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## ROCKETBORNE MEASUREMENT OF MESOSPHERIC H<sub>2</sub>O IN THE AURORAL ZONE

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Abstract. Infrared emission spectra of the longwavelength wing of the 6.3  $\mu$ m water band between 6.7 and 7.6  $\mu$ m have been measured in the mesosphere using a cryogenic rocketborne spectrometer. The resulting zenith radiance profile has been compared with a high altitude radiance model resulting in a volume mixing ratio of  $3.5^{\pm}$  2.2 ppm between 49 and 70 km.

A liquid-helium-cooled, long-wavelength infrared (LWIR) spectrometer was launched by the Air Force Geophysics Laboratory onboard a Black Brant rocket on 22 March 1973 from the University of Alaska's Poker Flat Research Range at Chatanika, Alaska. The spectrometer, which employed a circular-variable filter (CVF), was developed by AFGL and Utah State University (USU) and provided the first measurements of the altitude profile of the infrared spectrum of the upper atmospheric emissions between 7 and 24 µm.

The dominant emission features measured were from the 15  $\mu$ m carbon dioxide ( $\nu_2$ ) band between 65 to 150 km and from the 9.6  $\mu$ m ozone ( $\nu_{3}$ ) band between 45 and 100 km initial results of which were reported by Stair et al., [1974]. Final analysis of the rocket flight has been completed by Rogers et al., [1976] revealing emission data on the long-wavelength wing of the 6.3µm water band between 6.7 and 7.6 µm.

The spectrometer has been described in detail elsewhere [Wyatt, 1975] and so only a brief summary of its salient features will be included here. The spectral region from 6.7 to 24 µm is scanned at a rate of two scans per second with a full-angle field of view of 2 degrees. The entire optical subsection is cooled to liquid-helium temperature, including an arsenicdoped silicon detector which is thereby operated with an effective zero radiation background. A key element of the spectrometer is the Optical Coating Laboratory, Inc., (OCLI) rotating circular variable interference filter. The spectral resolution of the filter was measured at AFGL by Condron [1973] and the best estimate of the half-bandwidth for the flightconfigured spectrometer was 2.63 percent for the short wavelength filter half and 2.92 percent for the long wavelength filter half. A dynamic range of 3x10<sup>4</sup>

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was achieved by providing four data channels with different gains. The spectrometer is the least sensitive at the shorter wavelengths with a noise equivalent spectral radiance (NESR) of 2x10<sup>-10</sup> watt  $cm^{-2}sr^{-1} \mu m^{-1} at 7\mu m$ .

The payload was launched during a post-breakup auroral glow which had persisted at the IBC II level for nearly 10 minutes. Vertical viewing data were obtained during ascent from 96 km to apogee at 185 km while descent data were obtained down to 45 km. The overhead photon radiance at 3914A as measured by an onboard photometer was about 5 KR below 100 km.

Figure 1 shows a typical emission spectrum obtained at 54 km for the short wavelength half of the rotating filter in the spectrometer. The dominant feature at 9.6  $\mu$ m is due to the  $\nu_3$  band of ozone. The emission between 6.7 and 7.6  $\mu$ m has been attributed to the long wavelength wing of the  $\nu_2$  fundamental band of H<sub>2</sub>O. The altitude dependence of these two spectral features from 75 to 50 km is shown in Figure 2 by a three-dimensional presentation of the zenith spectral radiance versus wavelength as a function of decreasing altitude. The 9.6 µm ozone emission has been intentionally drawn off scale in the figure to show the weaker H<sub>2</sub>O emission at the shorter wavelengths.

The altitude dependence of the zenith radiance obtained by integrating the spectral data from 6.7 to 7.6  $\mu$ m is shown by the open boxes in Figure 3. The dashed curves are the ± 30 error bars associated with the spectrometer data. The larger spread in the error bars at the higher altitudes indicates that the spectrometer's lower sensitivity level is being reached.

The solid curve in Figure 3 is a theoretical calculation for the water radiance profile in this wavelength region using the model of Degges [1974]. In this calculation 20% of the total 6.3 µm water band emission lies between 6.7 and 7.6 µm. This estimate was obtained by calculating the spectral bandshape using the line positions and intensities of McClatchey et al., [1973] and integrating to obtain the percentage of resulting emission in the wavelength region of interest. The effects of temperature on this percentage are small since the result varied by only one percent over the range from 200 to 250°K. The primary excitation processes for  $H_2O$  at these

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Fig. 1. Spectral radiance values between 6.7 and 12.4  $\mu$ m during descent at 54 km.

altitudes are the absorption of earthshine or radiation emitted lower in the stratosphere,

$$h\nu + H_0O(000) \rightarrow H_2O(010)$$
;

vibrational energy exchange with molecular oxygen through the near resonant collision mechanism

$$H_2O(010) + O_2(v=0) \neq H_2O(000) + O_2(v=1)$$

and interconversion of thermal and vibrational energy by collision with other molecules,

$$H_2O(010) + M \neq H_0O(000) + M$$

De-excitation of vibrationally excited  $H_2O$  is by spontaneous radiation which occurs with an Einstein A coefficient of 20.3 s<sup>-1</sup> (<u>McClatchey et al.</u>, [1973].) The rate coefficients used for collisional de-excitation are those given by <u>R. L. Taylor</u> [1975], 2.70 x 10<sup>-11</sup>  $T^{-0.5}$  cm<sup>+3</sup> molecule <sup>-1</sup> s <sup>-1</sup> for exchange with O<sub>2</sub> and 4.8 x 10<sup>-11</sup> exp(-43/T<sup>1/3</sup>) cm<sup>3</sup> molecule <sup>-1</sup>s <sup>-1</sup> for quenching by N<sub>2</sub> and CO<sub>2</sub>. Excitation rates are obtained from de-excitation rates by applying detailed balancing. Excitation by exchange of vibrational energy with O<sub>2</sub> is the strongest collisional process and is more effective than absorption of radiation at all altitudes below 75 km. Collisional de-excitation is faster than spontaneous emission at all altitudes below 85 km.

Accurate modeling of the H<sub>2</sub>O emission requires detailed knowledge of the altitude profiles of the atmospheric temperature, density [M] and water concentration  $[H_2O]$ . The calculation shown in Figure 3 used a standard arctic atmosphere with 3.5 ppm by volume of  $H_2O$  and a temperature profile obtained by an extrapolation of sounding rocket temperature measurements made during the night of the spectrometer flight. These temperatures were 216, 222 and 236K at 30, 40 and 50 km respectively (Atmospheric Sciences Laboratory, White Sands Missile Range, New Mexico, private communications). The mesopheric temperatures obtained by this extrapolation introduce a factor of two uncertainties in the theoretical radiance profile over a  $30^{\circ}$  temperature range.

The water concentration used was determined by at least square fit of the calculated to the measured radiance values. The resulting average H<sub>2</sub>O concentration is  $3.5 \pm 1$  ppm by volume between 49 and 70 km. The error bars are increased to  $3.5 \pm 2.2$  by the additional error of a factor of two introduced by the temperature uncertainty.

The mixing ratio of H<sub>2</sub>O has been measured extensively in the stratosphere. <u>Mastenbrook</u> [1968], [1971] has performed a comprehensive stratospheric humidity sounding program carried out with an automatic frost-point sensor and has reported volume



Fig. 2. Altitude dependence of the spectral radiance between 6.7 and 12.4  $\mu$ m during descent from 75 to 50 km. The 9.6  $\mu$ m ozone emission has been intentionally drawn off scale in the figure to show the weaker H<sub>2</sub>O emission at the shorter wavelengths.



Fig. 3. The altitude dependence of the zenith radiance obtained by integrating the spectral data from 6.7 to 7.6  $\mu$ m. The experimental data is shown by the open boxes with the  $\pm$  3 $\sigma$  error bars associated with the spectrometer data shown by the dashed curves. The solid curve is a theoretical calculation based on an arctic model atmosphere with a H<sub>2</sub>O concentration of 3.5 ppm by volume. Twenty percent of the total  $\nu_2$  fundamental band emission occurs between 6.7 and 7.6  $\mu$ m at the temperatures of interest.

mixing ratios for  $H_2O$  from 2-3 ppm between 16 and 22 km. At higher altitudes, <u>Ehhalt et al.</u>, [1975] have used a rocketborne cryogenic air sampler to collect a large air sample between 40 and 50 km and water vapor concentrations of 4.0  $\pm 10.8$  ppm by volume were recovered.

Mesopheric measurements using ground based microwave sounding have recently been reported by <u>Radford et al.</u>, [1977]. They derived a water vapor concentration of 10 ppm at 50 km with a maximum value of 15 ppm at 60 km. As they point out, the results are much higher than expected from the theoretical predictions of <u>Crutzen</u> [1974] which show a maximum H<sub>2</sub>O concentration of 6 ppm at 50 km decreasing to 3-4 ppm at 70 km. The higher theoretical value takes into account the postulated formation of water by the reaction of vibrationally excited H<sub>2</sub> with O (<sup>3</sup> P).

The water concentration measurements mentioned here have all been obtained at mid-latitudes. In a review on water vapor in the stratosphere, <u>Harries</u> [1976] summarizes that there is a trend in the latitudinal distribution of water vapor in the lower stratosphere (below 20 km) showing a maximum near the equator with a decreasing mixing ratio at higher latitudes. However, he concludes from the limited data available above 20 km at higher latitudes, that there is no clear evidence for a rise over the tropics as occurs for lower altitudes.

A volume mixing ratio of  $3.5 \pm 2.2$  ppm for H<sub>2</sub>O between 49 and 70 km reported in this paper supports the predictions of atmospheric models such as <u>Crutzen</u> [1974] which show mesopheric water in concentrations from 2 to 6 ppm up to the 75-85 km altitude region where H<sub>2</sub>O is rapidly photodissociated.

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