GLOBAL OBSERVATIONS OF STRATOSPHERIC HEAVY OZONE ISOTOPOLOGUE ENRICHMENT WITH THE ODIN SUB-MILLIMETRE RADIOMETER

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ABSTRACT

The Odin Sub-Millimetre Radiometer (SMR) measures thermal emission lines of several minor ozone isotopologues such as symmetric and asymmetric O_3-18 and asymmetric O_3-17 at the atmospheric limb. The global data set spans over 11 years starting in late 2001. The basic characteristics and limitations of the observational data are presented and the global distribution of heavy ozone isotopologue enrichment as seen by Odin is described and discussed. Best results in terms of spatial coverage are obtained for asymmetric O_3-18, which shows a characteristic increase of the enrichment in the middle to upper stratosphere and the smallest enrichment in the polar lower stratosphere. Symmetric O_3-18 is characterised by a layer of maximum enrichment in the middle stratosphere.

Key words: Odin, SMR, stratosphere, heavy ozone isotopologue enrichment.

1. INTRODUCTION

The study of ozone isotopologue fractionation is important for our understanding of the oxygen cycle in the Earth middle atmosphere. An enrichment of (singly substituted) heavy ozone isotopologues with respect to the main isotopologue has been observed in the stratosphere employing balloon-borne mass-spectrometry [1], balloon-borne and space-borne infrared spectroscopy [2, 3], and airborne microwave spectroscopy [4]. The observed enrichment in the stratosphere has been explained by both mass-dependent and mass-independent effects in ozone formation ($O + O_2 + M \longrightarrow O_3 + M$) [5].

With the launch of the Odin Sub-Millimetre Radiometer (SMR) in 2001 [6], the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) on Envisat in 2002, the Atmospheric Chemistry Experiment Fourier Transform Spectrometer on SCISAT in 2003 [7], and the Superconducting Submillimeter-Wave Limb-Emission Sounder (SMILES) in 2009 [4, 8] several new global data sets have become available in the last decade for the study of ozone isotopologues in the stratosphere. This paper focuses on the global observations of stratospheric heavy ozone isotopologue enrichment with the Odin Sub-Millimetre Radiometer. We describe the different Odin data sets and their basic characteristics in Section 2. The enrichment of the heavier ozone isotopologues compared to the main isotopologue has been calculated and the global distribution of the enrichment is presented in Section 3. The main characteristics and limitations of the data such as spectroscopic retrieval uncertainties are summarised and discussed in Section 4.

2. ODIN/SMR MEASUREMENTS

The Odin/SMR instrument [9] measures thermal emission lines of several minor ozone isotopologues at the atmospheric limb. Observations of spectral lines of symmetric and asymmetric O_3-18 (usually denoted $^{16}O^{18}O^{16}O$ and $^{16}O^{16}O^{18}O$, respectively) are performed in a narrow spectral band centred at 490.4 GHz (see Figure 1). Limited information from a small line of asym-

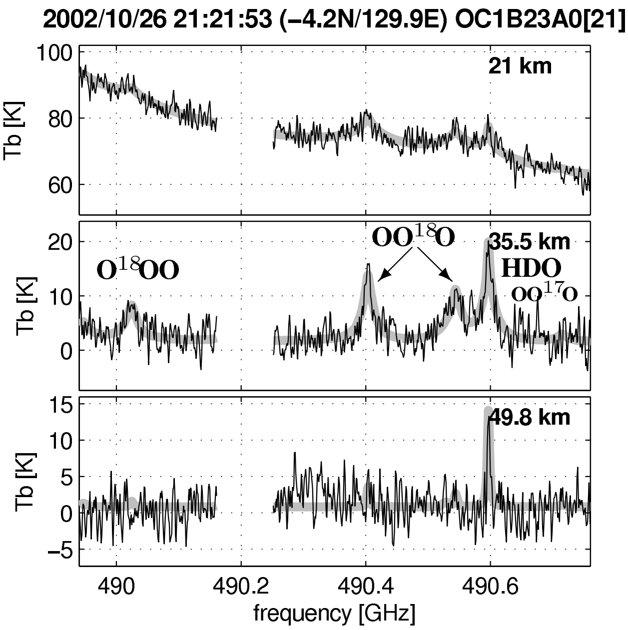

Figure 1. Typical Odin/SMR measurement of the 490.4 GHz band for different tangent altitudes (∼ 20, 35, 50 km) in the tropics (black thin line). The target lines can be best seen in the 35 km panel. The thick grey line is the result of a forward model calculation based on the retrieved trace gas profiles.

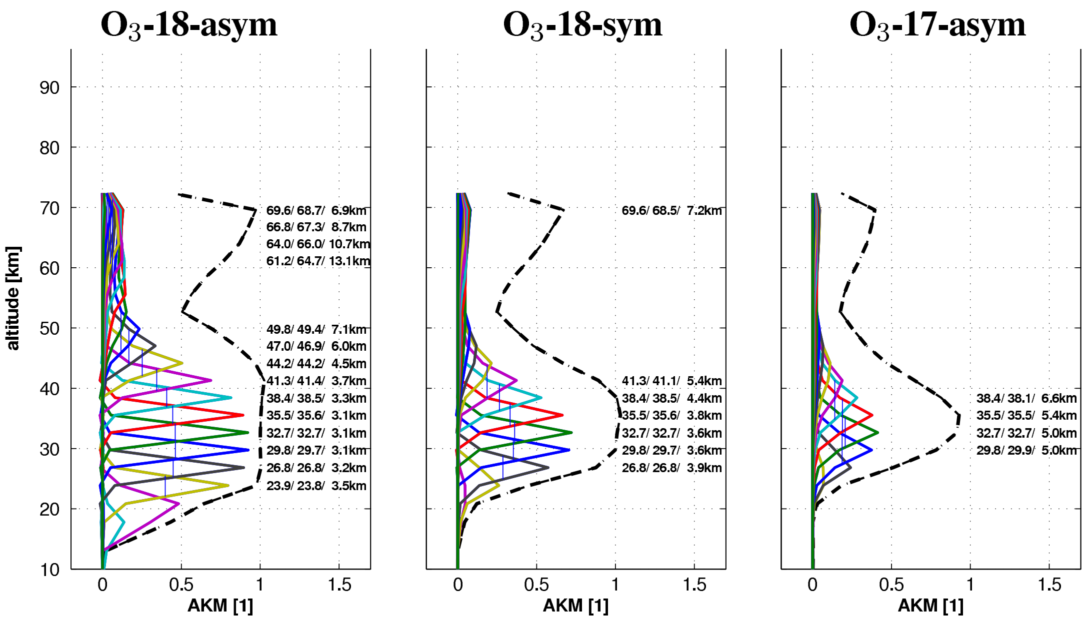

Figure 2. Averaging kernel functions corresponding to vertical profiles retrieved from the spectral lines shown in Figure 1 (coloured lines). Left and middle: asymmetric and symmetric O_3-18. Right: Asymmetric O_3-17. The full width at half maximum (FWHM) of the averaging kernel functions indicates the vertical resolution of the measurements at a given retrieval level. The sum over the averaging kernel functions of different retrieval parameters (volume mixing ratios) at a given altitude (measurement response) gives an indication of the altitude range where the measurements provide information (dashed black lines).

metric O_3-17 ($^{16}O^{16}O^{17}O$) is available from the same band. Measurements of the global distribution of stratospheric ozone isotopologues were performed on a regular basis of four observation days per month from late 2001 until April 2007 and about twice as often since then. The global Odin data set spans now a period of over 11 years.

Vertically resolved profiles can be retrieved in the stratosphere. The Odin/SMR operational retrieval processor [10, 11] employs an algorithm based on the Optimal Estimation Method [12]. The vertical ranges and resolutions of the volume mixing ratio profiles retrieved from individual limb scans are shown in Figure 2 for a typical measurement in the tropics. Asymmetric O_3-18 can be retrieved in the vertical region between about 25 and 45 km with a vertical resolution of 3-5 km. Information on symmetric O_3-18 is available in a slightly smaller vertical range from about 27 to 41 km and the vertical resolution is of the order of 4-6 km. Asymmetric O_3-17 can only be measured in a very narrow altitude range close to the ozone mixing ratio maximum (between 31 and 39 km, vertical resolution 5-6 km) and only in the tropics. The corresponding single-scan measurement precision in terms of volume mixing ratio is about 25 % for the three observed isotopologues.

3. GLOBAL DISTRIBUTION

The ratio of a minor ozone isotopologue measured by Odin/SMR with respect to the main isotopologue can be calculated using ozone observations from a different band. We use measurements of the 501.8 GHz band which were described and validated elsewhere [10, 13, 14, 15]. Measurements of the 501.8 GHz band are however not performed on the same days. In our analysis we first calculate monthly or seasonal averages for all latitudes and years and subsequently derive the isotope ratio from the obtained zonal mean fields. It should be noted that stratospheric ozone isotopologue enrichment is usually reported as an observed relative enrichment with respect to the isotope ratio of oxygen atoms found in Standard Mean Ocean Water (SMOW) of about $1:1/2700:1/500$ for $^{16}O:^{17}O:^{18}O$ and we will follow this convention (see e.g. [2] for further explanations).

Figure 3 shows the zonally averaged global distribution of the heavy ozone isotopologue enrichment for December-January-February (DJF) based on all Odin observations from December 2001 to February 2011. The global distribution of asymmetric O_3-18 (Figure 3, top) is characterised by an enrichment with respect to SMOW in the range of 5-20 % in the middle stratosphere at low and middle latitudes. The enrichment increases with altitude to roughly 20-30 % in the upper stratosphere. Minima of the enrichment are found in the polar regions below about 30 km with values smaller than 5 % and even indicating depletion, especially in the winter hemisphere. In contrast, the global distribution of O_3-18-sym (Figure 3, middle) shows a maximum layer of enrichment of roughly 10-20 % in the middle stratosphere and lower values in the upper stratosphere. As pointed out earlier, the Odin/SMR measurements of O_3-17-asym provide only information in the region of the tropical ozone volume mixing ratio maximum. The observed enrichment is of the order of 10 % around 35 km in the tropics.

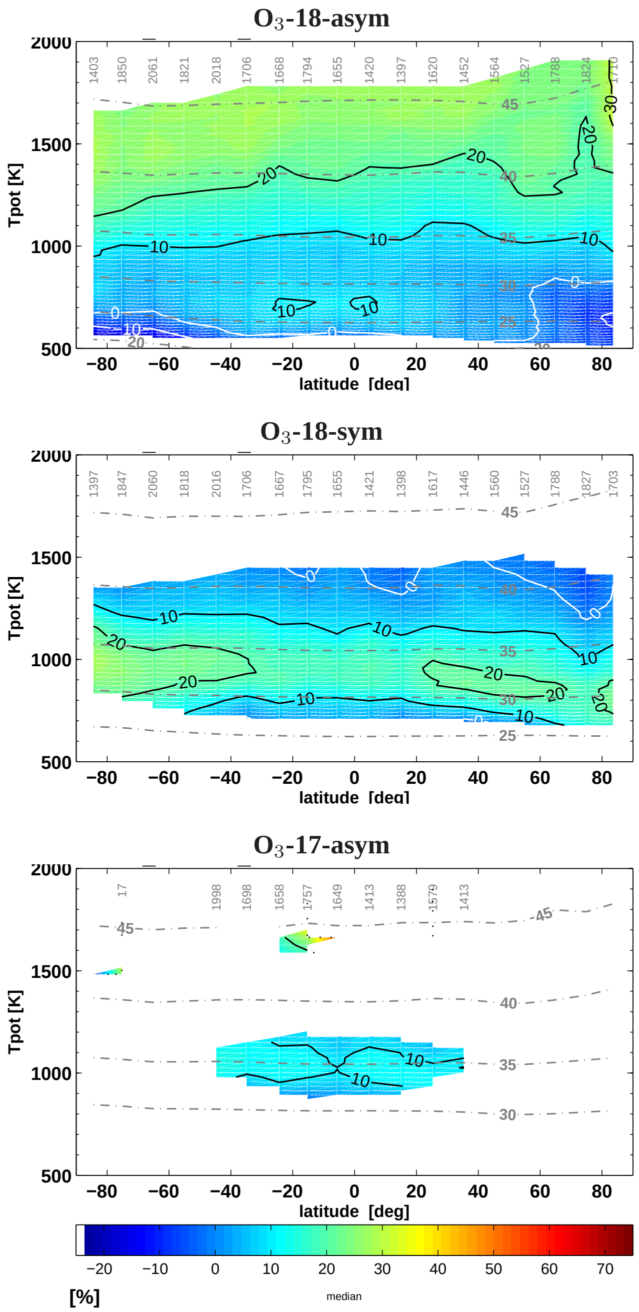

Figure 3. Global distribution of heavy ozone isotopologue enrichment derived from Odin/SMR observations between December 2001 and February 2011. Shown are seasonal results for the December-January-February (DJF) period for O_3-18-asym (top), O_3-18-sym (middle), and O_3-17-asym (bottom). Plotted is the enrichment in percent (colours and contours) as function of potential temperature and latitude. Averaged altitudes are indicated as dashed grey lines on the individual plots for the convenience of the reader. The numbers close to the top of the plots indicate the total number of vertical profiles averaged within each latitude band.

4. SUMMARY AND DISCUSSION

Global fields of ozone and its isotopologues (O_3-18-asym, O_3-18-sym and O_3-17-asym) have been retrieved in the middle stratosphere from 11 years of Odin/SMR measurements. The global distribution of the isotopic enrichment found in O_3-18-asym indicates an increase of the enrichment with altitude and minima in the polar regions below 30 km. Symmetric O_3-18 is characterised by a layer of maximum enrichment in the middle stratosphere.

Comparisons show that the derived isotope enrichment for asymmetric O_3-18 of about 12 % agrees within the errors with past balloon measurements at mid-latitudes [2] when averaged over the relevant 25-40 km altitude range (not shown). However, Odin/SMR measures quite different vertical profile shapes. Systematic errors arising from uncertainties in the radiometric calibration should cancel out to a first order as an isotopic ratio is calculated and the measurements were made with the same radiometer. A possible explanation is that the retrievals are affected by uncertainties in the spectroscopic parameters used by the forward model of the retrieval processor. These parameters are taken from the JPL (intensities, partition functions) and HITRAN (line-broadening parameters) spectral line catalogues [16, 17]. Figure 4 shows the retrieval errors arising from uncertainties of 5 % and 30 % in the collisional line broadening parameter of asymmetric O_3-18 (see [10] or [18] for a description of the calculation method). An uncertainty of about 5 % can typically be reached if the line broadening parameter is measured in the laboratory. In this case the resulting retrieval error is relatively small. This result can be compared to the assumption of a 30 % uncertainty (an illustrative worst case). Unfortunately the line-broadening parameters have not been measured for the spectral ozone isotopologue lines observed by Odin/SMR (Figure 1) and the actual uncertainty is at present unknown.

Further work is necessary in order to verify the spectroscopic data and to improve the retrieval algorithms with the aim to understand the mechanisms responsible for (1) the decrease of the isotopic ratio in polar winter, (2) the larger enrichment in the upper stratosphere of asymmetric O_3-18, and (3) to derive a reliable trend in heavy ozone isotopologue enrichment over the Odin period, considering instrumental drifts as well as natural changes in temperature and solar activity.

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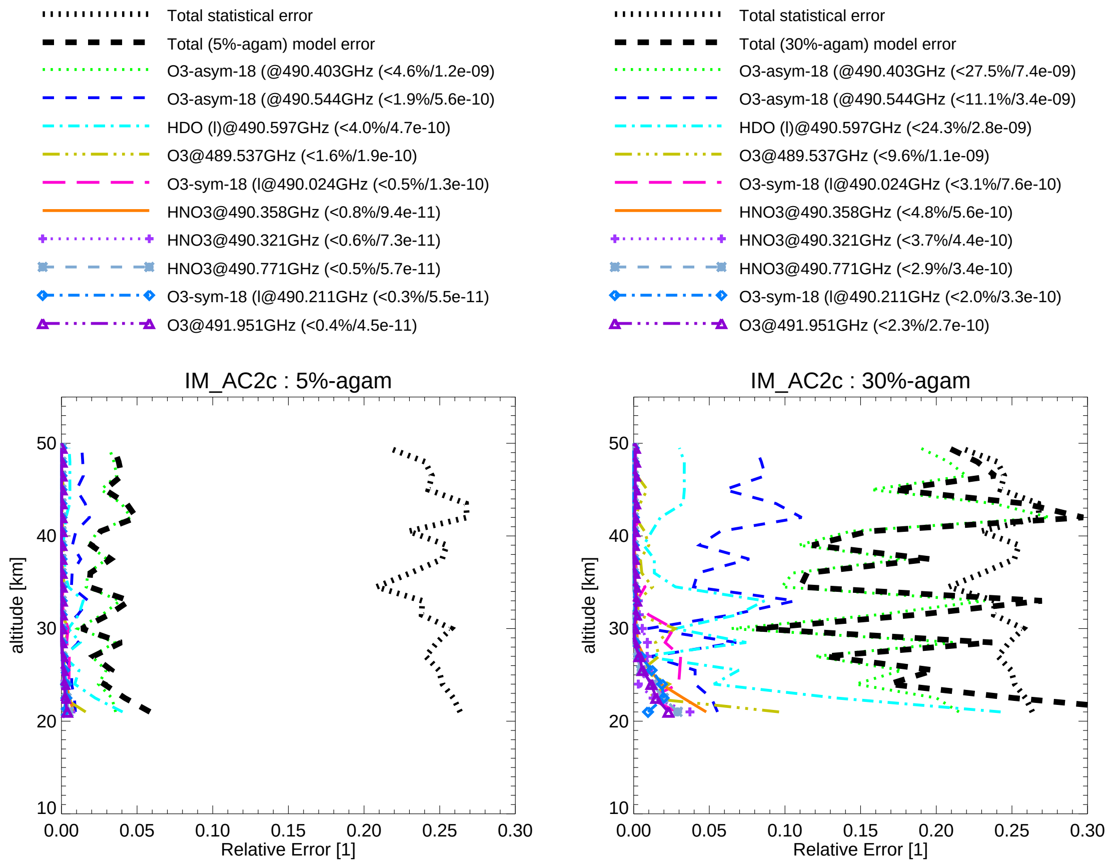

Figure 4. Estimated impact of spectroscopic retrieval errors for asymmetric O_3-18. Left: Retrieval errors resulting from a 5 % uncertainty in the collisional air broadening parameter (agam). Right, same but for a 30 % uncertainty. Errors resulting from individual lines as well as the combined (total) spectroscopic errors are plotted (see legend). The statistical retrieval error arising from noise in the Odin/SMR measurements is shown for comparison.

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