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HCl and ClO profiles inside the Antarctic vortex as observed by SMILES in November 2009: comparisons with MLS and ACE-FTS instruments

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Abstract

We present vertical profiles of hydrogen chloride (HCl) and chlorine monoxide (ClO) as observed by the Superconducting Submillimeter-Wave Limb-Emission Sounder (SMILES) inside the Antarctic vortex on 19–24 November 2009. The SMILES HCl value reveals 2.8–3.1 ppbv between 450 and 500 K levels in potential temperature (PT). The high value of HCl is highlighted since it is suggested that HCl was a main component of the total inorganic chlorine (Cly), defined as Cly ≃ HCl + ClO + chlorine nitrate ($ClONO_2$) inside the Antarctic vortex in spring, owing to low ozone values. To confirm the quality of two SMILES Level 2 (L2) data products provided by Japan Aerospace Exploration Agency (JAXA) and National Institute of Information and Communications Technology (NICT) from a view point of the partitioning of Cly, comparisons are made using other satellite data, from the Aura Microwave Limb Sounder (MLS) and Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS). HCl values from the SMILES NICT L2 product agree to within 10 % with the MLS HCl data between 425 and 650 K levels in PT and with the ACE-FTS HCl data between 425 and 575 K, respectively. The SMILES JAXA L2 product is 10 to 20 % smaller than that from MLS (ACE-FTS) between 400 (500 K) and 700 K. For ClO in daytime, the difference between SMILES (JAXA and NICT) and MLS was less than ±0.05 ppbv between 500 and 650 K with the ClO values less than 0.2 ppbv. $ClONO_2$ values as measured by ACE-FTS also reveal 0.2 ppbv at 475–500 K level, resulting in the HCl/Cly ratios of 0.91–0.95. The high HCl value and HCl/Cly ratio found from the three satellite instruments agree with the past observations inside the Antarctic vortex at this time (October to November) of year in the lower stratosphere.

1 Introduction

Hydrogen chloride (HCl) and chlorine monoxide (ClO) play an important role in the mechanism of ozone destruction in the stratosphere. The total inorganic chlorine (Cly) in the stratosphere is defined as the sum of Cl, 2 × Cl_2, ClO, HOCl, 2 × ClOOCl, OClO, chlorine nitrate ($ClONO_2$), and HCl. Observing the time evolution of these species in both of the Arctic and Antarctic vortices (lower stratosphere) is essential since HCl and $ClONO_2$ act as reservoirs for the chlorine radicals ($ClO_x = Cl + ClO + 2 \times ClOOCl$) that destroy ozone catalytically (e.g. WMO, 2007). It is also useful to evaluate model studies of the time series of Cly species (e.g. Santee et al., 2008b). The reservoirs, HCl and $ClONO_2$, are decomposed to yield Cl_2 through heterogeneous reactions occurring in/on sulfate aerosols and polar stratospheric clouds (PSCs) in winter. Consequently, ClO_x is elevated through photolysis/photochemical reactions in winter/spring, then it is deactivated into the reservoirs.

Increased amounts of HCl in the springtime Antarctic when ClO_x is deactivated were observed by ground-based Fourier Transform Infrared Spectroscopy (FTIR) instruments starting in 1987 (e.g. Murcray et al., 1989; Liu et al., 1992; Kreher et al., 1996). Using satellite instruments onboard the Upper Atmosphere Research Satellite (UARS) in 1990's, several studies showed the time evolution of ClO and HCl in the spring Antarctic vortices (e.g. Douglass et al., 1995; Santee et al., 1996; Chipperfield et al., 1996; Grooß et al., 1997; Mickley et al., 1997). On 3–12 November 1994, measurements from The Atmospheric Trace MOlecule Spectroscopy (ATMOS) on the Space Shuttle was conducted. Rinsland et al. (1996) and Michelsen et al. (1999) suggested that the high HCl/Cly ratio (∼ 0.9) was maintained in the Antarctic vortex. Also, in November 1996, it was reported that the high HCl and low $ClONO_2$ were observed by HALogen Occultation Experiment (HALOE) and Improved Limb Atmospheric Spectrometer (ILAS) satellite instruments, respectively (Hayashida and Sugita, 2007). All of these results confirm a theoretical study by Prather and Jaffe (1990) who showed a mechanism for increased HCl values after the "ozone hole" period in the Antarctic. This can be understood as low values of ozone shifting the partitioning of ClO_x into Cl, so that the reaction $Cl + CH_4$ that forms HCl proceeds faster than the reaction $ClO + NO_2 + M$ that forms $ClONO_2$, thus reaching a steady state with a high HCl/Cly ratio.

Simultaneous in situ aircraft measurements of ClO, HCl, and $ClONO_2$ have been available since 1997 (Bonne et al., 2000). At that time, an aircraft mission was performed in the 1999/2000 Arctic winter; the time evolution of the Cly species has been investigated (e.g. Wilmouth et al., 2006). Dufour et al. (2006) also showed an evolution of the Cly partitioning in the 2004/2005 Arctic winter from the Atmospheric Chemistry Experiment-Fourier Transform Spectrometer (ACE-FTS) measurements. However, the HCl value or HCl/Cly ratio in the Arctic was lower than those in the Antarctic (e.g. Santee et al., 2008b; Manney et al., 2011; Wegner et al., 2012), even with severe ozone loss in the 2010/2011 winter (e.g. Manney et al., 2011). An exception was the 1996/1997 Arctic winter when the breakup of the vortex occurred in May; an HCl/Cly ratio of 0.8–0.9 was observed by HALOE (Douglass and Kawa, 1999; Konopka et al., 2003).

In the Antarctic, no comprehensive aircraft campaign has been done since the 1994 mission (Tuck et al., 1997; Jaeglé et al., 1997), so satellite measurements are crucial to study the partitioning of Cly in the Antarctic. Although a high HCl/Cly ratio is usually seen in the upper stratosphere, it has also been seen in the lower stratosphere in the combined data from the Aura Microwave Limb Sounder (MLS) and ACE-FTS data (WMO, 2007; Santee et al., 2008b) or just in the MLS data (Santee et al., 2011; de Laat and van Weele, 2011) since 2004. This is due to the combination of diabatic descent of air inside the vortex, low ozone values, and isolation of the lower stratospheric vortex.

The International Space Station (ISS) borne instrument, the Superconducting Submillimeter-Wave Limb-Emission Sounder (SMILES), started operations in October 2009. SMILES observed latitudes between ∼ 66° N and ∼ 38° S. On 19–24 November 2009, there were, however, measurements including the Antarctic (∼ 38° N to ∼ 66° S) due to the ISS observation geometry. The breakup of the Antarctic vortex in the lower stratosphere occurred in December 2009 (NOAA, 2009), so that some measurements were taken inside the vortex where high HCl values are expected. In this paper, we focus on these SMILES measurements, and analyze the vertical profiles of HCl and ClO inside the Antarctic vortex to confirm those data quality from a view point of the partitioning of Cly, through comparisons with satellite data for the same time period from the MLS and ACE-FTS instruments. Comparisons performed when the HCl value is high (~ 3 ppbv) in the lower stratosphere will provide valuable information on the HCl measurements.

2 Measurements of SMILES

SMILES is a passive sensor to measure the limb of Earth's atmosphere in the frequency bands around 625 and 650 GHz. The instrument was attached to the Japanese Experiment Module (JEM) onboard ISS. The emission lines of O_3, HCl, ClO, HO_2, HOCl, BrO, and other molecules can be found in the low-noise spectra obtained with a 4-K mechanical cooler and superconductor-insulator-superconductor (SIS) mixers. The SMILES observations started on 12 October 2009 and ceased on 21 April 2010 due to the failure of a critical component in the submillimeter local oscillator. Results from SMILES have demonstrated its high potential for observing atmospheric minor constituents in the middle atmosphere, as shown in Kikuchi et al. (2010).

Since SMILES has three specified detection bands: 624.32–625.52 GHz (Band A), 625.12–626.32 GHz (Band B), and 649.12–650.32 GHz (Band C) with two acousto-optical spectrometers, observations of Bands A, B, and C are made on a time-sharing basis, such as Bands A + B, A + C, or B + C. In the period that we analyze, the Bands B + C measurements were performed. The $H^{35}Cl$ rotational transition ($J = 1$–0) is located at 625.9 GHz in Band B. The ClO transitions in the ground ro-vibronic state ($J = 35/2$–$33/2$) are located at 649.445 and 649.451 GHz in Band C. Vertical resolution is 3.5–4.1 km.

The SMILES Level 2 (L2) data processing systems (Takahashi et al., 2010; Baron et al., 2011) retrieve vertical profiles of the atmospheric minor constituents from the calibrated radiance observations (Level 1b data, hereafter referred to as L1b). In this paper, we used two SMILES L2 data products which are separately processed with different retrieval algorithms: the version 2.1 (hereafter referred to as v2.1) of JAXA

L2 products (Mitsuda et al., 2011) (hereafter SMILES-JAXA) and the v2.1.5 of NICT L2 products (Kasai et al., 2013; Sagawa et al., 2013) (hereafter SMILES-NICT). Both of the data sets were publicly released on 5 March 2012 (http://smiles.tksc.jaxa.jp/ for SMILES-JAXA and http://smiles.nict.go.jp/ for SMILES-NICT).

Both of the retrieval algorithms used same L1b data and similar instrument functions in the forward model calculations. The vertical inversion of the volume mixing ratio profiles are based on the optimal estimation method (e.g. Rodgers, 2000). However, several differences in the two algorithms made different data products (Kasai et al., 2013; Sagawa et al., 2013). Thus, it is of importance to compare both of the L2 products to investigate matters relating to the algorithms. Here, we briefly present the differences that affect mostly the lower stratosphere data quality.

One of the major differences is the spectral bandwidth used in the retrieval analysis. The SMILES-JAXA L2 algorithm uses a full bandwidth (~ 1.2 GHz) of each detection band to simultaneously retrieve all the observed species in that band. On the other hand, SMILES-NICT processing employs the sequential retrieval approach using a narrow bandwidth around the targeted species (Baron et al., 2011). For example, after O_3 and temperature profiles were retrieved, HCl is retrieved from the spectra extracted at ±300 MHz of the HCl line centre. ClO is retrieved from the spectra in Band C using a spectral bandwidth of 400 MHz centered on the ClO line. For the species of interest in this paper (HCl and ClO), the information in the lower stratosphere can be retrieved also from the far wings of their lines. Therefore, the usage of a limited spectral bandwidth results in a decrease of the sensitivity and degradation of the vertical resolution at lower altitudes. Whereas, for the full bandwidth approach, it could be difficult to deal with non-linearity of the detector in the far wings. Both of the approaches, thus, may have some disadvantages.

Different assumptions on a priori state can also introduce systematic biases on retrieved profiles, particularly when the measurement sensitivity is low. The a priori profile of SMILES-JAXA is based on mean profiles for month, latitude, (and day-night separately for ClO), using the MLS version 2.2 data. The ClO data has been bias-corrected as suggested by Santee et al. (2008a). While SMILES-NICT used a single common a priori profile for HCl or ClO for all observations.

For the modeling of continuum absorptions of H_2O and dry air, SMILES-JAXA uses the MPM-93 model (Liebe et al., 1993) with a scaling factor of 1.34. SMILES-NICT uses a model on the basis of Pardo et al. (2001). The dry air continuum absorption coefficient was increased by a factor of 20 % from the original formula, and retrieves H_2O as the continuum (Kasai et al., 2013). The above differences employed in each retrieval processing could be the cause of differences in the results of retrieved volume mixing ratio profiles. Other lower stratospheric differences in the SMILES-JAXA and -NICT processing are: the approach for measurement tangent height correction, a priori state of temperature and pressure, and so on. More detailed descriptions of these differences are found elsewhere (Kasai et al., 2013; Sagawa et al., 2013).

For spectroscopic parameters, the line frequency ν_0, the air broadening coefficient γ_{air}, and its temperature dependence n_{air}, there are also differences between the two forward model calculations. For $H^{35}Cl$ retrievals, SMILES-JAXA used $\nu_0 =$ 625 901.6584, 625 918.6975, and 625 932.0081 MHz (Perrin et al., 2005). SMILES-NICT used $\nu_0 =$ 625 901.6627, 625 918.7020, and 625 931.9977 MHz (Cazzoli and Puzzarini, 2004). For ClO retrievals, SMILES-JAXA used $\nu_0 =$ 649 445.250 and 649 451.072 MHz, $\gamma_{\mathrm{air}} = 2.11\,\mathrm{MHz\,hPa^{-1}}$, and $n_{\mathrm{air}} = 0.85$ (Oh and Cohen, 1994). SMILES-NICT used some modification to those values based on an original laboratory study: $\nu_0 =$ 649 445.040 and 649 451.170 MHz, $\gamma_{\mathrm{air}} = 2.15\,\mathrm{MHz\,hPa^{-1}}$, and $n_{\mathrm{air}} = 0.77$. A list of the parameters is found in Baron et al. (2011).

Finally, we present error analysis and validation results for both HCl and ClO. For previous versions of SMILES-JAXA, precision is less than 10 % for HCl at 20–60 km (Kikuchi et al., 2010; Shiotani et al., 2010). For ClO, precision is estimated to be 10 pptv between 19 and 28 km (Suzuki et al., 2012). This is evaluated from the standard deviation of the nighttime ClO measurements. The mean nighttime data also provides a good measure for a bias estimation of the JAXA ClO data products, which has already shown a validity for the BrO data product (Stachnik et al., 2013). This bias will be considered for ClO later (Sect. 4.2). The cause of this bias is unknown, but it seems to be due to an unexpected quadratic spectral baseline of the L1b data, which should be flattened by calibration, and a small residual between L1b and the forward model calculation, which could also arise from the quadratic continuum emission feature (Suzuki et al., 2012). The full-bandwidth approach used in the JAXA's processing system tends to be susceptible to these baseline-related effects. Validation studies for SMILES-JAXA are, to date, in preparation.

For HCl from SMILES-NICT (v2.1.5), the precision is estimated to be ∼ 1–2 % at the 10 hPa level and it increases to 8 % at 80 hPa. The validation of SMILES-NICT HCl was performed by comparing with MLS version 3.3 data and ACE-FTS version 3.0 data (Yokoyama et al., 2013): the absolute difference between SMILES-NICT and MLS amounts to −0.2 ppbv at 80 hPa and +0.1 ppbv at 10 hPa. The absolute difference between SMILES-NICT and ACE-FTS ranges from zero to +0.1 ppbv at 80–10 hPa levels. For ClO from SMILES-NICT (v2.1.5), the precision is estimated to be 100 and 30 pptv between 100 and 10 hPa (Sato et al., 2012). Sagawa et al. (2013) have also estimated the bias uncertainty of 5–10 pptv below 20 hPa level, and made a validation study: the absolute differences between SMILES-NICT and MLS version 3.3 data or Odin Sub-Millimetre Radiometre (SMR) Chalmers version 2.1 data (Murtagh et al., 2002) is within ±0.1 ppbv at 80–10 hPa levels.

3 Method

The 2009 Antarctic vortex at 450 K potential temperature (PT) level exhibited a typical seasonal pattern: it developed in mid-May, maximized around September, and then diminished in the late November/December period (NOAA, 2009). We analyzed SMILES data obtained on 19–24 November, when the measurements were conducted between 38° N and 66° S. During those days, the polar vortex was somewhat shifted toward South America, so that the edge of the vortex reached to around 50° S in the lower stratosphere (de Laat et al., 2010). Therefore, we extracted the SMILES data between 50 and 66° S. The profiles of SMILES (both JAXA and NICT) were discarded when flagged with “field-of-view interference” and “altitude error” according to a release note (http://smiles.tksc.jaxa.jp/l2data/index_e.html). For SMILES-JAXA, profiles with “convergence” value greater than 10 are discarded. Further, data points with precision larger than a half of a priori error are also discarded. For SMILES-NICT, profiles with “chi-square” value larger than 0.6 are discarded. Data points with measurement response smaller than 0.85 are also discarded.

In order to show the quality of the SMILES data for studying Cly partitioning inside the Antarctic vortex, we have made comparisons with other satellite data. In the same period as above, the MLS instrument on the National Aeronautics and Space Administration’s (NASA) Earth Observing System (EOS) Aura satellite (Waters et al., 2006) has operated between 82° N and 82° S. MLS measures millimeter- and submillimeter-wavelength thermal emission from the limb of Earth’s atmosphere. We used HCl and ClO data products retrieved with the version 3.3 data processing algorithm (Livesey et al., 2006) (http://mls.jpl.nasa.gov/). The quality of the HCl data is as follows (Livesey et al., 2011): vertical resolution in the lower stratosphere is ~ 3 km and precision is 0.3–0.2 ppbv at 100–10 hPa. The quality of the ClO data is as follows (Livesey et al., 2011): vertical resolution is 3.0–4.5 km, precision is ±0.3 ppbv at 147 hPa and ±0.1 ppbv at 100–22 hPa, and bias estimates are from −0.1 to zero ppbv at 50–70° S in November. In this study, we used data taken at 50–66° S. Data screening is also done according to Livesey et al. (2011).

ACE-FTS, the primary instrument on the SCISAT-1 satellite, is a high-resolution infrared Fourier transform spectrometer that measures solar occultation spectra between 2.2 and 13.3 µm (Bernath et al., 2005). It has also operated in the period of 19–24 November at latitudes between 65.7 and 69.3° S. This is a somewhat more southerly latitude than SMILES and MLS measurements used in this study. Vertical resolution is 3–4 km. We used HCl and $ClONO_2$ data products retrieved with the version 3.0 data processing algorithm (Boone et al., 2005) (http://www.ace.uwaterloo.ca/). The error analysis of the HCl data has not yet been evaluated, but the measurement variability that provides an upper limit on retrieval precision is estimated to be on the order of 5 % at 20–55 km (Mahieu et al., 2008). The fitting error of the $ClONO_2$ data is below 10 % at 20–30 km and increases to 40 % at 14 km (Wolff et al., 2008). Dufour et al. (2006) has provided an error budget for a single occultation: the total errors for HCl and $ClONO_2$ are, respectively, estimated to be 4–7 and 10–19 % between 16.5 and 28.5 km.

To extract observed data only inside the Antarctic vortex, we used Derived Meteorological Products (DMPs) (Manney et al., 2007). For both the MLS and ACE-FTS measurement locations and times, DMPs are produced to facilitate comparisons between different satellite instruments. They include PT, potential vorticity (PV), equivalent latitude (EqL, the latitude that would enclose the same area between it and the pole as a given PV contour Butchart and Remsberg, 1986), horizontal winds and tropopause locations. In this study, we used DMPs derived from the NASA Global Modeling and Assimilation Office's (GMAO) Goddard Earth Observing System (GEOS) data set (version 5), hereafter referred to as GEOS-5 (Reinecker et al., 2008). Using DMPs to view measurements with respect to their air mass characteristics is valuable in a study of chemistry and dynamics inside/outside the vortex. To compare SMILES measurements with those from MLS and ACE-FTS, we also obtained DMPs for the SMILES measurements for the study period.

We used PT for a vertical coordinate, since we focus on the observations in the lower stratosphere, where Cly has much longer chemical lifetime compared to the transport timescale at this time and location. In this study, we constructed averaged "vortex profiles" using data points at each vertical level that are poleward of the vortex edge center shown in the each DMP file. For instance, the vortex edge center is located at ~ −65° EqL (negative values are assigned in the southern hemisphere) at the 490 K PT level. The resulting EqL values around 490 K inside the Antarctic vortex are, on average, −71, −76, and −75° EqL for SMILES, MLS, and ACE-FTS observations, respectively. The volume mixing ratio of tracer species mapped with respect to the PT and EqL is nearly constant with EqL over a certain PT range within the vortex (e.g. Fig. 5 of Lingenfelser and Grose, 2002).

For the SMILES and ACE-FTS data, PT values from the DMPs are assigned at each altitude grid, whereas the MLS data, PT values from the DMPs are assigned at each pressure grid. The above averaged profiles are calculated from data contained within a 25 K wide PT bin. In general, intercomparison among satellite measurements is a difficult task because of the different vertical and horizontal resolutions considered (e.g. Ceccherini et al., 2003; Ridolfi et al., 2006). Thus, any interpolation of data onto specific PT levels has not applied in this study. In addition, the uncertainty in the meteorological data should also be concerned in such a comparison study. It was suggested that the GEOS-5 temperatures have about 1 K low bias (Lambert et al., 2012). The pointing uncertainty in the tangent altitude was estimated to be 340, 150, and 150 m for SMILES, MLS, and ACE-FTS, respectively (Kikuchi et al., 2010; Cofield and Stek, 2006; Harrison and Bernath, 2013) (for SMILES, it will be updated for the upcoming data versions; Ochiai et al., 2013). These uncertainties are inherent in the constructed average profiles. The individual profiles of SMILES HCl/ClO, MLS HCl/ClO, and ACE-FTS HCl/$ClONO_2$ used for the averages are found in Sugita et al. (2012), for reference.

Because the ISS is in a non sun-synchronous circular orbit with an inclination angle of 51.6° to the equator, the SMILES measurement at each tangent point occurs at various local solar times (LST). Therefore, it is crucial to consider the diurnal cycle of ClO and $ClONO_2$ in comparisons among SMILES, MLS, and ACE-FTS. In the following section, we consider SMILES and MLS ClO data only for daytime (solar zenith angles (SZAs) less than 85°), since the nighttime (SZAs larger than 95°) observations of ClO reveal values around zero. Although the number of data points is small, the SMILES ClO values in the SZA range between day and night times ($85° < \mathrm{SZA} < 95°$) reveal zero to 0.05 ppbv between 400 and 700 K levels (not shown). Figure 1 shows measurement latitudes as a function of LST used in this study (data taken inside the vortex). The daytime measurements of SMILES occurred at LSTs between 03:00 and 09:00 LST whereas the daytime measurements from MLS occurred at LSTs of 14:00–15:00 LST. These LST differences should be carefully treated in the following discussion. Data with SZAs larger than 85° are color-coded in black for each symbol. For

ACE-FTS measurements, all of the occultations occurred at a SZA of 90° from the satellite sunset for this time and location.

4 Results and discussion

4.1 HCl

Figure 2 shows the vortex profiles of SMILES and MLS HCl volume mixing ratios taken on 19–24 November 2009 at latitudes between 50 and 66° S as a function of PT. PTs of 400, 500, 600, and 700 K approximately correspond to altitudes of 15, 19, 23, and 26 km, respectively. Data are averaged within the PT bin between 400–425 and 675–700 K. At first, we focus on the results from SMILES. The average and one sigma standard deviation are shown for both of SMILES-JAXA and -NICT data in the left panel (the blue and green squares, respectively). The absolute difference between the SMILES-JAXA and -NICT averages in each bin is also shown in the same panel (the black cross). Precision was calculated as an average from the measurement precision reported for the respective data files, and is shown as the dashed line: it is quite small compared to the retrieved value. The right panel shows the relative difference expressed as the absolute difference divided by the mean of SMILES-JAXA and -NICT data (the black circle). The data values used in each bin are shown on the right-side of the right panel. Since the PT interval is narrower than the retrieval grids, these numbers are non-uniform in the vertical range.

The peak value of SMILES HCl reveals 2.8 and 3.1 ppbv at 450–475 K bin (JAXA) and 475–500 K bin (NICT), respectively. The relative difference between SMILES-JAXA and -NICT amounts to 7–21 % between 425–450 and 550–575 K bins (the absolute difference up to 0.5 ppbv), and −10 to 2 % between 575–600 and 650–675 K bins, resulting in a better agreement than in the lower PT levels. The cause of these differences of up to 20 % found in the lower levels is unclear, but it is related to the different retrieval approaches described in Sect. 2. Besides the difference in the spectral bandwidth used in the retrieval analysis, another possible cause could be the different a priori profiles used for each algorithm. We have tested JAXA's processing system using a more realistic a priori (constructed from the version 2.2 MLS HCl data inside the Antarctic vortex). However, the result was almost the same because the measurement sensitivity is sufficiently high that the retrieval is insensitive to the a priori. Another contributing factor may be the difference in dealing with the modeling of continuum absorptions (see Sect. 2). A further investigation to quantify the difference is ongoing along with updates of both retrieval algorithms.

To compare these SMILES HCl data products with those from other satellite instruments, the Aura MLS and ACE-FTS HCl data are analyzed. Figure 2 also shows the vortex profiles of MLS HCl (the red squares). The maximum mixing ratio of MLS is seen at 475–500 K bin with a value of 3.4 ppbv. The shape of the profile is similar to that of SMILES-NICT. Better agreement is found between SMILES-NICT and MLS (the green cross) than from SMILES-JAXA and MLS (the blue cross). The relative difference between SMILES-NICT and MLS ranges from −5 to 13 % between 425–450 K and 575–600 K bins, but it increases to 19 % at 650–675 K bin. For SMILES-JAXA and MLS, the relative difference amounts to 7–23 % between 400–425 and 675–700 K bins, except for 33 % at 425–450 K bin. In the period of November 2009, Band 13 (primary to detect $H^{35}Cl$) of the MLS instrument was not used and HCl was retrieved using Band 14; it has been suggested that retrievals of HCl from Band 14 have a high bias at high HCl values (above ∼ 3 ppbv) (Livesey et al., 2011). The positive difference found in both of the comparisons between SMILES and MLS at 450–500 K might reflect an artifact of the MLS Band 14 measurement to some extent (as mentioned just below, the ACE-FTS HCl value also does not exceed 3 ppbv at the peak).

Figure 3 shows the vortex profiles of SMILES and ACE-FTS HCl taken on 19–24 November 2009 as a function of PT. The SMILES data are identical to those shown in Fig. 2. Since the ACE-FTS measurements are made using solar occultation, observations are limited to a certain latitude in a certain period (see Sect. 2 and Fig. 1). The number of observations (occultation events) inside the vortex for ACE-FTS is 26. Since we used the 1 km altitude grid data, some profiles have two data points in a bin. Thus, the number of data used in the 400–425 K bin is 29 at the maximum. The maximum mixing ratio of ACE-FTS is seen at 500–525 K bin with a value of 2.9 ppbv that is similar to the results from SMILES and MLS, with a slightly higher PT level than those of SMILES and MLS.

The relative difference between SMILES-JAXA and ACE-FTS (the blue circle) ranges from –5 to 19 %. The relative difference between SMILES-NICT and ACE-FTS (the green circle) ranges from −9 to −1 % between 425–450 and 500–575 K bins, but it increases to 16–22 % between 575–600 and 650–675 K bins. In summary, a general feature is that SMILES-JAXA HCl is somewhat lower than both of MLS and ACE-FTS, whereas SMILES-NICT HCl shows better agreement with both MLS and ACE-FTS between 425 and 575 K. Above 575 K, SMILES-NICT HCl is, however, lower than that from both MLS and ACE-FTS.

4.2 ClO

Figure 4 shows the vortex profiles of the daytime ClO volume mixing ratios as measured by SMILES and MLS on 19–24 November 2009 at latitudes between 50 and 66° S as a function of PT. Generally, both of the SMILES-JAXA and -NICT profiles show increasing values up to 0.1–0.2 ppbv at 625–700 K with increasing altitudes. Below 600 K level, the ClO values were less than 0.1 ppbv. For the SMILES-JAXA ClO product, the bias should be corrected, as mentioned in Sect. 2. For this latitude and period considered here, the bias is estimated to be –33 pptv at 16 km and negligibly small above 19 km. Thus, this value (33 pptv) is added before taking the average at 16 km (400–450 K level). The absolute difference between SMILES-JAXA and -NICT ClO is ±0.05 ppbv between 450–475 and 600–625 K bins, although the difference between the two products amounts to −0.07 ppbv at 625–675 K and 0.07 ppbv at 425 K. The ClO values themselves are as small as 0.1–0.2 ppbv, making large relative differences.

In Fig. 4, the MLS ClO is also averaged only from measurements taken with SZAs less than 85° (see Fig. 1). Similar to the results from SMILES, the daytime MLS ClO values increased to ∼ 0.35 ppbv at 675–700 K bin. The reported precisions and one sigma standard deviations of the data are smaller in SMILES than in MLS, reflecting the difference in system noise temperatures between the two instruments (Kikuchi et al., 2010). It should be noted that some negative values in the MLS data are found below the 500 K level. We have bias-corrected with 0.09–0.15 ppbv (added) below 500 K (100 and 68 hPa levels) depending on latitudes (5° bin) for each ClO data before taking the averages as suggested by Livesey et al. (2011). The absolute difference between SMILES and MLS ClO is almost ±0.05 ppbv between 400 and 650 K, although the values themselves are again below 0.15 ppbv, making large relative differences.

As shown in Fig. 1, the MLS observations were taken at 14:00–15:00 LST, whereas the SMILES observations were taken at 03:00–09:00 LST. Thus, a large difference (0.14 ppbv) between SMILES-JAXA and MLS at 675–700 K bin (∼ 26 km) could be partly associated with diurnal changes in ClO. At this altitude, time, and location, the diurnal cycle of ClO is thought to take place via the photolysis of $ClONO_2$ (Santee et al., 2008b). Indeed, if we take an average only using the SMILES-JAXA ClO data with SZAs less than 55° in the 675–700 K bin (the number of data is 5 out of 57), the ClO value of 0.21 ppbv (SZAs less than 85°) varies to 0.30 ppbv. As a result, the absolute difference between SMILES-JAXA and MLS in the 675–700 K bin becomes from 0.14 to 0.05 ppbv, revealing better agreement (not shown). Therefore, the SZA or LST difference found in comparison of ClO within the daytime should be carefully treated at this altitude.

4.3 Cly partitioning

Since Cly in the stratosphere in 2009 was ∼ 3.3–3.5 ppbv (WMO, 2011), it is clear that HCl dominates Cly in this altitude range and time period. Such a feature is usually not seen in the lower stratosphere, but seen in the upper stratosphere (Cly $\simeq$ HCl) (e.g. WMO, 2011). This anomalously high HCl occurs as a result of low O_3 values ("ozone hole") in October, as follows (e.g. Mickley et al., 1997). The two competing reactions of the NO radical:

$$\mathrm{NO + ClO \rightarrow Cl + NO_2} \tag{R1}$$

$$\mathrm{NO + O_3 \rightarrow O_2 + NO_2} \tag{R2}$$

are the key to understanding this behavior. Under "ozone hole" conditions with low O_3 and high ClO values, Reaction (R1) becomes faster than Reaction (R2), allowing increased values of Cl. The low O_3 values directly slow the reaction:

$$\mathrm{Cl + O_3 \rightarrow ClO + O_2}\,. \tag{R3}$$

Thus, the production of HCl occurs gradually from September to October through the reaction:

$$\mathrm{Cl + CH_4 \rightarrow HCl + CH_3} \tag{R4}$$

as shown in e.g. Santee et al. (2008b). HCl production is thus sensitive to the amount of O_3. However, HCl values in mid-October in 2004 (when a smaller degree of ozone loss was found) were slightly lower than those in 2005 (when the degree of ozone loss was normal) (Santee et al., 2008b). HCl is destroyed through reactions with the OH radical and on the surface of sulfate aerosols, but both are so slow that the chemical lifetime of HCl is long compared to the transport time in November (i.e. high HCl values should be found as long as the vortex exists).

Figure 5 (left panel) shows vertical profiles of ClO, $ClONO_2$, and HCl inside the vortex on 19–24 November 2009, summarizing Figs. 2, 3, and 4. The bars show the one sigma standard deviation of data for each average. For the ACE-FTS $ClONO_2$ data, the same occultation measurements are used in the average as were used for the HCl data. It is of importance to examine Cly species other than HCl and ClO. Because of the Reactions (R1) through (R4), the formation of $ClONO_2$ by $ClO + NO_2 + M$ should also be suppressed significantly inside the vortex where the HCl value is high at 450–500 K level (e.g. Prather and Jaffe, 1990). Indeed, the ACE-FTS $ClONO_2$ values are less than 0.3 ppbv below 475–500 K bin, as shown in Fig. 5. Above the 500–525 K level, $ClONO_2$ increases with altitude to 1.1–1.2 ppbv at 650–700 K.

The ratio of HCl to Cly is shown in the right panel of Fig. 5. We take Cly values as below:

$$\text{Cly(SMILES)} = \text{HCl(SMILES)} + \text{ClO(SMILES)} + \text{ClONO}_2\text{(ACE-FTS)} \tag{1}$$

$$\text{Cly(MLS)} = \text{HCl(MLS)} + \text{ClO(MLS)} + \text{ClONO}_2\text{(ACE-FTS)} \tag{2}$$

$$\text{Cly(ACE-FTS)} = \text{HCl(ACE-FTS)} + \text{ClONO}_2\text{(ACE-FTS)}\,. \tag{3}$$

For ClO, values during daytime (SZAs less than 85°) are used (Fig. 4). $ClONO_2$ values for the LSTs of SMILES and MLS observations would be needed for a precise calculation of Cly. Since the $ClONO_2$ values measured by ACE-FTS at sunrise are higher than they would be during daytime, adding the ACE-FTS $ClONO_2$ value to the sum of ClO and HCl values for SMILES or MLS results in an overestimate of Cly values. Conversely, for ACE-FTS, we just take the sum of the HCl and $ClONO_2$ values, because ClO values at sunrise (SZA of 90°) are expected to be small (less than 0.05 ppbv, as was mentioned in Sect. 3). But, this does result in a slight underestimate of the ACE-FTS Cly values, especially above 600 K. Considering these two opposite directions, the HCl/Cly ratio difference between ACE-FTS and others above 600 K in the right panel of Fig. 5 can be partly explained. In addition, the vertical shape of the observed Cly values is consistent with deduced Cly values from N_2O measurements made by MLS and ACE-FTS (see Appendix A).

Although the contribution of HOCl to Cly should be discussed, the data quality of this version of HOCl has not been thoroughly evaluated. Thus, we do not address the diurnal cycle of HOCl here. More importantly, since nitrogen dioxide (NO_2) measured by ACE-FTS revealed a value of 2.7 ppbv on average in the 675–700 K bin, ClO observed in daytime could be converted to $ClONO_2$ during sunset. As mentioned in Sect. 2, the nighttime value of ClO was also almost zero. Therefore, the diurnal cycle of ClO from zero (nighttime) to 0.35 ppbv (14:00 LST) in the 675–700 K bin (26 km) should be mainly caused by the photolysis of $ClONO_2$.

Generally, the HCl/Cly ratios are in good agreement among the three sensors for all PT levels between 400 and 700 K. The HCl/Cly ratio is 0.91–0.95 in the peak HCl PT levels (450–500 K) from the three sensors. This strongly suggests that even in the late November period HCl dominates Cly inside the Antarctic vortex in the lower stratosphere, when the vortex situation was somewhat distinct compared to other years (de Laat et al., 2010). The result is consistent with the previous observations in November from the different sensors (Rinsland et al., 1996; Michelsen et al., 1999; Hayashida and Sugita, 2007) and the Aura MLS alone (de Laat and van Weele, 2011). The lower values of ClO in those PT levels also confirm that it was deactivated to reform HCl through the Reactions (R1)–(R4). The data quality of SMILES HCl in the lower stratosphere is sufficient to capture this high HCl/Cly ratio phenomenon.

5 Conclusions

SMILES measured several chemical species in the middle atmosphere between October 2009 and April 2010. It was onboard the ISS/JEM platform and successfully measured the low-noise emission spectra with a 4-K mechanical cooler and SIS mixers. We focus on HCl and ClO measured inside the late spring Antarctic vortex on 19–24 November 2009. The data are sorted by EqL fields with the aid of DMPs, and data inside the vortex are averaged. To confirm the quality of SMILES-JAXA and -NICT data products from the view point of partitioning of Cly, comparisons are made with Aura MLS and ACE-FTS satellite data taken in the same time period.

The SMILES HCl reveals 2.8–3.1 ppbv between 450 and 500 K, along with a result that the NICT product is, at the maximum, 0.5 ppbv larger than the JAXA product between 425 and 575 K. The SMILES-NICT HCl agrees within 10 % with the MLS HCl between 425 and 650 K and with the ACE-FTS HCl between 425 and 575 K, respectively. Above 575 K, the SMILES-NICT HCl values are, however, 16–22 % smaller than those from ACE-FTS. The SMILES-JAXA HCl is 10 to 20 % smaller than that from MLS between 400 and 700 K and ACE-FTS between 500 and 700 K, respectively.

For ClO in daytime (SZAs less than 85°), SMILES-JAXA and -NICT agree to within ±0.05 ppbv between 450 and 625 K, although the difference between the two products amounts to −0.07 ppbv at 625–675 K and 0.07 ppbv at 425 K. The difference between MLS and SMILES-JAXA or -NICT was less than ±0.05 ppbv between 500 and 650 K with ClO values less than 0.2 ppbv.

Considering the low ClONO$_2$ values also inside the Antarctic vortex as measured by ACE-FTS, HCl was the main component of Cly below the 500 K level in November 2009. All three sensors show high values ($>$ 0.9) of the HCl/Cly ratios, in agreement with the past observations inside the spring Antarctic vortex. These results can be useful for assessing the performance of recent Chemistry-Climate Models (CCM) in CCM validation activities (e.g. Fig. 6.32 of SPARC CCMVal, 2010). The results from SMILES-JAXA (v2.1) and -NICT (v2.1.5) suggest the validity of both HCl and ClO data in the lower stratosphere, where HCl values were as high as 3 ppbv during the period studied here.

Appendix A

Comparison between Cly and Cly*

The measured Cly values are compared with a surrogate for Cly, which is referred to as Cly*. Figure A1 shows vertical profiles of N$_2$O measured by Aura MLS and ACE-FTS (left panel). The right panel of Fig. A1 shows observed Cly values from SMILES, MLS, and ACE-FTS (see Sect. 4.3) and the Cly* values. The Cly* values are deduced from the N$_2$O data from MLS and ACE-FTS and a polynomial expression between Cly* and N$_2$O (Wetzel et al., 2010). This expression is based on in-situ balloon observations of N$_2$O and several organic chlorine species (Engel et al., 2006), and is dedicated to the Arctic stratosphere for mean age of air less than 6 yr (Engel et al., 2002). The retrieval method and validation results for MLS and ACE-FTS N_2O measurements are described in Lambert et al. (2007) and Strong et al. (2008), respectively. Precision and accuracy of the version 3.3 MLS N_2O data are estimated to be 8–9 % and 13–25 %, respectively, for 46.4–100 hPa levels (Livesey et al., 2011). Fitting errors for the version 2.2 ACE-FTS N_2O data are estimated to be 4 % for 5–35 km (Strong et al., 2008).

The Cly* values calculated from the expression are then decreased to the year 2009 as was done in Wetzel et al. (2012), since the expression is valid for the years 2000–2003. It should be noted that recent satellite measurements of SF_6 from the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) onboard Envisat suggested an inter-hemispheric difference in “apparent age” of air inside the Arctic and Antarctic vortices, being older ages in the Antarctic than in the Arctic (Stiller et al., 2008, 2012). For N_2O data less than 20–30 ppbv (above $\sim$ 500 K), they could be influenced by intrusions of mesospheric air into the Antarctic vortex. The real age of air in the Antarctic vortex might be different from that in the Arctic, making the Cly* values estimated uncertain. Thus, the Cly* values presented here are solely reference values.

Absolute differences between MLS and ACE-FTS N_2O data amount to 30 ppbv. It could be partly due to a different degree of descent of air. Such differences reflect the Cly* values; those from MLS are larger than those from ACE-FTS below 500 K levels. The Cly and Cly* values from ACE-FTS (purple and gray lines) agree well below 500 K levels. The result presented here provides some sort of uncertainty range for the Cly level in the Antarctic stratosphere in this time period.

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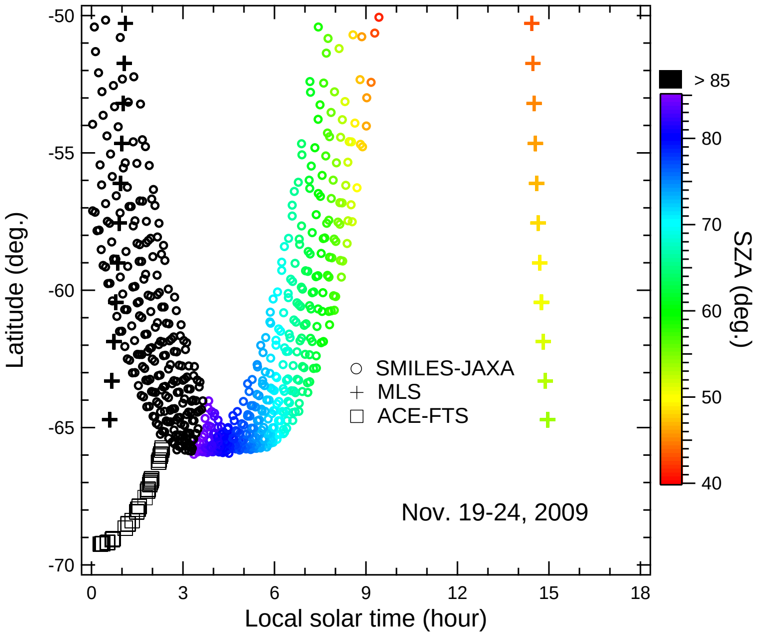

Fig. 1. Measurement latitude as a function of local solar time for SMILES (open circles), MLS (crosses), and ACE-FTS (open squares). Solar zenith angle is color-coded between 40 and 85°. Above 85°, the symbols are colored in black. Data only inside the Antarctic vortex (within the edge center) between 19 and 24 November 2009 are used.

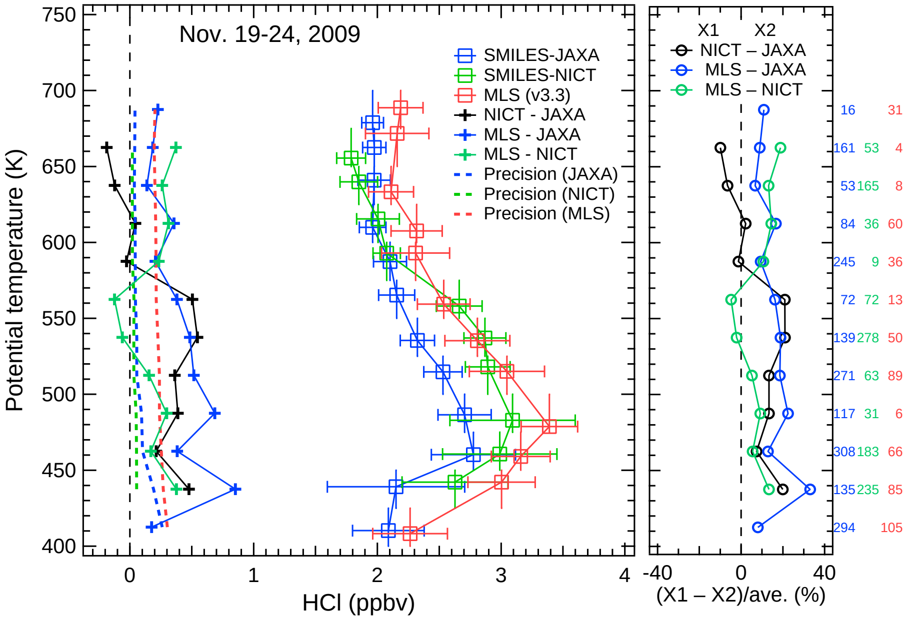

Fig. 2. Vertical profiles, as a function of potential temperature (PT), of HCl volume mixing ratios as measured by SMILES and MLS on 19–24 November 2009 inside the vortex (within the edge center). The left panel shows average profiles of HCl in each 25 K PT bin (each vertical bar corresponds to 25 K) for the SMILES-JAXA (blue), -NICT (green), and MLS (red) data products. Squares and horizontal bars show average and $\pm 1\sigma$ standard deviations, respectively. Dashed lines show reported precisions. The black crosses show the absolute difference between the two SMILES products. The blue and green crosses show the absolute difference between the MLS and SMILES-JAXA or -NICT, respectively. The right panel shows the relative difference with regard to the absolute difference divided by its mean value of SMILES-JAXA and -NICT, MLS and SMILES-JAXA, or MLS and SMILES-NICT products. The numbers of data used in the averages are also listed outside of the right panel: SMILES-JAXA (blue), -NICT (green), and MLS (red).

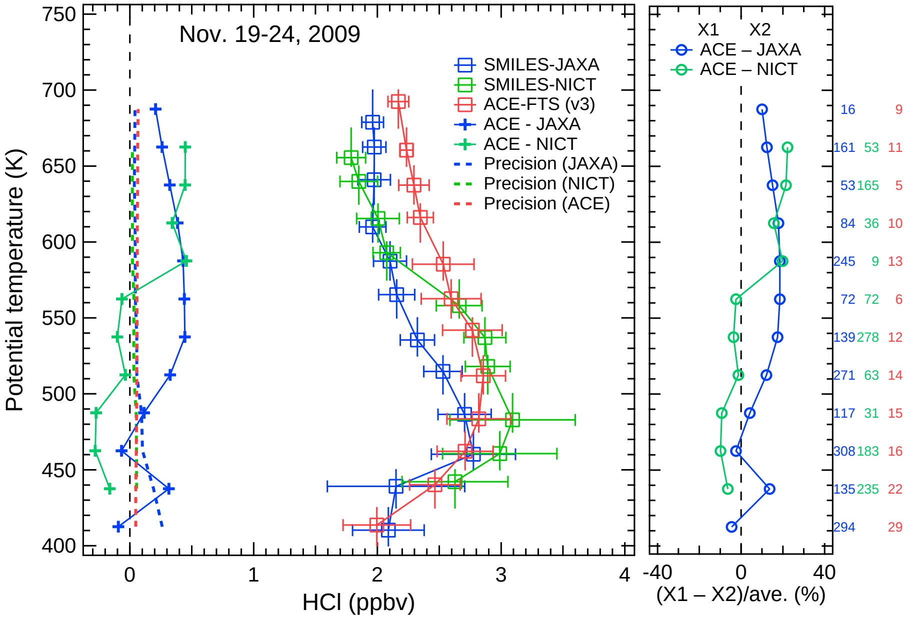

Fig. 3. Same as Fig. 2, but for SMILES and ACE-FTS. The absolute and relative differences between SMILES-JAXA and -NICT are omitted.

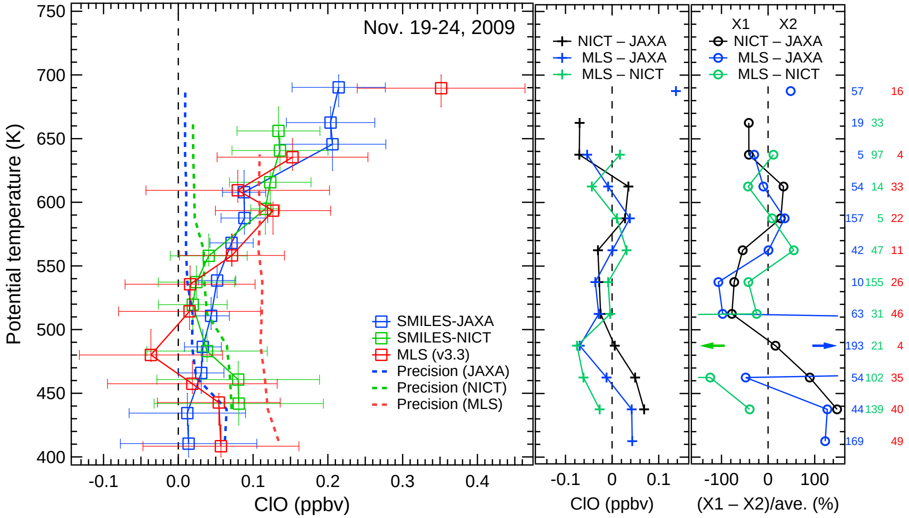

Fig. 4. Same as Fig. 2, but for ClO volume mixing ratios as measured by SMILES and MLS. For clarity, absolute differences are shown in the middle panel. Measurements with solar zenith angles less than 85° (daytime) are used. Arrows in the right panel show larger relative differences owing to the averages close to zero.

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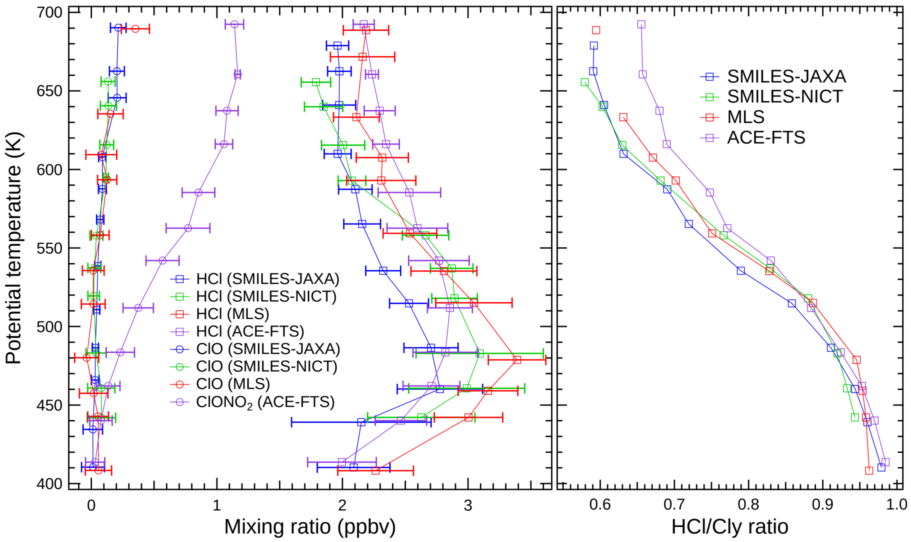

Fig. 5. Vertical profiles of ClO (measured by SMILES and MLS), ClONO$_2$ (measured by ACE-FTS), and HCl (measured by SMILES, MLS, and ACE-FTS) on 19–24 November 2009 inside the Antarctic vortex (within the edge center) as a function of potential temperature (left panel). Horizontal bars show ±1σ standard deviations of the averages. HCl/Cly ratios computed for each of the three instruments are shown in the right panel. For a derivation of Cly, see Sect. 4.3.

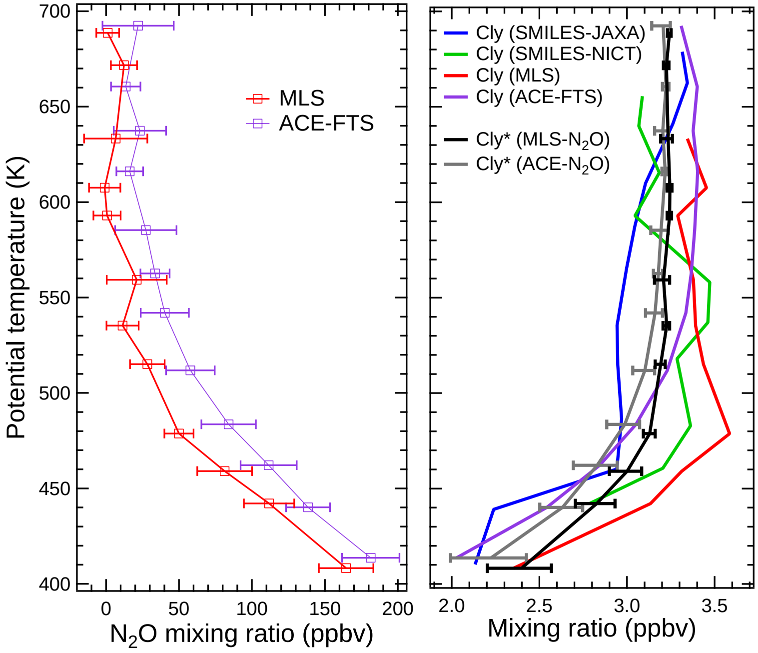

Fig. A1. Vertical profiles of N_2O measured by MLS (red) and ACE-FTS (purple) on 19–24 November 2009 inside the Antarctic vortex (within the edge center) as a function of potential temperature (left panel). The average and ±1σ standard deviation in each 25 K PT bin are shown. Observed Cly values calculated from SMILES, MLS, and ACE-FTS (see Sect. 4.3) and the Cly* values deduced the N_2O values from MLS and ACE-FTS (see Appendix A) are shown in the right panel. Bars show corresponding Cly* values estimated from the standard deviations of N_2O data.