INFRARED SPECTROMETRY OF ATMOSPHERIC GASES : 1969-1992

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The proposal of using limb absorption spectrometry for atmospheric studies (Ackerman, 1963) dates back to the founding days of the Belgian Institute for Space Aeronomy (BISA). The use of this technique was suggested for measuring the vertical distribution of N_2O, CH_4, H_2O, O_3 and CO_2 in the earth's atmosphere from balloon gondolas and, later on, artificial satellites. This early program has continued to have a high priority today and it still constitutes the core of the present ESA Envisat program.

In order to achieve the aforementioned objective, the first eight years were spent in a survey program used first to determine the infrared solar flux at balloon altitudes. In 1967, flights of a spectrometer designed at BISA began using a stabilised balloon gondola. The resolution of 2.5 cm-1 allowed to resolve the manifolds of methane in the 3 μ ν 3 fundamental band.

These early flights permitted to achieve the observation of the vertical distribution of methane (Ackerman et al, 1972) but also led to the first observation of stratospheric NO_2 (Ackerman and Frimout, 1969) and the quantitative determination of its vertical distribution (Ackerman and Muller, 1972). A 1970 spectrum of lower stratospheric nitrogen dioxide, on figure 1, illustrates this early period. These NO_2 results had a large influence on the "Climatic Impact Assessment Program" related to supersonic aircraft : it was proven that an active nitrogen chemistry was already present in the atmosphere before the existence of anthropogenic perturbations. This argument is still correct in 1994 and the conclusions of the new studies of supersonic aircraft impact confirm most of the reports that were issued at the end of the international "Climatic Impact Assessment" study of 1975. In 1972, BISA decided to replace the early Ebert-Fastie instrument by a high resolution grille spectrometer developed in co-operation with the group of Dr. Girard at ONERA (France). With a prototype flown on the Concorde aircraft in 1973 as well as on balloon gondolas nitric oxide could be observed and its vertical distribution was published for the first time (Ackerman et al, 1973, 1974). Later flights confirmed these early observations and gave the first values of the ratio of the ratios of the nitrogen oxides in the stratosphere. In 1975, the grille spectrometer measured the first HCl vertical distribution (Ackerman et al, 1976); almost simultaneously two different American groups obtained similar results. Unfortunately, these observations of hydrogen chloride came too late to determine the pre-industrial stratospheric chlorine value and most modelers have interpreted these results as being consistent with anthropogenic releases of chlorofluoromethanes. The simultaneous observation of methane and hydrogen chloride also led to an indirect determination of chlorine monoxide (Ackerman et al, 1977).

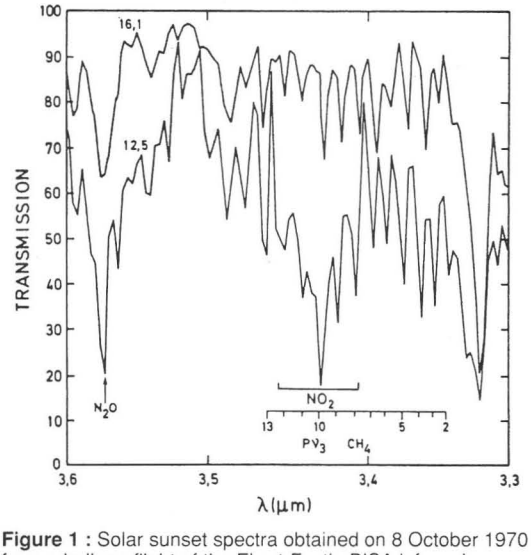

Figure 1 : Solar sunset spectra obtained on 8 October 1970 from a balloon flight of the Ebert-Fastie BISA infrared spectrometer. The flight was performed at the CNES ballooning station of Aire-sur-l'-Adour. The grazing ray altitude is indicated above the spectra, the lower spectrum shows a spectacular enhancement of nitrogen dioxide in the lower stratosphere. Similar phenomena combining chemistry and dynamics are at the core of present theoretical stratospheric research

In 1975, ONERA and BISA decided to start together the study of a space-borne grille spectrometer for the ESA SPACELAB which at that time was in an early stage of development. The design evolved together with SPACELAB, starting from the idea of a simple airborne instrument installed in the pressurised module and ending up

in a pallet instrument driven by a technologically advanced microprocessor control. This model of the grille spectrometer was accepted for flight by ESA in 1977 and developed by Belgian industry in close collaboration with ONERA and BISA.

Simultaneously in the 1980-1982 period a 0.1 wavenumber resolution interferometer was deployed at the "Pic du Midi" in order to study the seasonal and diurnal variation of trace gases. This experiment led to an independent estimate of the atmospheric growth of freon 11 at a time when industry was reporting an important decrease in production (Lippens and Muller, 1981, 1982). In order to complete the series, this observation has been repeated in 1994 from the Caucasian observatory of Seljensjuk; the results are currently processed.

Meanwhile, the SPACELAB instrument was ready for its first flight on the 14 of July in 1980. It was accepted for four demonstration flights and twenty operational flight, but unfortunately, its first flight was in November of 1983 and the second and last happened in March of 1992 during the ATLAS 1 mission. This effectively limited number of flights is due to delays in Space Shuttle operations and also to the ESA ministers decision to give up SPACELAB operations after the first test flight despite its success. The first flight yielded the vertical distributions of 10 atmospheric species over a wide range of latitudes from mid-latitude north to 68° S due to the choice of the launch date near the solstice. In particular, the secondary mesospheric maximum of NO could be observed at high southern latitude while an anti-correlation of water vapour and carbon monoxide was given evidence of. The ATLAS-1 flight, near equinox, led to a much larger set of exploitable experimental data but the zone covered was confined to a southern tropical and mid-latitude belt. The spectral data are still being interpreted; the first published results indicate a growth of HCl in the upper stratosphere since 1985 that is of order 25% at 40 km altitude (De Mazière et al, 1993). The results of this ongoing analysis will constitute an important database that will allow, among others, an evaluation of stratospheric changes in the past and in the future. Figure 2 shows the published HCl profile and compares it to our 1975 first observation of HCl. Moreover, the grille data are accepted for taking part in the UARS (NASA's Upper Atmosphere Research Satellite) correlative program.

In 1988, at the first announcement of opportunity for the ESA polar platform, BISA participated in four proposals : SCIAMACHY, GOMOS, MIPAS and AMAS. The three first instruments by now are accepted for flight on the first ENVISAT payload and AMAS is considered for future platforms. BISA retains scientific participation in these instruments which cover the entire range of atmospheric chemistry by studying radiative atmospheric interactions from the ultra-violet to the microwave.

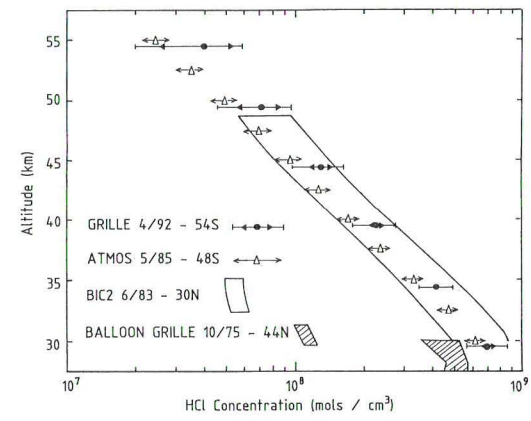

Figure 2 : Hydrogen chloride observations from its discovery in 1975 (Ackerman et al, 1975) to the ATLAS 1 flight of the grille spectrometer (De Mazière et al, 1993), the first observations are still fundamentally correct and a limited growth of stratospheric chlorine appears from these results. The ATMOS and BIC results correspond to the JPL high resolution infrared interferometer and to a joint hydrogen chloride observation campaign performed in 1983. Series of observations together with comparison of results obtained by different techniques will permit to monitor global atmospheric trends in the future

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