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MIPAS measurements of stratospheric Fluorine and Chlorine

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Abstract

Stratospheric fluorine and chlorine both have important effects on the atmosphere, especially chlorine on ozone depletion and fluorine in long-lived greenhouse gases. Atmospheric fluorine and chlorine are mainly comes from man-made species, such as CFCs, HCFCs. Here we study fluorine and chlorine inventories measured by Michelson Interferometer for Passive Atmospheric Sounding (MIPAS). MIPAS is a Fourier transform spectrometer instrument operated on the Envisat satellite from 2002-2012, operating in the near- to mid-infrared region at the Earth's limb. We present results of retrievals of these species using the Oxford processor, MORSE, to establish climatologically distributions.

1. Introduction

The Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) instrument, which operated on the Envisat satellite from 2002-2012 is a Fourier transform spectrometer for the measurement of high-resolution gaseous emission spectra at the Earth's limb. It operates in the near to mid-infrared, where many of the main atmospheric trace gases have important emission features. The initial operational products were profiles of Temperature, H_2O, O_3, CH_4, N_2O, HNO_3, and NO_2, and this list was recently extended to include N_2O_5 , $ClONO_2$, CFC-11 and CFC-12. Here we present preliminary results of retrievals of the third set of species under consideration for inclusion in the operational processor: HCN, CFC-14 (CF_4), HCFC-22, COF_2, CCl_4, SF_6, OCS, HOCl and C_2H_6. Stratospheric fluorine and chlorine both have important effects on the atmosphere, especially chlorine on ozone depletion and fluorine in long-lived greenhouse gases. Atmospheric fluorine and chlorine are mainly comes from man-made species, such as CFCs, HCFCs. Here we study fluorine and chlorine inventories measured by MIPAS.

2. Data and Method

The Oxford L2 processor MORSE has been used to retrieve the Volume Mixing Ratio (VMR) of halogenated compounds in the stratosphere. Here we analyzed these fluorine and chlorine species to their global vertical profiles and the total fluorine and chlorine VMRs. The fluorine species measured by MIPAS are COF_2, CFC-11, CFC-12, CF_4, CFC-22 and SF_6, while the chlorine species are CFC-11, CFC-12, CFC-22, CCl_4 , HOCl and $ClONO_2$. For each gas, we eliminated those data which there are cloud-affected and a priori has a significant contribution, which is the measure random error greater than 70% of the retrieval. A 5σ global spike test is then applied to each species to eliminate anomalous retrievals. Here we consider one month data for March 2010.

The total fluorine measured by MIPAS can be computer as:

$$[mF] = 2[COF_2] + [CFC\text{-}11] + 2[CFC\text{-}12] + 4[CF_4] + 2[CFC\text{-}22] + 6[SF_6] \quad (1)$$

with the '[ ]' representing the VMRs. [1] shows that most of the stratospheric fluorine species eventually form Hydrogen Fluoride (HF), which is not measured by MIPAS. Other fluorine containing species are small compared with these. Hence the total fluorine considered here is represented as following:

$$[F] = [mF] + [HF] \quad (2)$$

Fig. 1 shows that the expected HF The total [mF] decreases with altitude as fluorine ends up as HF, which is not measured by MIPAS. The expected HF zonal plot for 2010 March is constructed by the following:

$$[mHF] = [maxF] - [mF] \quad (3)$$

where [Fmax] is the maximum value of [mF]. Constructing the monthly zonal mean for expected HF from MIPAS, [mHF], is compared with the zonal mean from SLIMCAT, [sHF] in Fig. 2. Similarly for chlorine budget, the total chlorine measured by MIPAS can be computer as:

$$[mCl] = 3[CFC\text{-}11] + 2[CFC\text{-}12] + [HCFC\text{-}22]$$

+ 4[CCl_4] + [$ClONO_2$] + [HOCl] (4)

There are three major chlorine species, HCl, CH_3Cl and CFC-113,that are not measured by MIPAS. However, has spectral range of 0 – 3198 cm^{-1} and CFC-113 has 780-995 cm^{-1} and 1005-1232 cm^{-1} spectral ranges, which are the potential retrieval candidates of MIPAS. The approach of constructing expected [HF] can be applied for expected [HCl]+[CFC-113]+[CH_3Cl]. Data provided by ACE-FTS will be included for comparison in the future.

3. Results and Discussions

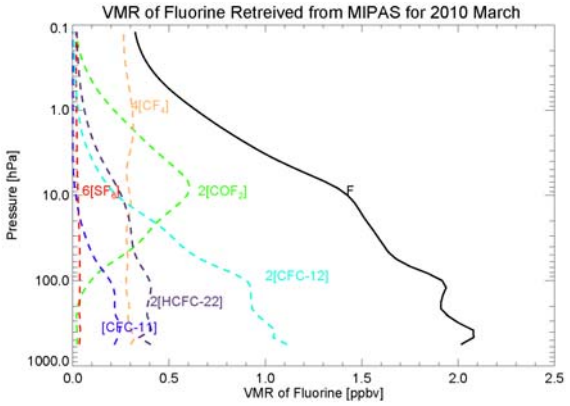

Figure 1: Global fluorine contained in species measured by MIPAS

Fig. 1 shows the vertical profiles with global average of all the fluorine species and total fluorine (mF) measured by MIPAS. In the lower stratosphere and upper troposphere, the atmospheric fluorine is mainly contributed by CFC-12. COF_2 contributes nearly half of the fluorine at 10hPa pressure level, while CF_4 dominates in the upper stratosphere. CF_4 remains mainly constant in the atmosphere.

Constructing the monthly zonal mean for expected HF from MIPAS by Equation 3, is compared with the zonal mean from SLIMCAT, [sHF], shown in Figure 3. The plots are consistent with each other. If adding the vertical profile of HF modelled by SLIMCAT in Figure 1, a vertical line of total [F] is expected.

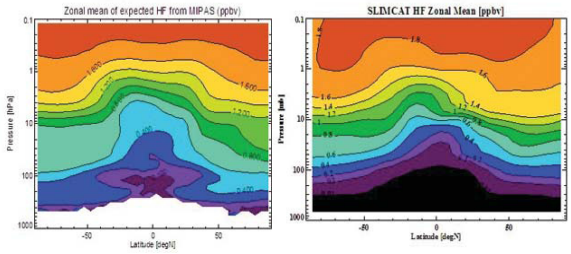

Figure 2: 2010 March zonal mean of HF deduced from MIPAS and modeled by SLIMCAT.

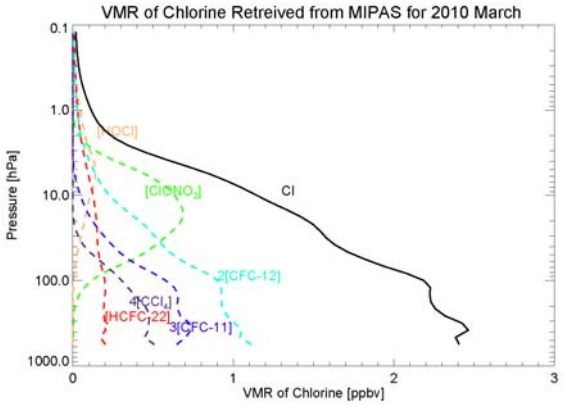

Figure 3: Global chlorine contained in species measured by MIPAS.

The solid line, representing the amount of [mCl], is decreasing with altitude, starting with about 2.4ppbv in the troposphere. Around half of the total chlorine, [mCl], in the troposphere is contributed by CFC-12 and around one third by CFC-11. $ClONO_2$ dominates the chlorine VMR at round 20hPa pressure level without considering the concentrations of HCl, CH_3Cl and CFC-113.

5. Future work

The preliminary results of the fluorine and chlorine budgets measured by MIPAS are presented. These results will be further compared with the ACE-FTS fluorine and chlorine budgets. Any trends detectable in the MIPAS 10-year dataset will be studied.

References

[1] Mahieu, E. and Duchatelet, P., et. al.: Validation of ACE-FTS v2.2 measurements of HCl, HF, CCl_3F and CCl_2F_2 using space-, balloon- and ground-based instrument observations, Atmospheric Chemistry and Physics, Vol. 8, pp. 6199–6221.