A PROMISING TECHNIQUE TO MEASURE WATER VAPOR CONTENT IN THE POLAR ATMOSPHERE: LYMAN-α/OH FLUORESCENCE HYGROMETER

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Abstract: For monitoring the water vapor content in the polar troposphere and stratosphere a Lyman–α/OH fluorescence hygrometer is very useful owing to its rapid response, high sensitivity and wide dynamic range. Cooperative measurement of Lα/OH fluorescence hygrometry with other monitoring techniques of atmospheric water vapor such as satellite, Raman scattering lidar, balloon-borne carbon humidity sensor and so on will produce abundant results which help understand the water vapor concentration in the polar atmosphere.

1. Introduction

Measurement of water vapor concentration in the polar atmosphere is one of the important tasks for study of polar atmospheric physics since (1) water vapor play an important role in the radiative energy transfer in the atmosphere, and information on its distribution is indispensable in the study of the energy budget of the polar atmosphere, and (2) the polar stratosphere is possibly an important sink of water vapor, and water vapor measurement in the polar stratosphere can help understand the global budget of stratospheric water vapor.

The model proposed by BREWER (1949) still has a large influence on the

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studies of the transport process of minor constituents in the stratosphere. The essence of his model is that there will be "a circulation in which air enters the stratosphere at the equator, where it is dried by condensation, travels in the stratosphere to temperate and polar regions, and sinks into the troposphere (BREWER, 1949)". The dryness of the stratosphere is attributed to "the cold trap" at the equatorial region in this model. Independently, STANFORD (1973, 1977) proposed a theory that stratospheric clouds frequently reported in the winter Antarctic Continent will be a possible sink of stratospheric water vapor on a global scale. IWASAKA (1981) also reported that this is a possible mechanism of extraction of water vapor in the height range from the stratosphere into the troposphere.

There have been few satisfactory measurements of water vapor content in the Antarctic atmosphere though this is a very important parameter. This is because the absolute value of water vapor concentration in the Antarctic atmosphere is very low due to low atmospheric temperature, so that it is difficult to obtain accurate data in humidity measurements with the use of conventional humidity sensors.

We already reported on the humidity measurement technique using the fluorescence effect of the excited state OH radicals produced through the photodissociation of H_2O by the Lyman-α line (121.6 nm) (IWASAKA et al., 1981). This type of humidity sensor, called the Lα/OH fluorescence hygrometer, has the following advantages:

(1) fast response (probably on the order of 10 ms at a water vapor content of 1000 ppmV),

(2) high sensitivity,

(3) wide dynamic range (useful in the troposphere and the stratosphere).

Such advantages must be very useful in various scientific fields. It is indispensable to know the rapid variations of water vapor content in the atmosphere for studies on boundary-layer meteorology, agricultural meteorology, air-sea interaction processes and so on. High sensitivity of instrument is always desired when we observe water vapor concentration in a very dry atmosphere such as the stratosphere and
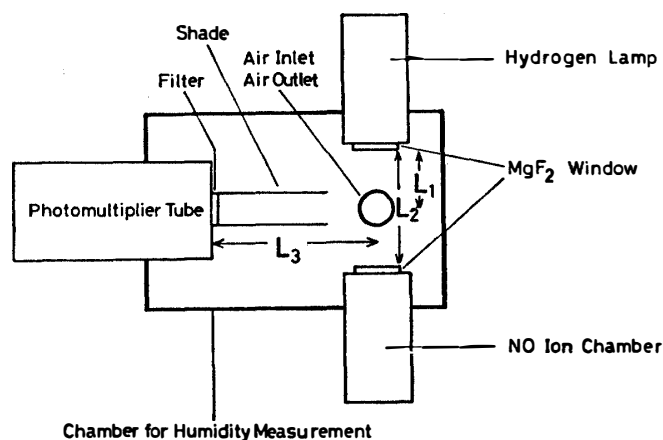

Fig. 1. Configuration of Lα/OH fluorescence hygrometer. The nitric oxide ion chamber and the photomultiplier (PMT) are used to detect the attenuation of the Lα line intensity and to monitor the fluorescence intensity from $OH(A^2\Sigma^+)$.

the polar atmosphere.

In this paper we would like to show the usefulness of the Lα/OH fluorescence hygrometer in the water vapor measurement of the polar atmosphere.

2. Hygrometry Using Fluorescence from OH($A^2\Sigma^+$)

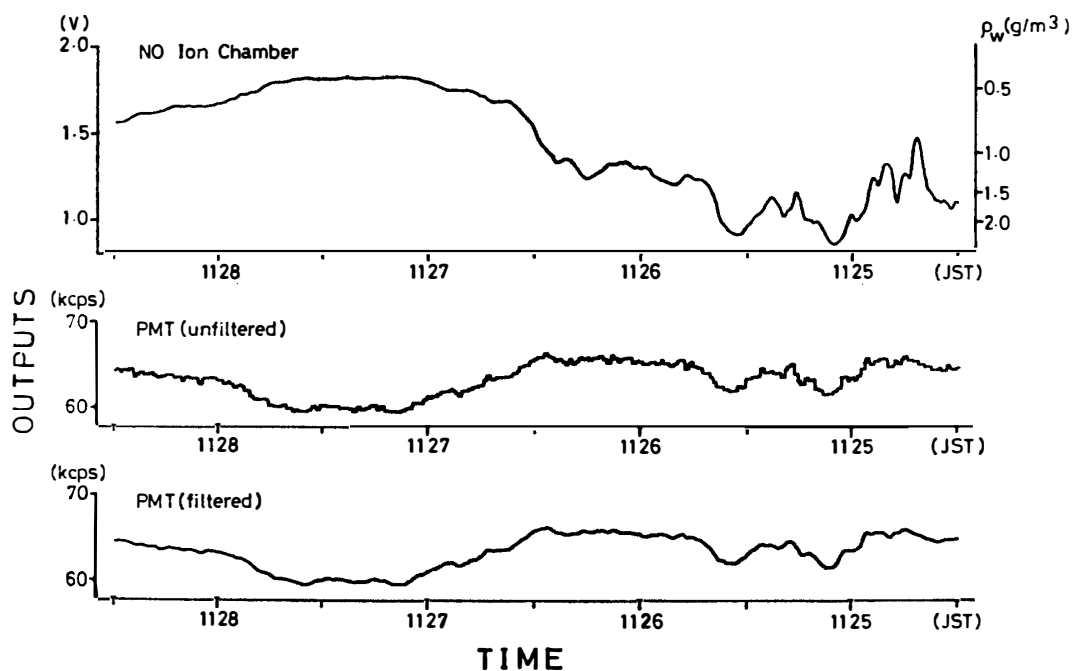In the Lα/OH fluorescence hygrometer, the following chemical reactions are used:

Fig. 2. Results of an aircraft measurement which was made to test the efficiency of the Lα/OH fluorescence hygrometer.
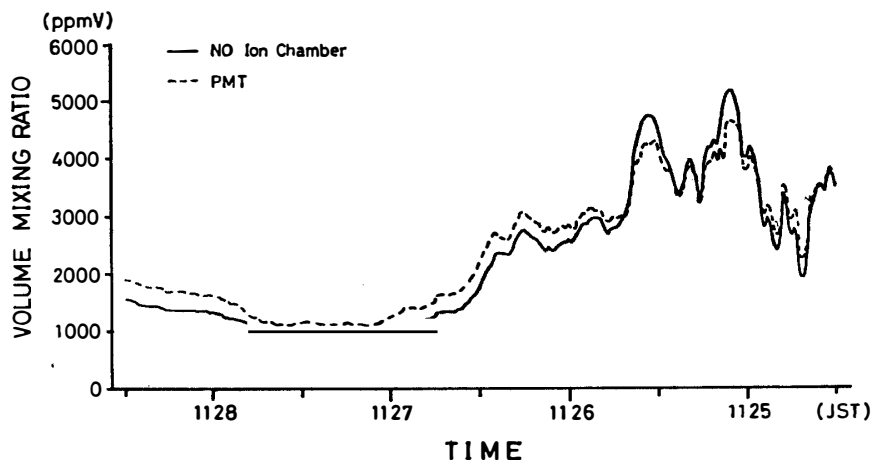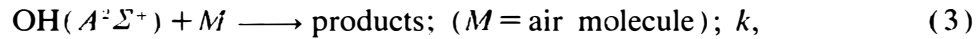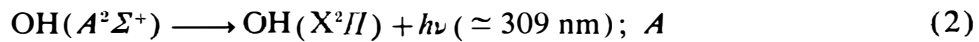

Fig. 3. Comparison of the output power from the ion chamber with the correction value of the PMT output which is obtained by a data reduction procedure (see MURABAYASHI et al., 1982). Both curves show good agreement, and suggest that the output power from the PMT is adequate for the lower atmosphere.

$$H_2O + \text{Lyman-}\alpha \longrightarrow (121.6 \text{ nm}) \quad OH(A^2\Sigma^+) + H(^2S); \quad J \qquad (1)$$

$$OH(A^2\Sigma^+) \longrightarrow OH(X^2\Pi) + h\nu \ (\simeq 309 \text{ nm}); \quad A \qquad (2)$$

$$OH(A^2\Sigma^+) + M \longrightarrow \text{products}; \quad (M = \text{air molecule}); \quad k, \qquad (3)$$

where

J : photodissociation rate,

A : transition probability,

k : deactivation rate constant.

As the value of ($A + k$ $[M]$; $[M]$ means number density of M) is for greater than 10^{-6}s^{-1}, OH radicals in an excited state can be considered to be in chemical equilibrium with water vapor molecules on a time scale greater than 10^{-6} s. Therefore the intensity of fluorescent light $OH(A^2\Sigma^+)$ can be expressed in the following form in the troposphere and the stratosphere;

$$I = [OH(A^2\Sigma^+)]A$$
$$= JeA(1/k)_\mu \qquad (4)$$

where

e : quantum efficiency of reaction (1),

μ : volume mixing ratio of H_2O ($= [H_2O]/[M]$).

From eq.(4), the mixing ratio of water vapor can be obtained by measuring the intensity of fluorescent light from OH radicals produced in the photodissociation of H_2O. In addition to this, the attenuation of Lα light intensity due to the absorption by H_2O (reaction (1)) is also used to monitor the concentration of H_2O molecules. The details of the instrument have already been described in other papers (IWASAKA et al., 1981; MURABAYASHI et al., 1982), and we will not describe them here. The configuration of the instrument is schematically shown in Fig. 1.

An example of the output signal of the instrument is shown in Figs. 2 and 3. Curve a in Fig. 2 shows logarithmically amplified output from a nitric oxide ion chamber (measurement of the attenuation effect of Lα light). Curve b of Fig. 2 shows the direct output from the photon counter (measurement of the fluorescence from the excited state OH radical). Curve c in Fig. 2 shows output of the photon-counter smoothed by a low pass filter having a cut-off frequency of 0.2 Hz. And the output of the instrument which was obtained through the data reduction procedure is compared with the signal from the ion chamber in Fig. 3 (the data reduction procedure was described in the paper by MURABAYASHI et al., 1982). These results apparently show the advantages of this type hygrometer.

3. Water Vapor Concentration in the Antarctic Atmosphere

Figure 4 shows the monthly humidity variation range observed at Mizuho Station from March to December 1979 (WADA et al., 1981). The humidity sensor used here was an alminum oxide hygrometer manufactured by Parametric Co., Ltd., U.S.A. The minimum value of the volume mixing ratio of water vapor was observed in June, and it corresponds to winter season of the Antarctic Continent. Monthly minimum values of the volume mixing ratio of water vapor gradually increased as the season proceeded from winter to spring and to summer. The seasonal varia-
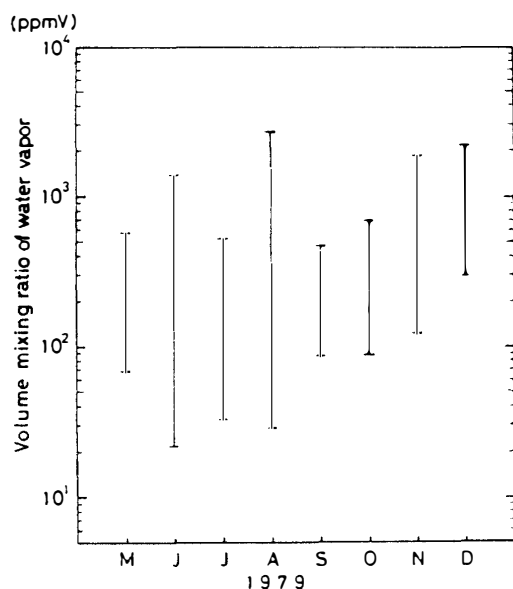
Fig. 4. Monthly data for the humidity variation ranges observed at Mizuho Station, Antarctica in 1979 (WADA et al., 1981).
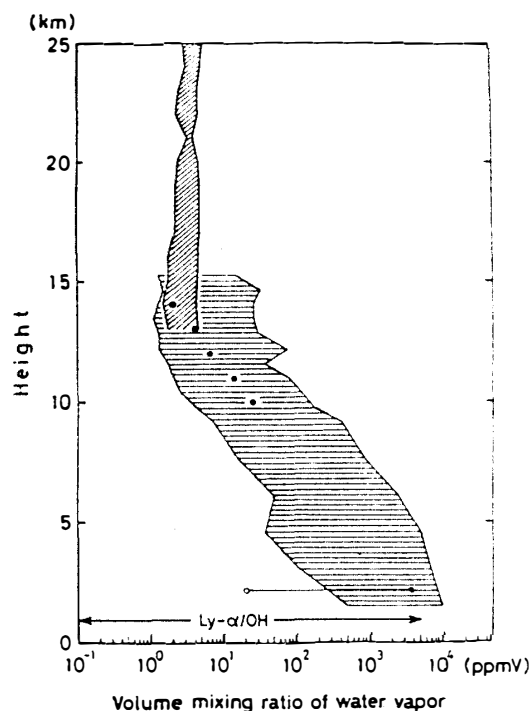

Volume mixing ratio of water vapor

Fig. 5. Vertical distribution of water vapor concentration. The slant hatching region is the result by HARRIES (1976) (30°–58°N), horizontal hatching by MURGATROYD et al. (1955) and HELLIWELL et al. (1975) (Southern England), full circles by MURGATROYD (1965), and open circles by WADA et al. (1981). (Mizuho Station on the Antarctic Continent). Arrows indicate the measurable range of the Lα/OH fluorescence hygrometer described here.

tion of the monthly maximum value also showed a similar tendency although the maximum value was also observed in summer. The volume mixing ratio of water vapor covered three orders in humidity variation range, and the minimum value was as low as 20 ppmV. This means that it is difficult to observe water vapor concentration on the Antarctic Continent by using conventional type humidity sensors such as a psychrometer.

In the upper-air observations made at Syowa Station, carbon humidity sensors have been used to measure water vapor concentration. However, its measurable range is insufficient to obtain a vertical distribution of water vapor concentration up to the stratosphere, and we cannot obtain information about the water vapor transport in the stratosphere at present. Hatched areas shown in Fig. 5 are the variability of water vapor concentration obtained by HARRIES (1976), HELLIWELL et al. (1975), and MURGATROYD et al. (1955) in the mid-latitude regions of the Northern Hemisphere.

These areas indicate that the water vapor concentration gradually decreases with height and that the stratosphere is very dry, as dry as several ppmV. Again if one desires to measure water vapor concentration vertically from the the earth's surface to the stratosphere, he should use a humidity sensor having a measurable range as wide as five orders of magnitude. Full circles in Fig. 5 are the results obtained by MURGATROYD (1965) at 70°S. Open circles are the same results as Fig. 4 obtained by WADA et al. (1981) on the Antarctic Continent. These circles imply that the air over the Antarctic Continent is drier than that in the mid-latitude regions of the Northern Hemisphere. HARRIES (1976) stated in his review work that there is a tendency of decrease of water vapor concentration toward higher latitude in both hemispheres in the lower stratosphere. These observational results may support the idea that the Antarctic winter is an important sink for stratospheric water vapor (STANFORD, 1973, 1977; IWASAKA, 1981).
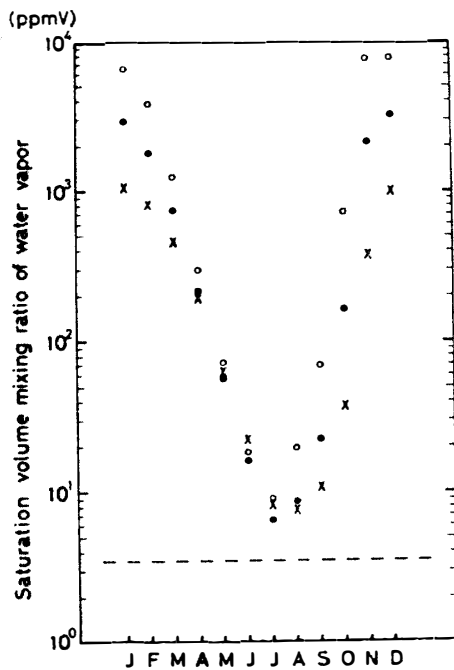

Fig. 6. Monthly values of saturation volume mixing ratio of water vapor with respect to ice for levels of 30 mb (●), 50 mb (○), and 100 mb (×) over Syowa Station on the Antarctic Continent. The horizontal line is the volume mixing ratio of water vapor at 20 km according to HARRIES (1976).

In order to check this possibility of a water vapor sink in the Antarctic stratosphere, we illustrated in Fig. 6 the monthly values of saturation volume mixing ratio of water vapor with respect to ice for levels of 30, 50 and 100 mb over Syowa Station (JAPAN METEOROLOGICAL AGENCY, 1981). Annual mean heights of individual pressure levels are 22764, 19566 and 15240 gpm respectively. In the winter season, the saturation volume mixing ratios of water vapor at individual levels become very low values, less than 10 ppmV. However, the values for a height of 20 km in the mid-latitude regions of the Northern Hemisphere obtained by HARRIES (1976) as shown by the broken line in Fig. 6 are still smaller than these winter saturation values over Syowa Station. Therefore, precipitation may not take place in the stratosphere over Syowa Station even in the winter season. However, as shown in the work by STANFORD (1973), the temperature at the

50 mb level becomes as low as $-90°C$ at the South Pole in winter. This means that it is required to undertake a precise observation of water vapor concentration in the Antarctic stratosphere in order to examine the theory proposed by STANFORD (1973, 1977) and IWASAKA (1981).

4. Discussion and Conclusion

The use of a Lα/OH fluorescence hygrometer will certainly make an important contribution toward understanding the water vapor content in the polar atmosphere and its role in the study of the physics of the polar atmosphere. There are many plans to monitor the water vapor content of the polar troposphere and stratosphere during the Japanese Middle Atmosphere Program (MAP). Of course, a comparison of Lα/OH fluorescence hygrometry with other different techniques is also desired to establish the accuracy of the instruments, and to complement defects of these instruments.

The Raman scattering lidar and satellite are used to monitor the water vapor content of the polar atmosphere in addition to the balloon-borne carbon humidity sensor which has been used to detect the water vapor concentration of the polar troposphere.

The basic formula for the scattered power for a Raman lidar is given by:

$$P(\nu_2, z) = P_0(\nu_1)\{KT_1(z)T_2(z)\beta_{12}\}/z^2 , \qquad (5)$$

where

$P(\nu_2, z)$: received power at wavelength ν_2,

z : vertical height,

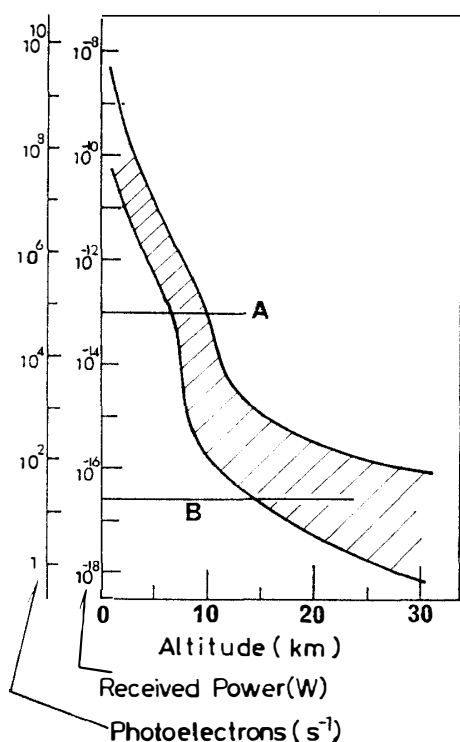$P_0(\nu_1)$: output power of laser pulse,

Fig. 7. *The shaded area shows the received power estimated for the various water vapor contents. Lines A and B show the noise level of the night sky and the detection limit assuming a S/N level of 3, respectively.*

T_1, T_2 : atmospheric transmission at wavelengths ν_1 and ν_2 respectively,

β_{12} : backscattering coefficient of the Raman scattering of water vapor molecule,

K : system constant.

The Raman backscattering coefficient is defined by:

$$\beta_{12}(z) = [H_2O(z)]\left(\frac{d\sigma}{d\Omega}\right)_{Ram},\qquad(6)$$

where

$[H_2O(z)]$: water vapor density at altitude z,

$(d\sigma/d\Omega)_{Ram}$: Raman differential backscattering cross section.

Figure 7 shows the height range where the Raman scattering lidar can detect the atmospheric water vapor. The shaded area indicated the range of the backscattered power which is estimated for the various water vapor contents summarized by HARRIES (1976) under the condition that output energy power of laser pulse = 0.8 J/pulse (wavelength = 694.3 nm; Ruby laser), receiving telescope diameter = 70 cm, quantum efficiency of photomultiplier = 0.4 (wavelength = 347.2 nm, in this lidar system the second harmonic wave of Ruby laser is considered), $K = 1$, and $(d\sigma/d\Omega)_{Ram} = 9.5 \times 10^{-30}$ cm^2 sr^{-1} (MELFI, 1972).

It can easily be found that the Raman lidar technique is very powerful in remote measurement of the water vapor content in the troposphere, but this technique is not suitable for the monitoring of the water vapor concentration above 10 km height.

The estimation of the vertical profile of the water vapor content on the basis of the radiance from the atmospheric water vapor infrared emission (8.30–6.70 μm) measured by the polar orbital meteorological satellite can be made by so-called "inversion method" (SMITH, 1968). However, in practice, the monitoring of the water vapor concentration is not done this way, but by a statistical method, the so-called "regression method" in which the humidity data obtained by radio sonde are essential to determine the regression coefficients (AOKI et al., 1981). Hence, the humidity estimated from the satellite is dependent on the accuracy and quality of the measurements of the radio sonde (HAYDEN et al., 1981). The balloon measurements of water vapor using the Lα/OH fluorescence hygrometer certainly will provide valuable information for satellite water vapor measurements.

Acknowledgments

References

AOKI, T., NAKAJIMA, S. and KATO, K. (1981): Kyoku-kidô kishô êsei (TIROS-N) dêta shori shisutemu (Data processing system for TIROS-N satellite series). Tenki, **28**, 531–542.

BREWER, A. W. (1949): Evidence for a world circulation provided by the measurements of helium and water vapour distribution in the stratosphere. Q. J. R. Meteorol. Soc., **75**, 351–363.

HARRIS, J. E. (1976): The distribution of water vapor in the stratosphere. Rev. Geophys. Space Phys., **14**, 565–575.

HAYDEN, C. M., SMITH, W. L. and WOOLF, H. M. (1981): Determination of moisture from NOAA polar orbiting satellite sounding radiance. J. Appl. Meteorol., **20**, 450–466.

HELLIWELL, N. C., MACKENZIE, J. K. and KERLEY, M. J. (1975): Some further observations from aircraft of frost point and temperature up to 50,000 ft. Q. J. R. Meteorol. Soc., **83**, 257–262.

IWASAKA, Y. (1981): Stratospheric "cist" and water vapor budget in the stratosphere. Mem. Natl Inst. Polar Res., Spec. Issue, **19**, 188–194.

IWASAKA, Y., MURABAYASHI, S. and YAMASHITA, Y. (1981): A new method to measure water vapor content of the polar middle atmosphere using the photochemical reaction "$H_2O + h\nu$ (121.6 nm) → $OH^* + H$". Mem. Natl Inst. Polar Res., Spec. Issue, **19**, 195–209.

JAPAN METEOROLOGICAL AGENCY (1981): Climatological data at Syowa Station, Antarctica. Nankyoku Shiryô (Antarct. Rec.), **73**, 273–303.

MELFI, S. H. (1972): Remote measurements of the atmosphere using Raman scattering. Appl. Opt., **7**, 1605–1610.

MURABAYASHI, S., IWASAKA, Y. and YAMASHITA, Y. (1982): A note on the new type hygrometer using the photochemical reactions: $H_2O + h\nu$ (Ly-α line) → $OH^* + H$, and OH^* → $OH + h\nu$ ($\lambda = 309$ nm). Mem. Natl Inst. Polar Res., Spec., Issue, **22**, 239–246.

MURGATROYD, R. J. (1965): Ozone and water vapor in the upper troposphere and lower stratosphere. Meteorological Aspects of Atmospheric Radioactivity, ed. by W. BLEEKR. Geneva, World Meteorol. Organ., 68–94, (WMO Tech. Note, 68).

MURGATROYD, R. J., GOLDSMITH, P. and HOLLINGS, W. E. H. (1955): Some recent measurements of humidity from aircraft up to height of about 50,000 ft over southern England. Q. J. R. Meteorol. Soc., **81**, 533–537.

SMITH, W. L. (1968): An improved method for calculating tropospheric temperature and moisture from satellite radiometer measurements. Mon. Weather Rev., **96**, 387–396.

STANFORD, J. L. (1973): Possible sink for stratospheric water vapor at the winter Antarctic Pole. J. Atmos. Sci., **30**, 1431–1436.

STANFORD, J. L. (1977): On the nature of persistent stratospheric clouds in the Antarctic. Tellus, **29**, 530–534.

WADA, M., YAMANOUCHI, T., MAE, S., KAWAGUCHI, S. and KUSUNOKI, K. (1981): POLEX South data, Part 2. Micrometeorological data at Mizuho Station, Antarctica in 1979. JARE Data Rep., **62** (Meteorol. 9), 321 p.