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# Observation of HD<sup>18</sup>O, CH<sub>3</sub>OH and vibrationally-excited N<sub>2</sub>O from Odin/SMR measurements

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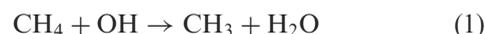
The sub-millimetre radiometer (SMR) instrument aboard the Odin satellite launched in February 2001 is dedicated to the observation of trace gas profiles in the stratosphere and in the mesosphere, in particular water vapour and its isotopic species HDO, H<sub>2</sub><sup>17</sup>O and H<sub>2</sub><sup>18</sup>O. By averaging more than 400 000 Odin/SMR spectra over the period from November 2002 to March 2003, weak lines have been detected in the atmosphere. These weak lines have been attributed to water isotopic species HD<sup>18</sup>O, to a vibrationally-excited N<sub>2</sub>O, and to CH<sub>3</sub>OH.

## 1. Introduction

Water vapour is a good tracer for studying dynamics in the stratosphere due to its long lifetime [1] and also because the temperature at the tropopause controls the flow of water vapour into the stratosphere [2, 3]. Less abundant H<sub>2</sub>O isotopic species, namely HDO, H<sub>2</sub><sup>17</sup>O and H<sub>2</sub><sup>18</sup>O, can be used to study the dynamics in the middle and upper atmosphere in the same way as the main isotope. The isotopic ratio, i.e. the amount of heavy isotopic species compared to the main isotope, determines the origin of an air mass. The main mechanism controlling the isotopic fractionation of water vapour in the troposphere is the lower vapour pressure of the heavier isotopic species. A low deuterium to hydrogen (D/H) ratio will therefore indicate that the air parcel has its origin in the troposphere, while a high ratio implies H<sub>2</sub>O to be formed in the stratosphere by oxidation of methane [4].

Methane represents one of possible sources of water molecules in the stratosphere. Methane is produced only

at the surface of the Earth by anaerobic processes and biomass burning. In the troposphere, methane is a relatively long-lived species, its lifetime is greater than one year. Methane comes into the stratosphere – probably in the tropics – in the same way as water vapour. In the stratosphere, the reaction with OH dominates and causes the loss of methane:



The reaction with O(<sup>1</sup>D) is also significant at around the altitude of 30 km and starts to be more important than reaction (1) above 50 km. This reaction leads mainly to CH<sub>3</sub> production but both formaldehyde (CH<sub>2</sub>O) and molecular hydrogen (H<sub>2</sub>) can be generated in such a way.

Methanol is the second most abundant organic trace gas in the Earth's atmosphere after methane. Measurements of methanol are performed on the base of ground, airborne, and satellite instruments [5, 6].

The knowledge of the stratospheric distribution is based on very few measurements of isotopic water in the stratosphere, which are very limited in time and space. Recent measurements of stratospheric HDO, H<sub>2</sub><sup>18</sup>O and

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also  $\text{H}_2^{17}\text{O}$  have become to be available. There are thus only a few data sets in existence and they are usually limited in time and space. The ATMOS instrument [7–9] measured isotopic species of water vapour and methane with a rather large latitudinal coverage. The problem is that the measurements were performed on different occasions so that there is no real global coverage available. It has flown four times on the Space Shuttle since 1985, first on Spacelab 3 in April 1985 and then on ATLAS 1, 2 and 3 shuttles in March 1992, April 1993 and November 1994, respectively. ATMOS measurements include, among other gases,  $\text{H}_2\text{O}$ , HDO,  $\text{H}_2^{18}\text{O}$ ,  $\text{H}_2^{17}\text{O}$ ,  $\text{CH}_3\text{D}$  and  $\text{CH}_4$ . Results show that there is a large depletion in HDO in the upper troposphere and lower stratosphere [9, 10]. The decrease in the D depletion is slightly smaller than expected on the base of theories described above. This discrepancy can be explained by the fact that the production rate of HDO from oxidation of  $\text{CH}_3\text{D}$  is slower than the corresponding production of  $\text{H}_2\text{O}$  from  $\text{CH}_4$ . Irion *et al.* [9] found, using ATMOS data, that the production of HDO is about 20% slower than the production of  $\text{H}_2\text{O}$ . Measurements of isotopic water vapour have been performed from balloon-borne platforms in the far infrared spectral region [11, 12] and new measurements are, therefore, of a growing importance.

The data from ODIN satellite launched in February 2001 offer a new tool for studying various processes in the stratosphere by determination of the isotopic ratio of water vapour. In this paper, we first show zonal means of water vapour ( $\text{H}_2\text{O}$ ) and its main isotopic species (HDO,  $\text{H}_2^{18}\text{O}$ ). We present the methodology used in order to detect very weak lines by averaging of sub-millimetre radiometer (SMR) spectra in order to increase the signal-to-noise ratio. This technique, which employs a millimetre-wave superheterodyne receiver and multichannel filter spectrometer, was previously used to observe rotational transitions of stratospheric molecules as chlorine monoxide and also for observations of other stratospheric trace gases such as  $(^{16}\text{O})_3$ , vibrationally excited  $(^{16}\text{O})_3$ ,  $(^{18}\text{O})_2(^{16}\text{O})$ ,  $\text{N}_2\text{O}$ ,  $\text{HO}_2$ , and  $\text{HCN}$  [13]. The SMR on the Odin satellite observes species as  $\text{O}_3$ ,  $\text{ClO}$ ,  $\text{N}_2\text{O}$ , and  $\text{HNO}_3$  using two bands centred at 501.8 and 544.6 GHz [14].

## 2. The Odin data

The Odin mini-satellite is a Swedish-led project funded jointly by Sweden, Canada, France, and Finland [15, 16]. It was placed into a 600 km Sun-synchronous, terminator orbit by a START-1 rocket on 20 February 2001, and is still operational. Odin includes two instruments. The first one is a UV-visible and infrared

instrument called Optical Spectrograph and Infrared Imager System (OSIRIS) and enabling detection of  $\text{O}_3$ ,  $\text{NO}_2$ , and aerosol particles in the scattered sunlight ranging from wavelength 280 nm up to 800 nm. The related oxygen airglow emission at  $1.27\ \mu\text{m}$  [17] is also measurable. The second instrument is the SMR [15], which can measure  $\text{O}_3$ ,  $\text{ClO}$ ,  $\text{N}_2\text{O}$ ,  $\text{HNO}_3$ ,  $\text{H}_2\text{O}$  and isotopic species, and  $\text{CO}$  in the frequency domain 480–580 GHz.

In the spectral range of the SMR (about 450–600 GHz) there are two  $\text{H}_2\text{O}$  lines, two  $\text{H}_2^{18}\text{O}$  lines and one HDO line [16], that can be used for studies of stratosphere–troposphere exchange and different mixing mechanisms. The two  $\text{H}_2\text{O}$  lines are located at 488.491 and 556.936 GHz. For studies of the stratosphere the line at 488 GHz is the most suitable due to its moderate line strength [16]. The second line, at 557 GHz is very strong and is more suitable for mesospheric studies.  $\text{H}_2^{18}\text{O}$  follows the same pattern with one strong line at 547.676 GHz and a weaker one at 489.054 GHz. Again the weaker line is the best to use for studies of the stratosphere but the stronger line is also useful [16]. The HDO line, located at 490.596 GHz is of moderate strength and is suitable for studies in the stratosphere [16].

## 3. Results and discussion

The origins of air parcels can be deduced by observation of the deuterium to hydrogen ratio (HDO/ $\text{H}_2\text{O}$ ). The water vapour produced by methane oxidation in the stratosphere will have a different isotopic ratio from tropospheric water vapour. Results of water vapour and its isotopes measurements are depicted in figure 1. Figure 1 shows zonal means of water vapour isotopic species as measured in the bands centred at 488.9 GHz ( $\text{H}_2\text{O}$  and  $\text{H}_2^{18}\text{O}$ ) and at 490.4 GHz (HDO) by Odin/SMR on 15 March 2002. The three fields show a tropical dehydrated lower stratosphere induced by the troposphere to stratosphere intrusion of air masses, and an increase in upper stratosphere by methane oxidation. At high latitudes in the southern hemisphere, the upper stratosphere–lower mesosphere is wetter than at other latitudes. This feature is induced by the meridional mean circulation.

In the present analysis, Odin/SMR spectra have been used from the FM01 band (dedicated to the measurement of  $\text{ClO}$  and  $\text{O}_3$ ) i.e. from 501.16939 to 501.59139 GHz. Measurements from November 2002 to March 2003 have been averaged within 5 km vertical layers from 0 to 100 km in order to increase the signal-to-noise ratio for the detection of very weak lines. We first concentrated on the tropical band from  $30^\circ\text{S}$  to  $30^\circ\text{N}$  and

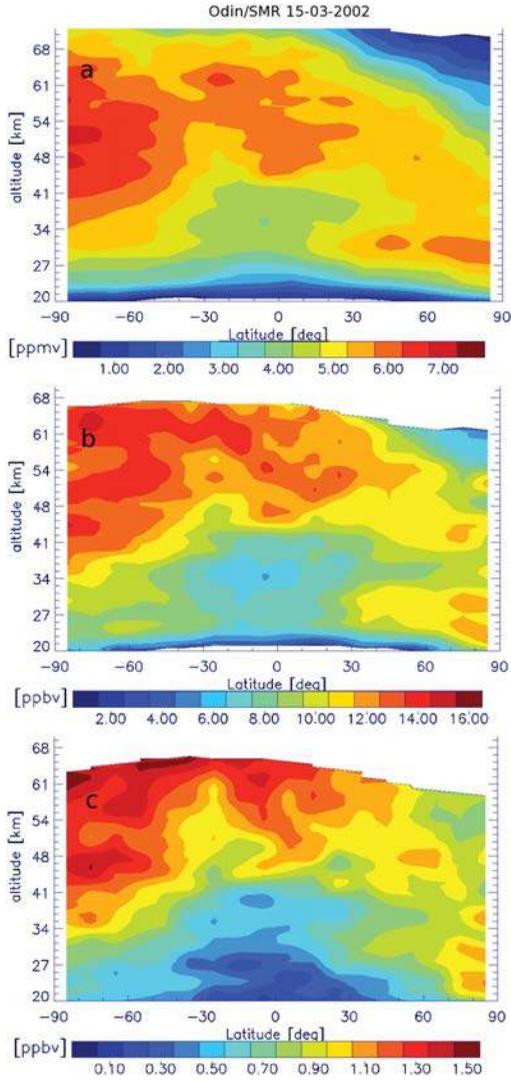


Figure 1. Latitude–height cross-sections of various water vapour isotopic species as measured by the Odin/SMR instrument on 15 March 2002: (a)  $\text{H}_2\text{O}$ , (b)  $\text{H}_2^{18}\text{O}$ , and (c) HDO.

then on the northern and southern hemispheres, and finally on the whole Earth. The number of averaged spectra in particular vertical layers is documented in table 1.

Figure 2 shows measured spectra averaged within different 5 km layers from 2.5 to 52 km. Both the  $\text{O}_3$  and  $\text{ClO}$  lines are visible in all the altitude layers above 17.5 km. The lower panel shows in the frequency micro-windows, namely 501.52–501.59 GHz, few very weak lines. Indeed, several structures in the selected micro-window are present in the averaged measured spectra (figure 3). A second-derivative Pearson function was used to fit the strongest lines and their associated

Table 1. Number of spectra averaged within the 5 km deep vertical layers.

Altitude layer/km	Northern hemisphere	Southern hemisphere	Tropics	Global
2.5	1184	1324	1160	3668
7.5	15153	16042	13993	45188
12.5	16247	16048	16280	48575
17.5	16460	16346	16624	49430
22.5	16534	16409	16546	49489
27.5	16499	17131	17140	50770
32.5	16548	16389	16593	49530
37.5	16641	16494	16891	50026
42.5	16458	16371	16567	49396
47.5	12364	13098	12371	37833
52.5	5762	5934	5653	17349
57.5	5588	5421	5550	16559
62.5	5597	5409	5503	16509
67.5	3914	4554	3607	12075

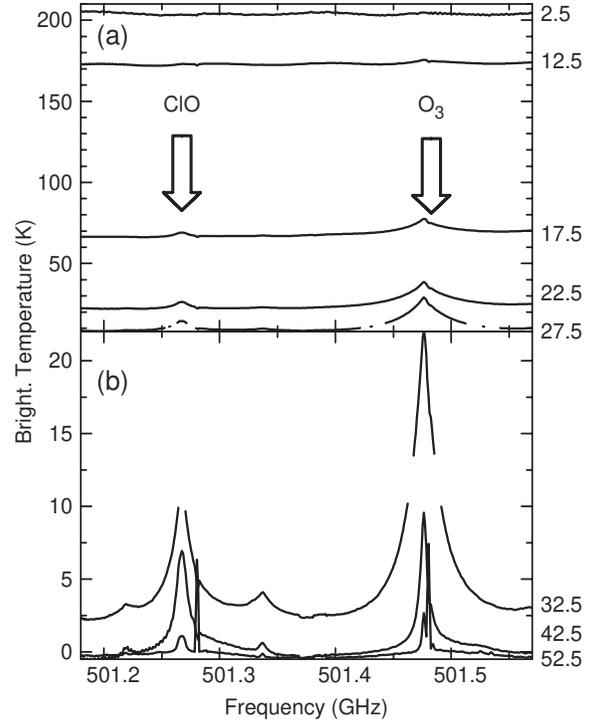


Figure 2. Average of more than 400 000 sub-millimetre wave spectra measured by the Odin/SMR instrument in different altitude layers: (a) 2.5, 12.5, 17.5, 22.5, 27.5, and (b) 32.5, 42.5 and 52.5 km.

continua within and around this particular micro-window (figure 3). The averaged spectra from 17.5 to 67.5 km and the theoretical fits, are displayed in figure 4 for different latitudinal bands, namely in the southern

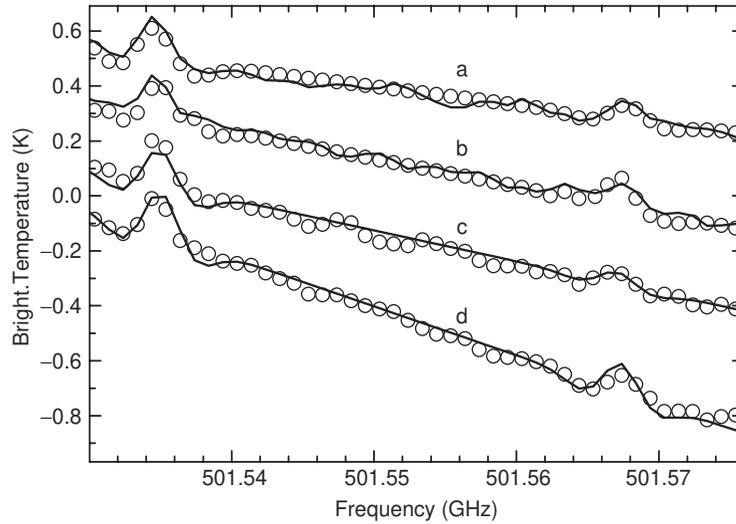


Figure 3. Averaged spectra over the 501.53–501.58 GHz micro-window superimposed on a second-derivative Pearson function fitting the strongest lines and their associated continua within and around this particular micro-window: (a) 52.5, (b) 57.5, (c) 62.5 and (d) 67.5 km, curves (a) and (b) are offset.

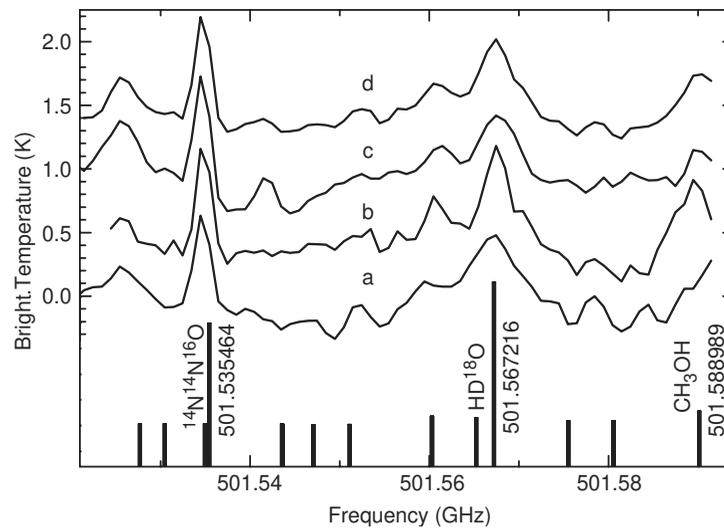


Figure 4. Residuals of sub-millimetre wave spectra over the 501.52–501.59 GHz micro-window minus modelled fit: (a) southern hemisphere, lat.  $< 30^{\circ}\text{S}$ ; (b) global,  $90^{\circ}\text{S} < \text{lat.} < 90^{\circ}\text{N}$ ; (c) northern hemisphere, lat.  $> 30^{\circ}\text{N}$ ; (d) tropics,  $30^{\circ}\text{S} < \text{lat.} < 30^{\circ}\text{N}$ . Data were averaged over the range 17.5–67.5 km and curves (a), (b) and (c) are offset. Superimposed are the theoretical lines taken from [20] (vertical bars).

hemisphere mid- and high latitudes ( $< 30^{\circ}\text{S}$ ), northern hemisphere mid- and high latitudes ( $> 30^{\circ}\text{N}$ ), tropical latitudes ( $30^{\circ}\text{S} - 30^{\circ}\text{N}$ ), and global latitudes.

In all latitudinal bands, three lines are well above the noise level located at 501.53439, 501.56739 and 501.59039 GHz. From spectroscopic databases, they can be attributable to:  $\text{N}_2\text{O}$  in vibrationally-excited state ( $E_{\text{vib}} \cong 1800 \text{ cm}^{-1}$ )  $20 - 19 (1, 1^{\text{d}}, 0)$  (501.535464 GHz) [18, 19], to ground state  $\text{HD}^{18}\text{O}$  ( $J', K'_{-1}, K'_{+1} - J'', K''_{-1}, K''_{+1}$ : 1, 1, 0–1, 0, 1) [20] (501.567216 GHz [21]), and to ground state of  $\text{CH}_3\text{OH}$  ( $J', K'_a, K'_c - J'', K''_a, K''_c$ : 9, 2,

8–9, 1, 9) (501.588989 GHz [22, 23]), respectively. Spectral resolution of the ODIN radiometer is in the range 0.1–1 MHz and because we have processed very small signals we could expect the higher level of frequency uncertainties about 1 MHz. We have carefully checked that there were no strong lines in the image sideband that could contaminate the signal sideband.

A very important part of the future work is to include the less abundant species and isotopes of water vapour in the chemical reaction scheme of a more sophisticated model. Each detected line could contribute to this

research. Simultaneously, investigation of deuterium and  $^{18}\text{O}$  with the help of  $\text{HD}^{18}\text{O}$  could contribute to the following research. It is for example, such studies as the latitudinal behavior of isotopic ratios, isotope exchange between water and molecular hydrogen and the  $[\text{O}^{18}]/[\text{O}]$  ratio of stratospheric water vapor affected by O-atom exchange with the oxygen reservoir, initiated by the reaction with  $\text{O}(^1\text{D})$ .

A simple calculation can be done in order to investigate what accuracy we need to be able to resolve the depletion of deuterium and of  $^{18}\text{O}$ . Isotopic compositions of two samples are usually expressed using delta notation, in which the isotopic ratios (for example deuterium  $D$  and hydrogen  $H$  species) of the sample and reference are related by:

$$\delta D = 1000 \left( \frac{(D/H)_{\text{sample}}}{(D/H)_{\text{ref}}} - 1 \right) \quad (2)$$

The reference ratio is usually taken from standard mean ocean water isotope ratios ( $[\text{O}^{18}]/[\text{O}^{16}] = 2.0052 \times 10^{-3}$  and  $[\text{D}]/[\text{H}] = 1.5576 \times 10^{-4}$  [24]. Atmospheric measurements show that  $\delta(\text{HDO})$  is approximately in the range from  $-700\%$  to  $-200\%$  increasing with altitude [25–29]. The range from approximately  $-150\%$  to  $+20\%$  was observed for  $\delta(^{18}\text{O})$ . Equation 2 shows that an estimated error of 10% in the line profile measurements results in an error of  $\delta(\text{HDO})$  approximately 50%.  $\delta(^{18}\text{O})$  requires higher accuracy of the measurements and an error of 10% causes an error of  $\delta(\text{HD}^{18}\text{O})$  in the range from 90 to 180%.

#### 4. Conclusions

More than 400 000 Odin/SMR spectra have been averaged from the FM01 band (initially dedicated to the measurement of  $\text{ClO}$  and  $\text{O}_3$ ) in order to increase the signal-to-noise ratio. Three very weak lines have been detected at frequencies of 501.53439, 501.56739 and 501.59039 GHz that can be assigned to  $\text{N}_2\text{O}$  in vibrationally excited state [18, 19],  $\text{HD}^{18}\text{O}$  [20, 21], and  $\text{CH}_3\text{OH}$  [22, 23], respectively.

$\text{HD}^{18}\text{O}$  is a suitable tracer for studying the isotopic substitution and dynamics in the stratosphere and can be used for comparison of tropical and high latitude regions. The line of vibrationally-excited  $\text{N}_2\text{O}$  in the 501-GHz region in combination with the line of  $\text{N}_2\text{O}$  in ground state in the 502-GHz region provides possibility to use the ratio  $\text{N}_2\text{O}(501)/\text{N}_2\text{O}(502)$  as an indicator of the number of molecules in the vibrationally-excited state. Finally, information contained in the methanol

line within different latitude bands might help characterizing its space distribution on a global scale. These very encouraging results will be spread to other time periods within the lifetime of the Odin satellite (from November 2001 to the present time) and to other frequency bands, e.g. the 544.6 and 576.6 GHz bands.

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