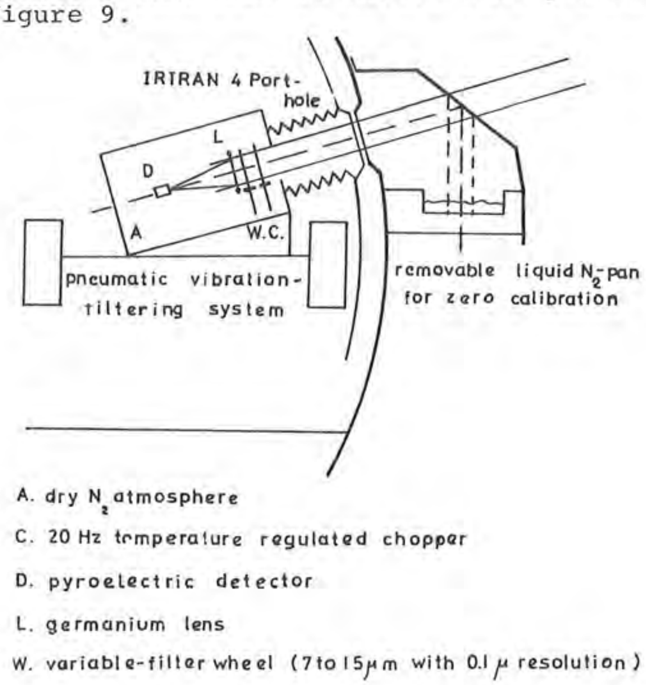

fig. 8 : schematic diagram of IR radiometer

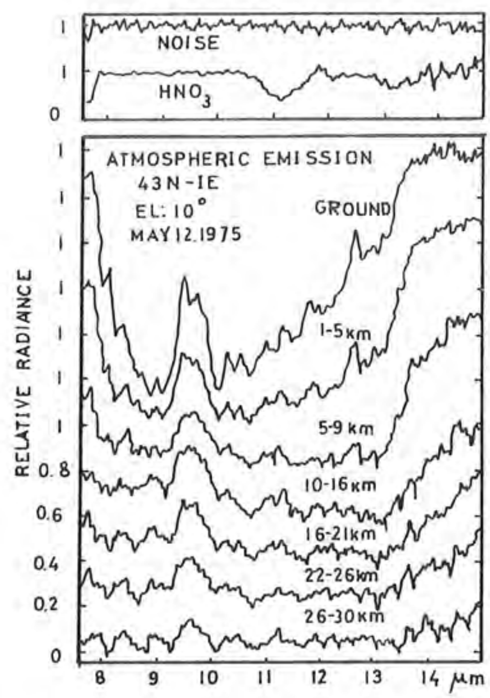

fig. 9 : spectra obtained during a balloon-flight

Thermal radiance is a combination of emission and absorption occuring along the optical path . For a given wave length, it can be written :

$$L_{sky} = \int_0^\infty k \cdot n(x)\, B_o\,(T(x))\, e^{-\int_0^x kn(r)\,dr}\, dx$$

k : absorption coefficient
n (x): concentration at distance x
B_o(T): Planck emission of Black Body at temperature T.

In the case of the ozone band at 9,6 microns the interpretation of the spectra as vertical profiles is not easy and is not yet achieved.

The HNO_3 emission band at 11,3 microns, for which the theorical spectrum is not well-known, seems still more appropiate for the determination of an order of magnitude of vertical HNO_3 thickness. As the concentrations are very low L_{sky} can be approximated by

$$L_{sky} \approx \int_0^\infty kn\,(x)\; B_o\,(T(x))\,dx$$

relation in which k can be taken form experimental data and corresponds to an absorption over the entire band.

Concluding remarks

1) The results obtained by spectrometry for NO2 latitudinal variations are in accordance with previous measurements (ref. 10). Radiometric measurements demonstrate the same relative increase for NO_2 vertical column density from high latitude in the winter hemisphere to high latitude in the summer hemisphere ; however the quantities calculated from the radiometric measurements are roughly 10 times greater than the spectroscopic ones.

   This can be attributed to experimental difficulties with the absolute calibration system of the radiometer.

   It is also noteworthy that measurements in the visible spectrum of NO_2 have led up to now to greater concentrations than IR measurements.

2) Spectrometric measurements are a very powerful tool for the knowledge of the stratosphere. They require well known theorical spectra for the studied species and for the species which have contaminating lines in the recorded spectral range. For O_3, HNO_3 and chlorofluoromethanes much spectroscopic work has still to be done. Radiometers have other advantages : they are easy to operate (at any time of the day) and were initially designed to fly on board commercial aircraft.

We intend to make further experiments of the same type in order to detect possible climatic variations - the seasonnal trends have also to be checked.

3) Comparisons are necessary with data of the ozone network and with data collected by NIMBUS 7 wich has been launched late october 1978. A study of the meteorological situations during the measurements period is also very useful to check against anormalous dynamic transport effects.

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