P11.198 Diurnal variations of stratospheric/mesospheric trace species, ClO, BrO, and HO_2 derived from 4K cooled submm limb sounder ISS/JEM/SMILES

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1. INTRODUCTION

The Superconducting Submillimeter-Wave Limb-Emission Sounder (SMILES) (Kikuchi et al., 2010) is one of the early science program onboard Japanese Experimental Module/Exposed Facility (JEM/EF) of the International Space Station (ISS). SMILES has been developed jointly by JAXA and NICT. SMILES measures Earth's limb in the 625-650 GHz frequency region by using 4-K cooled Superconductor-Insulator-Superconductor (SIS) junction device, which results $T_{sys} \simeq$ 400K and *NEDT* $\simeq$ 0.4 K, i.e. 10 times higher sensitivity compared to previous limb atmospheric measurements in the mm/submm wavelength region, such as AURA/MLS (Waters et al., 2006) and ODIN/SMR(Frisk et al., 2003).

SMILES can measure several key species related to chemistry of O_3 layer; O_3 (O_3, ^{17}OOO, $O^{17}OO$, ^{18}OOO, and $O^{18}OO$), HCl ($H^{35}Cl$ and $H^{37}Cl$), ^{35}ClO, HO_2, $HO^{35}Cl$, ^{81}BrO, and CH_3CN. The 350-400 km and 52° inclined ISS orbit shows 88-90 days precession period, which means that SMILES can measure diurnal variation of ClO, HOCl, HO_2, BrO and mesospheric O_3 within 45 days observation. The high sensitivity (T_{sys}~400 K) and diurnal variation of the SMILES should open opportunity to investigate details of photochemistry of stratosphere and mesosphere which have not been investigated well before.

Profiles of SMILES data (O_3, HCl, etc), i.e. Level 2 (L2) data, are expected to have better precision (smaller error bar) compared to previous satellite measurements, because of better T_{sys} (~400K) and $NE\Delta T$ (~0.4 K) of the SMILES instrument. Accuracy of SMILES L2 data may not be as good as its precision, due to many reasons; such tangent height determination (Verdes, 2002; Takahashi et al., 2010), poor knowledge on instrument characteristics, uncertainty of spectroscopic errors (Verdes et al., 2005A and 2005B), approximations of algorithms, etc.

It is well known that the trace species, such as ClO, BrO, HOCl, HO_2, are controlling the photochemistry of stratosphere and mesosphere with diurnal variation. Observations of these species have been difficult due to reasons; 1) weak signals require integration for satellite observations, 2) limited local time from sun-synchronous satellites, and/or 3) limited space/time coverage from balloon instruments with enough sensitivity. The sensitivity of SMILES can provide opportunity to derive single scan retrieval of ClO, BrO, HOCl, and HO2. By plotting these species over 30-45 days, it is possible to derive diurnal variation of these species in the stratosphere and/or mesosphere.

This paper describes current status of SMILES ClO, BrO, HO_2 retrieval, their validation, features in diurnal variation.

2. SMILES OBSERVATION

2.1 OBSERVATION

SMILES was launched on Sep. 11, 2009 as a cargo payload of H-II Transportation Vehicle (HTV) by using H-IIB launcher from Tanegashima Space Center. SMILES have been operated from Oct. 12, 2009 to Apr. 21, 2010 when the submm local oscillator of the SMILES aborted operation.
Orbit characteristics of the ISS is 350-400 km in altitude and 52° inclination. Attitudes of ISS have varied frequently as much as ± 5° for pitch, yaw, and roll angles due to docking/undocking and other events.

SMILES observes 45° left from the forward direction of the ISS, which results nominal geographical coverage 38°S-65°N (Fig.1). Scanning antenna covers -30 to +160 km altitude region at tangent points. The observation cycle is 53s; the first 29.5 s is atmospheric measurement of ~2 km altitude interval at the tangent point in 0.5 s step. After the 29.5 s, the antenna was scanned quickly to the 160 km for the cold sky calibration. Internal room temperature hot load calibration and the frequency calibration have been carried out at every 53 s observation period.

Due to the precession of the ISS orbit, ISS orbit plane rotates with a period of 88-90 days against Sun. Thus, it is possible to plot the diurnal variation of observed parameters by using 45 days data set. Figure 2 shows local time coverage by using data in January 2010.

Figure 3 shows the expected performance of the SMILES assuming the T_{sys} ~ 500 K (design target) for the mid-latitude atmosphere (Buhler et al, 2005; Takahashi et al., 2010; Kikuchi et al., 2011). In this estimation, covariance of *a priori*, $\boldsymbol{S_a}$, is assumed to be 100%, which means Error ratio S/S_a = 1.0 is equivalent to 100% error bar. O_3 and HCl can be retrieved <10% precision in the 20-65 km altitude range. Precision of HNO_3 retrieval can be expected better than 50% at the altitude region 20-35 km, but it can be improved by applying proper *a priori* restriction as described in the later section. Weaker species, such as BrO, can be retrieved ~ 50% in 30-40 km altitude range.

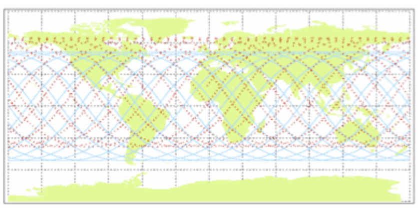


Fig. 1 Geographical Coverage of SMILES (red) and ISS nadir trajectory (blue)

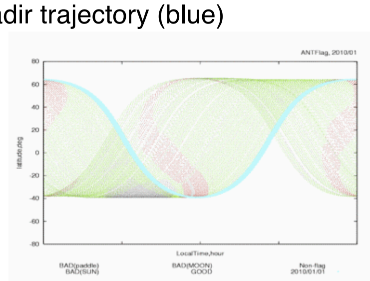


Fig. 2 Actual local time coverage of SMILES in January 2010 (blue: Jan 1st one day, green: all other days, red: not observed).

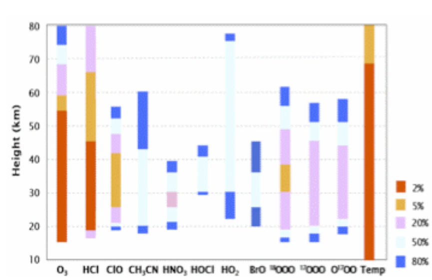


Fig. 3 Expected retrieval precision by considering only instrument noise.

2.2 INSTRUMENT

SMILES consists of (1) Scanning Antenna, (2) Single Side Band (SSB) Separator, (3) Local Oscillator, (4) SIS mixer, and (5) Acousto-Optic Spectrometer (AOS). Table 1 lists the major characteristics of the SMILES system.

Table 1. Characteristics of SMILES instrument.

Frequency coverage	Band A (624.32 – 625.52 GHz) Band B (625.12 – 626.32 GHz) Band C (649.12 – 650.32 GHz)
Freq. sampling	0.8 MHz
Freq. resolution	1.8 MHz (FWHM)
System noise temp.	350~400 K in orbit
Integration time	0.5 s for each obs. tangent point
Noise tquivalent brightness temperature	~ 0.4 K in orbit
Calibration accuracy	< 1.0 K (specification)
Obs. cycle	53 s
Obs. alt. range	0 - 160 km
Vertical sampling	~2 km *(nominal)*
Instrumental vertical resolution	3.5 - 4.1 km *(nominal)*
Latitude coverage	38°S - 65°N *(nominal)*

The SMILES IFOV pointing knowledge is given by the combination of (a) angle resolver of the primary telescope (0.0015° precision, i.e. 60 m at tangent point), (b) attitude of SMILES instrument determined by the 2 star sensors of SMILES (0.006° precision, i.e. 210 m at tangent point), and/or the ISS attitude telemetry data in the Erath Centered Inertial coordinate system. Since the ISS orbit motion (~90 minutes period), exact pointing knowledge requires accurate timing knowledge, 90 ms precision, which is equivalent to the 0.006° attitude knowledge. The ISS attitude is determined by the triangulation using three GPS receivers at the ISS truss structure far from JEM/SMILES. There are two other star sensors within JEM, one is for MAXI instrument next to SMILES, and the other is Inter satellite Communication System (ICS).

Altitude of tangent point is calculated from combinations of (a) ISS altitude telemetry data (< 100 m precision), (b) ray tracing through the geodetic spheroid with atmosphere. The tangent height knowledge should be determined in 340 m precision, if neglecting error in timing of attitude determination. This pointing/tangent height knowledge is only determined to the mechanical axis of the primary mirror, and there can be difference between true antenna axis and mechanical axis.

The primary mirror of the antenna is elliptical, 40 cm in vertical and 20 cm in horizontal, and the antenna beam pattern is elliptical, 0.09° in elevation and 0.18° in horizon, respectively. ISS is 3-axis stabilized satellite, but its attitude might change significantly ±15° in pitch, yaw, and roll (nominal specification). During the SMILES operation period, ISS attitude ranged within ±5° in pitch, yaw, and roll axis. This attitude envelope should be considered in the data retrieval.

2.3 RETRIEVAL ALGORITHMS

The retrieval algorithm (Takahashi et al. 2010; Imai et al., 2010) of operational L2 processing system is based on the OEM applied for atmospheric sounding (Rodgers, 1976). The maximum a posteriori estimate can be derived from statistical combination of a priori knowledge of a state vector $\boldsymbol{x}$ and the information on the measurement. We use a modification of the Gauss-Newton method called the Levenberg-Marquardt method (Levenberg, 1944; Marquardt, 1963). The retrieved state vector $\boldsymbol{x_{i+1}}$ at the iterative step i+1 is calculated as

$$x_{i+1} = x_i + \left(K_{xi}^T S_y^{-1} K_{xi} + S_a^{-1} + \gamma D\right)^{-1} \left\{K_{xi}^T S_y^{-1}\left[y - F\left(x_i, b\right)\right] + S_a^{-1}\left(x_i - x_a\right)\right\}, \tag{1}$$

where, $\boldsymbol{x}$ is a state vector of length $\boldsymbol{n}$, which contains concentrations of the species, atmospheric temperature, and pointing offset. The $\boldsymbol{y}$ is a measurement vector of length $\boldsymbol{m}$, which denotes the calibrated brightness temperature observed by the SMILES, and $\boldsymbol{F}$ is a forward model including both atmospheric radiative transfer and instrument characteristics. ***A priori*** knowledge is represented by the expected state $\boldsymbol{x}$ and its covariance matrix $\boldsymbol{S_a}$. $\boldsymbol{S_y}$ is the covariance matrix of $\boldsymbol{y}$. The matrix $\boldsymbol{K_{xi}}$ is a weighting function for each of the retrieval parameters evaluated at $\boldsymbol{x_i}$. The $\boldsymbol{x_a}$ normally corresponds to the initial guess $\boldsymbol{x_0}$, $\boldsymbol{D}$ is a scaling matrix that is usually assumed to be $\boldsymbol{S_a^{-1}}$, and $\boldsymbol{g}$ is a Levenberg-Marquardt parameter which is initially set to 10^{-3} in the released two datasets (vers. 005-06-0024, and 0032), respectively.

A priori value and their covariance have been prepared from monthly zonal mean (10° latitude bin) of Aura/MLS. Initial value of temperature, humidity, and pressure are meteorological data provided by NASA/GSFC (6 hours interval) (Rienecker, M. M. et al., 2007).

In the present study, SMILES ver.005-06-0150 (here after ver. 1.2) has been used. Major differences of the versions 0024 and 0032 from the algorithm theoretical basis document (ATBD) work (Takahashi et al., 2010) are initial tangent altitude guess. In the ATBD algorithm, initial tangent heights can be estimated within ±1km by using ancillary data. In the ver. 0024, initial tangent height is calculated from the smoothed value of Star Sensor of SMILES, and the line parameter of O_3, HCl are updated based upon the recent comparison (Perin et al., 2005). In the ver.0032, initial tangent heights are calculated using both Star Sensor of SMILES and the attitude data of ISS.

3. SMILES PRODUCT OVERVIEW

3.1 O_3, HCl, ClO AND HNO_3

Three data sets have been released until Sep. 2010. These differ only slightly; 1) initial tangent height estimation, 2) Spectral response function (FWHM), and there have been no major changes from ATBD before launch. Table 2 lists version numbers of three data sets. Internal version number gives information of sub system versions; (L0_data)-(L1B_data)-(L2_processing).

Table 2 SMILES operational Data set releases.

Version	Internal Version	Release Date
1.0	005-06-0024	Feb. 2010
1.1	005-06-0032	May 2010
1.2	005-06-0150	Sep. 2010

Validation of major species showed quite good agreements with Aura/MLS, SciSAT-1/ACE-FTS, and ENVISAT/MIPA (Suzuki 2010, Shiotani 2010). Validation of weak signal species (HO_2, BrO, HOCl, O_3-isotopes) are underway, but it looks SMILES has better sensitivity and/or precision against validation counter part. And simple validation comparison may not work for SMILES weak signal species.

3.2 CURRENT ISSUES

Currently we found several major issues regarding SMILES L1B data, and it looks SMILES L2 data set must be upgraded using upcoming corrected L1B data, before concluding validation studies and starting scientific works.

- Nonlinearity correction of RF signal chain and AOS.
- Spectral response function of AOS.

- Frequency calibration of AOS.
- Unidentified short term calibration error within 53 sec observation cycle.

Nonlinearity correction has not been applied to released L2 data sets (versions 1.0-1.2). It has been found that these L2 data sets showed similar negative temperature bias (-2% or -5K) against GMAO weather data and SABER temperature at the stratosphere. But if we apply non-linearity correction as a trial, this temperature bias disappeared. We expect products other than O_3 and HCl should have been showing 10-20% higher value due to “without non-linearity correction”.

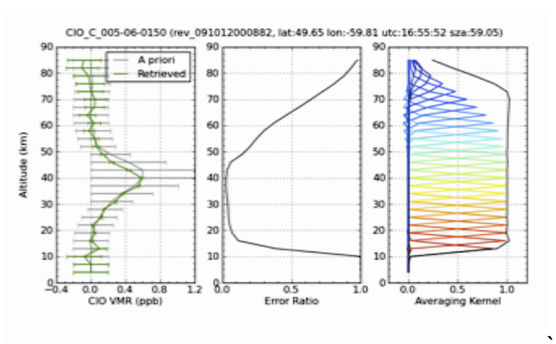
We found that spectral response function and frequency calibration of AOS have strong impact (as much as ±30%) to the SMILES data in high altitude (> 50 km). We have been conducting detailed investigation of these issues by using pre-launch test data, in-orbit calibration data, as well as trial tunings of L2 algorithm.

Recently, new issue ‘short term calibration error’ has found and it looks affecting all weak signal species at all altitude region. HOCl is not discussed in this paper, which showed unacceptable value in the versions 1.0-1.2 due to this short term calibration issue.

4. ClO

4.1 ClO RETRIEVAL

SMILES can measure ^{35}ClO at Band C. ClO has strong emission in the SMILES observation and it is relatively easy to retrieve. Fig. 4 shows ClO retrieval example in daytime and nighttime.



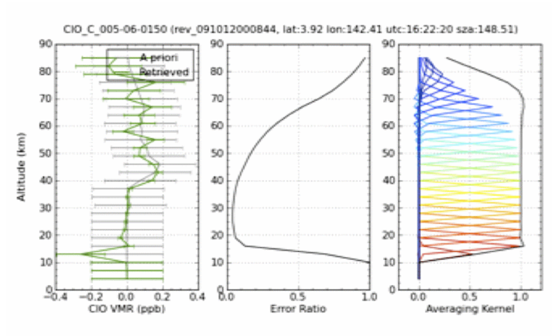


Figure 4 Retrieval example of ClO in daytime (upper) and nighttime (lower).

4.2 ClO VALIDATION STATUS

Validation of SMILES ClO has been carried out tentatively using ver. 1.2 against with coincidences of Aura/MLS ver 2.2 (Figs. 5 and 6). It looks agreement with MLS is acceptable, and formal validation studies are on-going with Aura/MLS team. The SMILES ClO L2 ver 1.2 data should be useful for scientific studies with some cautions at upper and lower altitude regions where SMILES L2 data is not compared with Aura/MLS.

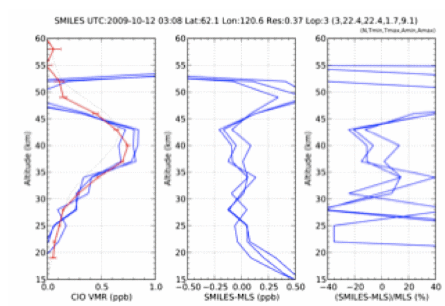


Figure 5. Coincidence event on Oct. 12, 2009 SMILES (red), MLS (blue lines) at 62.1°N 120.6°E (The first coincidence).

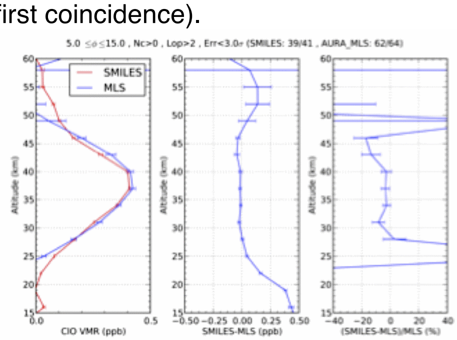


Figure 6. Coincidence Statistics 5N-15N, 39 SMILES vs. 62 MLS profiles. < 10% agreement with MLS 2.2 between 25-42 km.

4.3 ClO DIURANAL VARIATION

ClO diurnal variation can be plotted by using 30-45 days data as described above. Fig. 7 shows ClO diuranal variation at in the equatorial region.

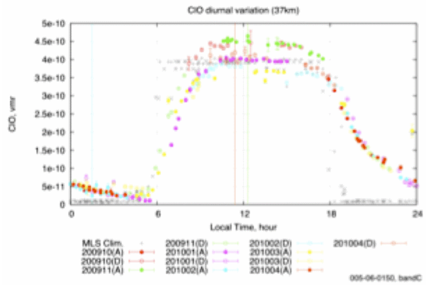


Figure 7. ClO diurnal variation at 37 km in the equatorial region (10N-10S) observed by SMILES.

5. BrO

5.1 BrO RETRIEVAL

SMILES is not specifically designed to measure BrO. BrO can be measured much easily at the other submm frequency regions (Urban 2003). SMILES Bands A and B cover weak BrO lines superposed by stronger emission of other molecules. Only Band C BrO covered by strong O3 isotope lines can be retrieved as shown in Fig. 8. Figure 9 shows retrieval example of BrO in daytime.

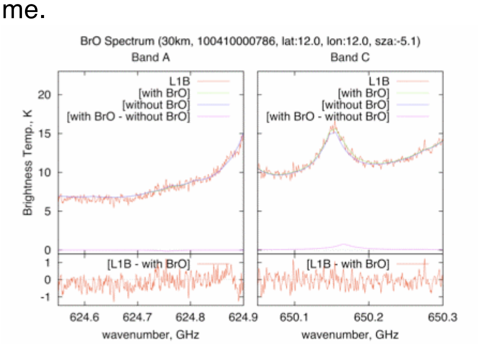


Figure 8. SMILES BrO Observation in Band A and C. (Upper: L1B spectrum and L2 fit, Middle: calculated BrO spectrum component, Bottom: noise residual).

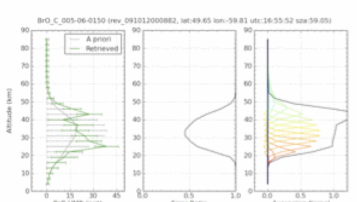


Figure 9. Retrieval example of BrO in daytime.

5.2 BrO VALIDATION STATUS

SMILES BrO showed larger BrO value compared to previous model and remote sensing studies. Validation of BrO will be carried out after the L2 update (after L1B update) in near future.

5.3 BrO DIURANAL VARIATION

Figure 10 shoes diurnal variation plot of BrO at 31 km altitude in the equatorial region. It should be very cautious on the absolute value of this plot, but temporal change looks to be acceptable. BrO is photolised around local time noon and shows small dip as predicted by the model studies.

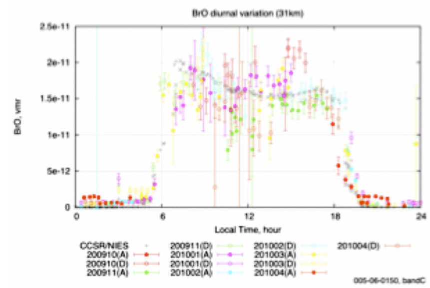


Figure 10. Diurnal variation plot of BrO at 31 km in the equatorial region (N10-S10).

6. HO_2

6.1 HO_2 RETRIEVAL

SMILES can measure HO2 both in Band B and C. Figure 11 shows detailed fitting example at Bands B and C. Currently we recommend to use Band C HO2 since spectral baseline around HO2 emission line is more flat in Band C, which should give better retrieval quality. Fig. 12 shows retrieval example of HO_2 in the daytime.

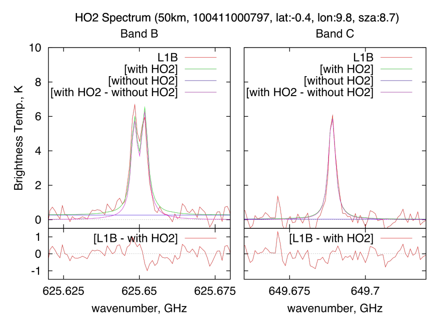


Figure 11. Spectral fitting of HO_2 in the Bands B and C.

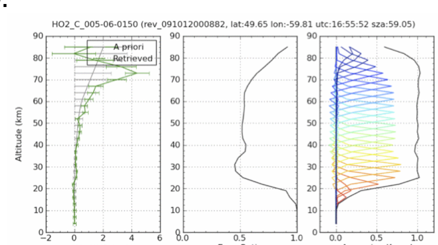


Figure 12. Retrieval example of HO_2 in daytime (Oct. 12, 2009, SZA=59.1°, 49.7°N)

6.2 HO_2 VALIDATION STATUS

HO_2 validation is under going with coincidences against Aura/MLS. Figure 13 shows coincidence statistics at the N5-S5 latitude region. Although it agrees quite well with Aura/MLS at the peak (75km), the difference in the 55-65km is large. And the coincidence statistics are slightly worth in the other latitude regions. So, we recommend not to use SMILES HO_2 before the conclusion of further validation studies.

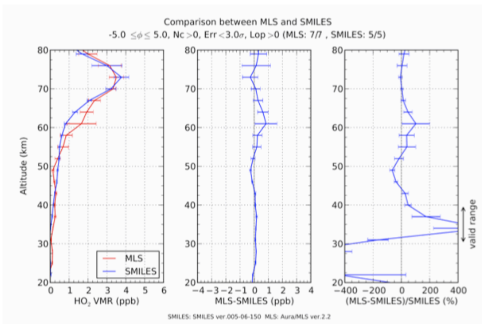


Figure 13 Coincidence statistics of SMILES Band C HO_2 with Aura/MLS in the N5-S5 latitude region.

6.3 HO_2 DIURANAL VARIATION

Figure 14 shows diurnal variation plot of HO_2 at 40 km in the equatorial region (N5-S5). The absolute value needs further validation and algorithm studies, the temporal feature of HO_2 can be compared with model studies.

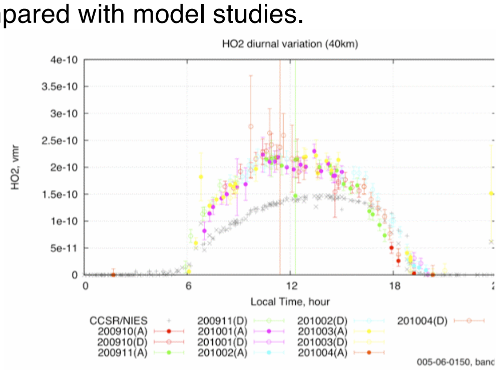


Figure 14. Plot of HO_2 diurnal variation using Band C data in the equatorial region (N5-S5).

7. SUMMARY

SMILES showed excellent sensitivity as expected from its performance of 4K cooled SIS mixer and AOS spectrometer. Data retrieval studies and validation have been on-going right now.

Extensive caution on L1B data should be paid to obtain high quality L2 data, as expected from the instrument performance. There are several L1B issue and L2 retrieval issue in the SMILES data products. O_3, HCl, and ClO products look quite good and there can be applied to scientific studies, with special cautions.

SMILES BrO and HO2 can be retrieved as expected by the theoretical calculation. These products must be verified by further studies. Diuranal variation of ClO, BrO, and HO_2 (HOCl and mesospheric O_3 are also possible) can be plotted by using 30-45 days SMILES data. And precision of these plots should open new dimension of understandings to the chemistry of atmosphere.

8. REFERENCES

Buehler, S. A. et al. (2005), Expected performance of the Superconducting Submillimeter-Wave Limb Emission Sounder compared with aircraft data, *Radio Sci.*, *40*, 3, 1-13, RS3016,

Von Clarmann, T. et al. (2009), Retrieval of temperature, H_2O, O_3, HNO_3, CH_4, N_2O, $ClONO_2$ and ClO from MIPAS reduced resolution nominal mode limb emission measurements., Atmos. *Meas. Tech. Discuss.,* 2, 181–236.

Dupuy et al. (2009), Validation of ozone measurements from the Atmospheric Chemistry Experiment (ACE). *Atmos. Chem. Phys.,* 9, 287-343

Frisk et al., (2003), The Odin satellite. I. Radiometer design and test, *Astronomy & Astrophysics*, 402, 3, 27-34

Froidevaux et al. (2008A), Validation of Aura Microwave Limb Sounder stratospheric ozone measurements. *J. Geophys. Res.,* 113, D15S20, 1-24.

Froidevaux et al. (2008B), Validation of Aura Microwave Limb Sounder HCl measurements, *J. Geophys. Res.*, *113*, D15S25, 1-23.

Imai, K., M. Suzuki, and C. Takahashi (2010), Evaluation of Voigt algorithms for the ISS/JEM/SMILES L2 data processing system, *Advances in Space Research*, *45*, 669-675.

Inatani,. J. et al., (2000), Submillimeter Limb-emission Sounder JEM/SMILES aboard the Space Station", *Proc. SPIE, 4152*, 243-254.

Kikuchi, K., et al. (2010), Overview and early results of the Superconducting Submillimeter-Wave Limb-Emission Sounder (SMILES), *Journal of Geophysical Research*, submitted.

Mahieu et al. (2008), Validation of ACE-FTS v2.2 measurements of HCl, HF, CCl_3F and CCl_2F_2 using space-, balloon-and ground-based instrument observations. *Atmos. Chem. Phys.*, *8*, 6199-6221.

Masuko, H., S. Ochiai, Y. Irimajiri, J. Inatani, T. Noguchi,

Y. Iida, N. Ikeda, N. Tanioka (1997), A Superconducting Sub-millimeter Wave Limb Emission Sounder (SMILES) on the Japanese Experimental Module (JEM) of the Space Station for Observing Trace Gases in the Middle Atmosphere, *Proceedings of Eighth Int. Symp. on Space Terahertz Tech.*, Harvard Univ., pp. 505-517.

Mencaraglia et al. (2006). Validation of MIPAS satellite measurements of HNO_3 using comparison of rotational and vibrational spectroscopy. *J. Geophys. Res., 111,* D19305, 1-8.

Perrin et al. (2005), Molecular Line Parameters for the “MASTER” (Millimeter Wave Acquisitions for Stratosphere/Troposphere Exchange Research) Database. J Atmos Chem, 51 (2), 161-205

Rienecker, M. M. et al. (2007), The GEOS-5 Data Assimilation System - Documentation of versions 5.0.1, 5.1.0, and 5.2.0. Technical Report Series on Global Modeling and Data Assimilation NASA/TM-2007-104 606, 27, edited by M. J. Suarez.

Rodgers, C. D. (1976), Retrieval of atmospheric temperature and composition from remote measurements of thermal radiation, *Rev. Geophys. Space Sci.*, *14*, 4, pp. 609-624.

Santee M. L. et al. (2007), Validation of the Aura Microwave Limb Sounder HNO_3 measurements, *J. Geophys. Res., 112,* D24S40, 1-22.

Shiotani et al. Recent results from the Superconducting submillimeter-wave limb-emission sounder (SMILES) onboard ISS/JEM. Proc. SPIE (2010) pp. 1-13
SMILES Science Team and SMILES Mission Team, 2002. *JEM/SMILES Mission Plan Version 2.1*.

Suzuki et al. EARLY RESULTS FROM 4K-COOLED SUPERCONDUCTING SUBMM WAVE LIMB EMISSION SOUNDER SMILES ONBOARD ISS/JEM. International Archives of the Photogrammetry, Remote Sensing and Spatial Information Science (2010) vol. 38 (8) pp. 104-109

Takahashi, C., et al. (2010), Operational retrieval algorithms for JEM/SMILES level 2 data processing system, *J. of Quantitative Spectroscopy & Radiative Transfer, 111*, 160–173.

Urban J., Optimal sub-millimeter bands for passive limb observations of stratospheric HBr, BrO, HOCl, and HO2 from space. Journal of Quantitative Spectroscopy and Radiative Transfer (2003) vol. 76 (2) pp. 145-178.

Verdes C. (2002), Deriving Atmospheric Temperature and Instrumental Pointing from Millimeter/Sub-millimeter Limb Sounding Measurements. Thesis, IUP Bremen, pp. 1-131.

Verdes et al. (2005A), A sensitivity study on spectroscopic parameter accuracies for a mm/sub-mm limb sounder instrument. Journal of Molecular Spectroscopy, 229 (2), 266-275

Verdes et al. (2005B), Partition function data and impact on retrieval quality for an mm/sub-mm limb sounder. *J. of Quantitative Spectroscopy and Radiative Transfer*, 90 (2), 217-238.

Waters, J.W., et al. (2006), The Earth Observing System Microwave Limb Sounder (EOS MLS) on the Aura satellite, *IEEE Trans. Geosci. Remote Sensing* 44, no. 5.

Wolff et al. (2008), Validation of HNO_3, $ClONO_2$, and N_2O_5 from the Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS). *Atmos. Chem. Phys.*, 8, 3529-3562.