Chemistry of stratosphere and mesosphere revealed by ISS/JEM/SMILES for Earth Diagnosis.

Makoto SUZUKI[1], Naohiro Manago[1]

(JAXA/ISAS, Chiba U./CeRES)

Naohiro Manago[1], Chihiro MITSUDA[2], Koji IMAI[3], Hideharu AKIYOSHI[4], Takuki SANO[1], Yoko NAITO[5], D. Kinnison[6], and Masato SHIOTANI[7]

[1]Institute of Space and Astronautical Science, JAXA

[2]Fujitsu FIP, [3]Tome R&D Inc.

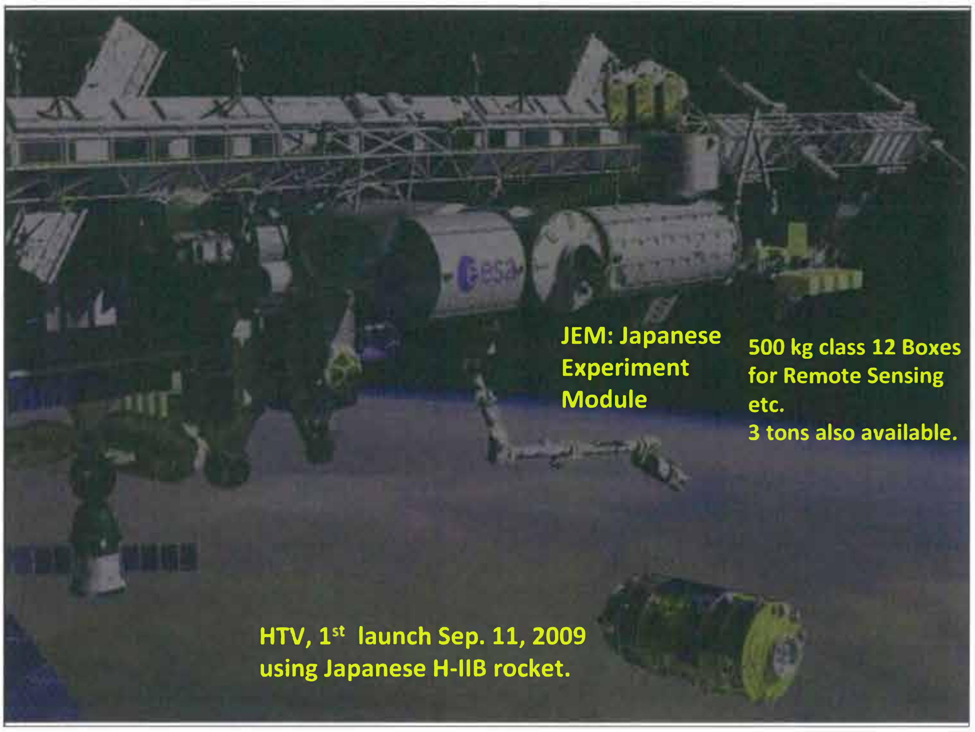
4National Institute for Environmental Studies

[5]Department of Geophysics, Kyoto University, [6]NCAR

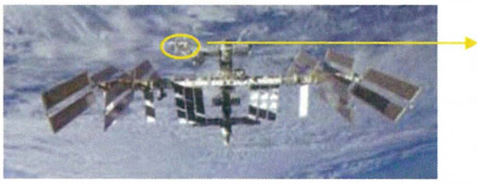
[7]Research Institute for Sustainable Humanosphere, Kyoto Universit y

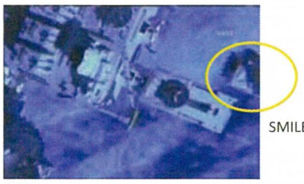
Passive microwave remote sensing in Japan

- AMSR, AMSR-E have shown excellent performance for remote sensing of SST, sea ice, etc.
- AQUA/AMSR-E had stopped operation last year, but AMSR-2 instrument and GCOM-W satellite is now waiting for launch (it was delayed from Feb. 2012, by the delay of another satellite provider).
- A sub-mm chemistry mission, SMILES had been conducted jointly by JAXA and NICT. It proposed around 1987 originally, and it finally onboard ISS on Sep. 18, 2009. It worked only 6 months, but it demonstrated breakthrough performance at 625, 650 GHz region, Tsys = 340K, using 4 K cooled SIS detector.
  - This is the my 3rd instruments that worked only 6 months.
    - ADEOS-I/ILAS, ADEOS-II/ILAS-II, ISS/JEM/SMILES (this talk)
    - And I suffered another satellite failure, not entering Venus orbit, on Dec. 9, 2010, but it failed. Venus Climate Orbiter, Akatsuki, is now wandering around 0.6-0.7 AU to reach Venus again around 2015/16.



JEM/SMILES Payload



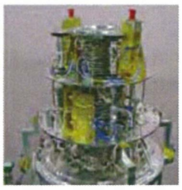


The SMILES was carried by the H-IIB with the H-II Transfer Vehicle (HTV) (Sep. 11); the HTV was attached to the ISS (Sep. 18); the SMILES was attached to the JEM (Sep. 25) (All dates in JST)

JEM/SMILES Mission

(JEM/SMILES: Superconducting Submillimeter-Wave Limb-Emission Sounder designed to be aboard the Japanese Experiment Module on ISS; Collaboration project of JAXA - Japan Aerospace Exploration Agency - and NICT - National Institute of Information and Communications Technology -)

1. Demonstration of superconductive mixer and 4-K mechanical cooler for the submillimeter limb-emission sounding in space



[Mechanical Cooler] Two-stage Stirling and J-T; 20mW @4K, 200mW @20K, 1000mW @100K; Power Consumption: <300 W; Mass: 90 kg

[SIS Mixer] RF: 640 GHz, IF: 11-13 GHz; Junction: Nb/AlOx/Nb, ~7 kA/cm^2; Fabricated at Nobeyama RO

2. Observation on atmospheric minor constituents in the middle atmosphere

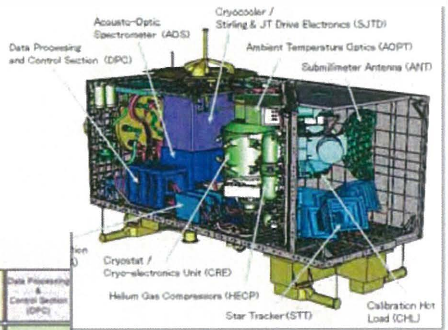
[Standard Products]

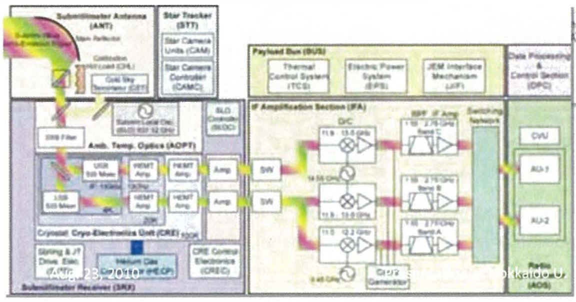
- 1 scan: O_3, HCl, ClO, CH_3CN, O_3 isotopes, HOCl, HNO_3
- Multi-scan: HO_2, BrO

[Research Products] UTH, Cirrus Clouds, volcanic SO_2, H_2O_2

SMILES Instrument

625, 650 GHz region, SSB system
4 K cooled, Tsys = 340 K
Two 1.2 GHz bandwidth spectrometers
0.8 MHz spectral resolution.
Comparable or even better than best laboratory instruments.



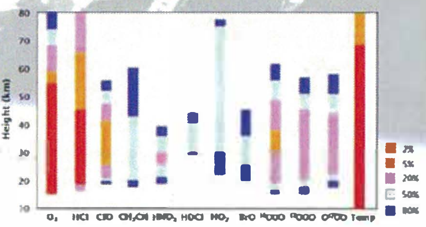


Scientific targets of SMILES

1. Inorganic Chlorine chemistry
   - ClO to HCl ratio
     (O_3 trend in the US)
   - HOCl production
     (O_3 trend in the LS)
   - Global ClO
     (background ClO)
2. Bromine budget (very short-lived source gas issue)
3. HO_x budget

etc.

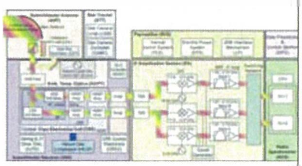
Simulated SMILES observation performance



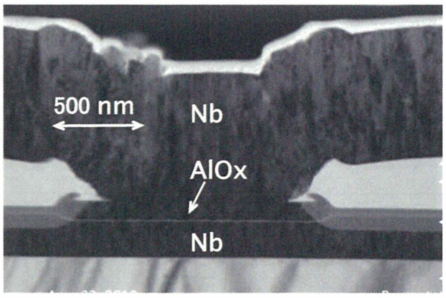
Error estimation for the mid-latitude case based on the single scan measurement

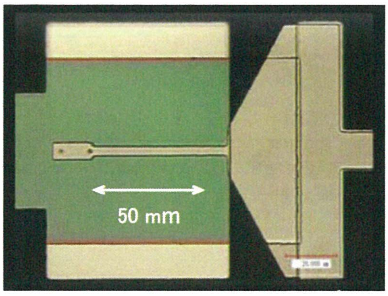
Overview of JEM/SMILES Instruments:

SIS Device



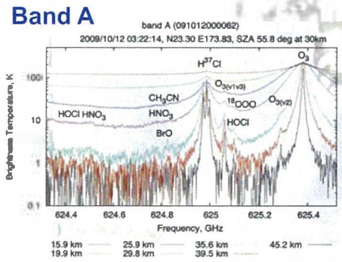
SIS Junction: Nb/AlOx/Nb
Junction Area: ~1 x 1 mm^2
Current Density: 6-7 kA/cm^2
Fabricated at Nobeyama Radio Observatory, JAPAN



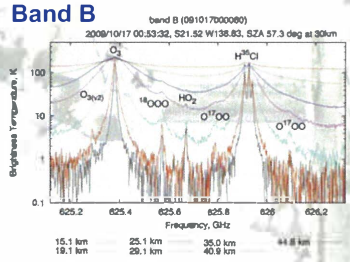


Two Bands among Band A, B, C can be observed.

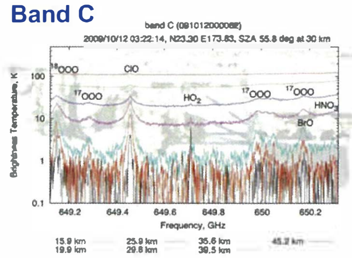
Band A



Band B



Band C



Frequency region has been selected by engineering interest, as high as possible, but 625-626 GHz region is the only frequency to measure HCl below 1 THz.

At 600 GHz troposphere is opaque in limb.

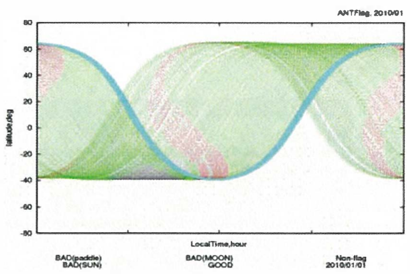
Tsys ~ 350 K, and Noise floor is ~0.4 K, given by

$$NE\Delta T_B = \frac{T_{sys} + T_{Atm}}{\sqrt{\Delta\nu \cdot \tau}}$$

Sep. 27-29, 2010 AURA meeting, M. Suzuki et al 9

ISS Orbit plane rotates in ~90 days, 45 days for diurnal coverage. It will be good platform for diurnal variation study and solar occultation, such as ISS/SAGE-III (2015 ?).

Actual Local Time Coverage of SMILES in January 2010. Blue: January 1st. Red: not observed.



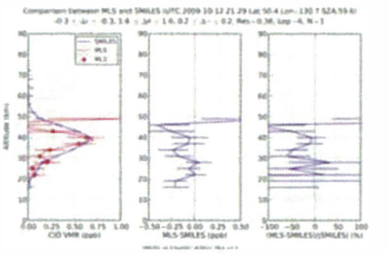
ISS orbit
- Low altitude: 350 km
- Moderate inclination: 51.2°

Sep. 27-29, 2010 AURA meeting, M. Suzuki et al 10

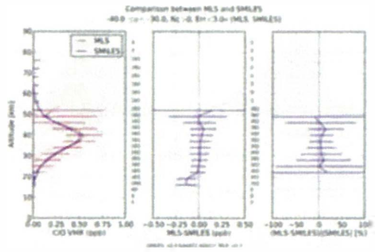
Comparison of SMILES ClO v2.0 with MLS 3.3

MLS coincidence, DSZA < 2°, **SMILES ver.2 release candidate.**

Coincidence event on Oct. 12, 2009 SMILES, MLS at 50.4°N 130.7°E (the first coincidence in current criteria). SZA= 59.6°



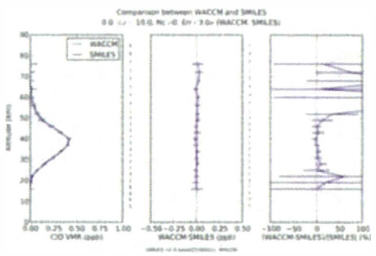
Coincidence Statistics 30S-40S, 161 SMILES vs. 301 MLS profiles, most coincidence cases in current criteria. < 10% agreement with MLS 3.3 between 22-48 km



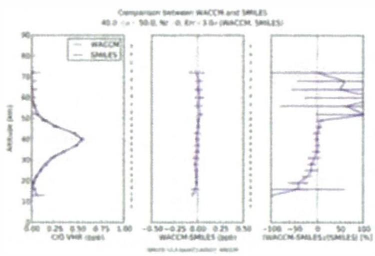
Daytime ClO comparison with NCAR WACCM

Other latitude slightly worth than these examples.

coincidences 0-10N, Oct. - Feb., SMILES vs. WACCM



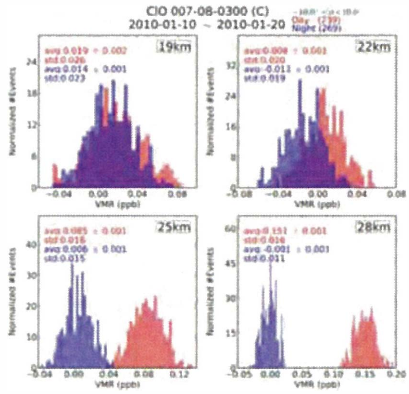
coincidences 30-40N, Oct. - Feb., SMILES vs. WACCM



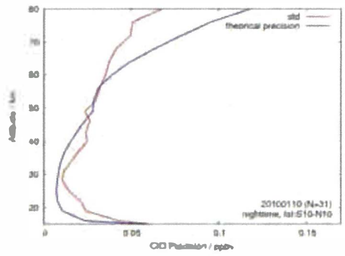
Detection limit of SMILES ClO, 15 ppt at 25 km in single shot, negative bias at 22 km in nighttime

Detection limit changes with pressure.

Daytime (noon) and Nighttime (midnight) difference at 10N-10S



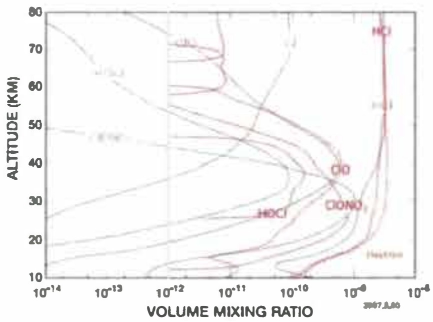
Theoretical detection limit and standard variation of nighttime. Below 35 km, it should be detection limit.



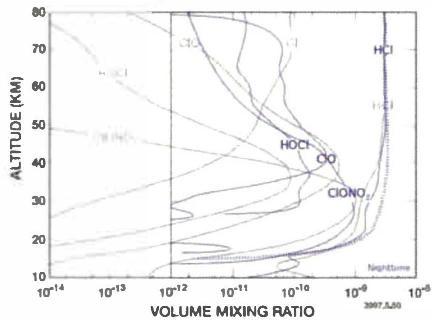
SMILES (+ MIPAS) can provide knowledge of chlorine partitioning at the background atmosphere based upon observations.

2010/10/12, local solar noon (53N-60N) and midnight (23S-33S),
$ClONO_2$: MIPAS IMK, day 51N-57N, night 50N-54N

Daytime profiles

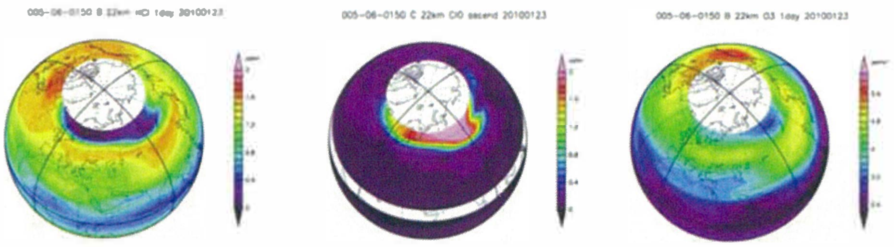


Nighttime profiles



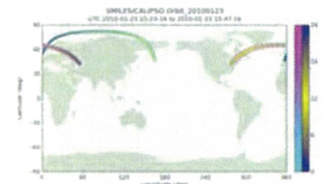
Brasseur and Solomon, pp.373

Fig. 1 HCl (left), ClO (center), and O_3 (right) distribution on Jan. 23, 2010 at the 22 km altitude in the northern hemisphere.



HCl is about 1.6 ppbt at outside polar vortex and it is almost entirely converted to the ClO (1.6 to 2.0 ppbt). O3 destruction has occurred as much as 20% (from 4 ppmv to 3.2 ppmv) after 3 weeks of heterogeneous chemical process.

Fig. 2 (a) shows trajectory of observation points of SMILES (large circles) from 15:23UT to 15:47 in Jan. 23, 2009, and CALIPSO observation points which passed north of Europe. Fig. 2(b) shows SMILES ClO vertical section.



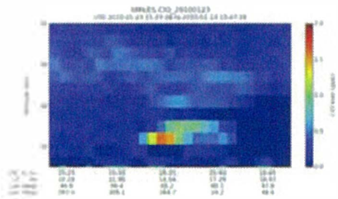
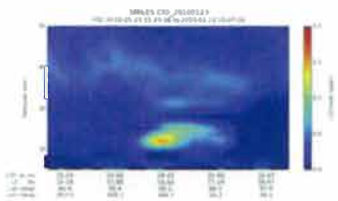
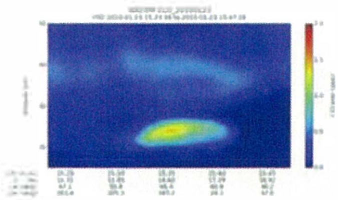


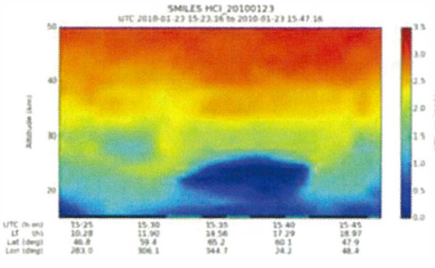
Fig. 2(b) shows SMILES ClO vertical section.



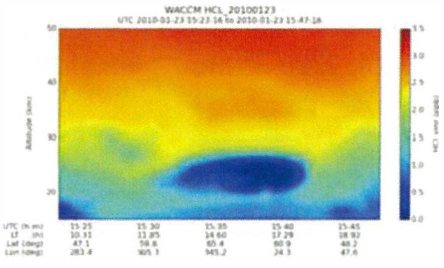
(c) SMILES ClO vertical section interpolated.



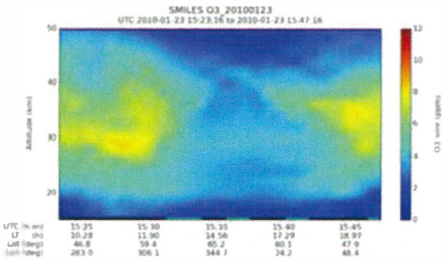
(d) SD-WACCM ClO vertical section.



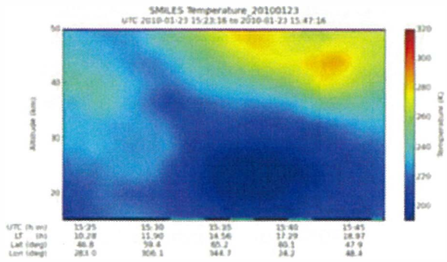
(e) SMILES HCl.



(f) SD-WACCM HCl

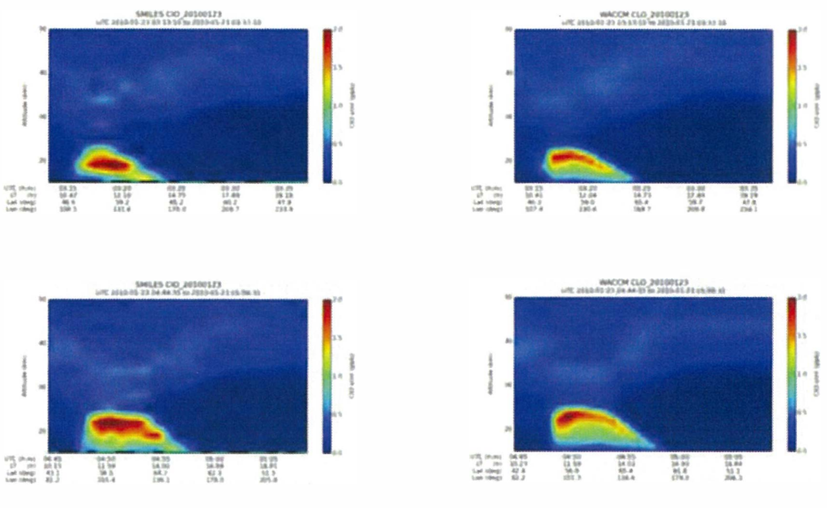


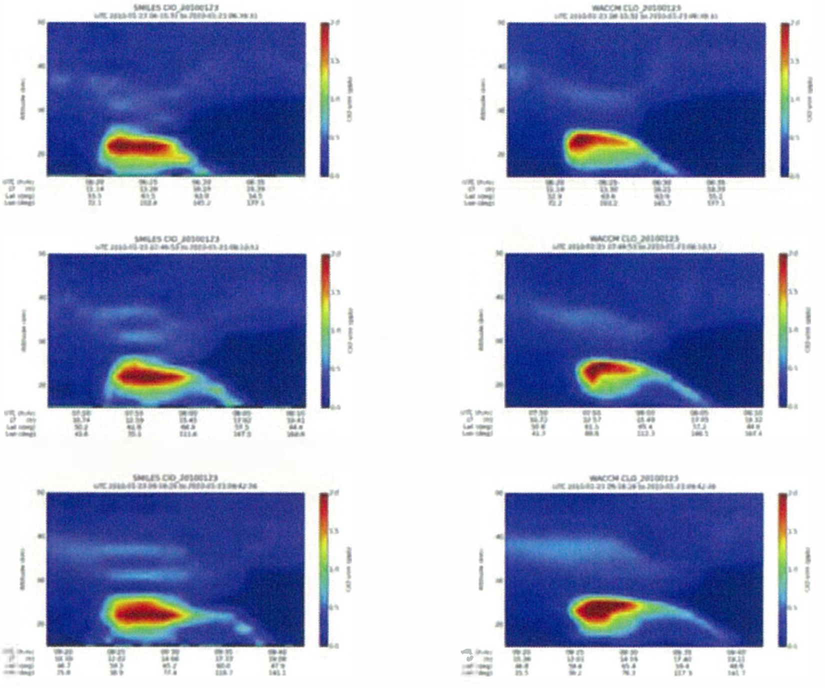
(g) SMILES O_3



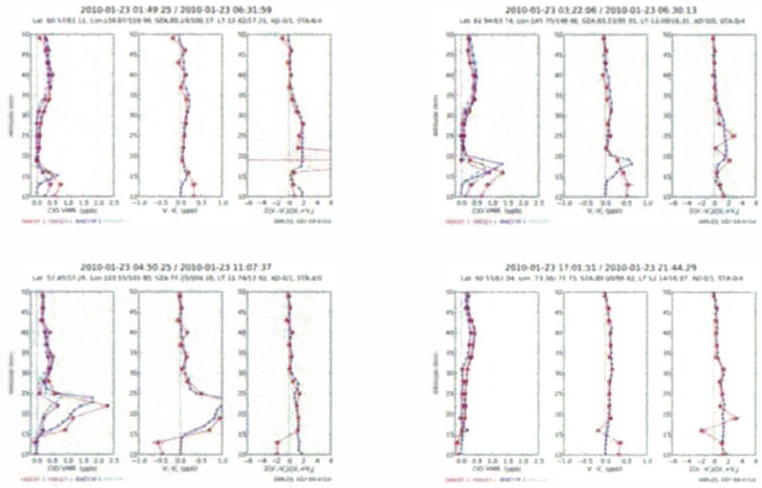
(h) SMILES Temperature

Checking ClO more thoroughly, Jan 23 at different orbits. (left) SMILES interpolated, (right) WCCAM interpolated.

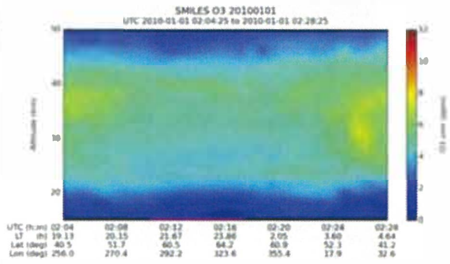


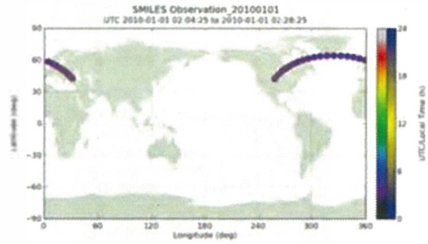


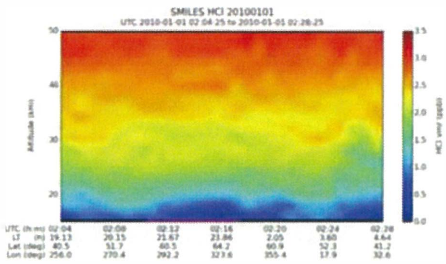
SMILES can measure same location at different local times. Jan. 23[rd] masurements showed, nighttime CLO decline generally agreed well with WACCM calculation. This suggest current knowledge of $(ClO)_2$ formation rate is acceptable.



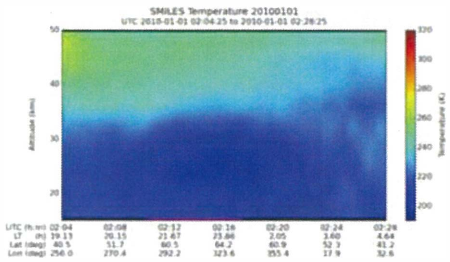
SMILES O_3(Left-Upper), Locations (Right-Upper) ClO (Left-Lower), HCl (Right-Lower)

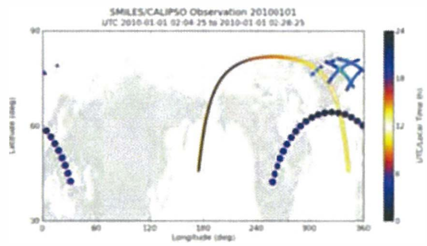


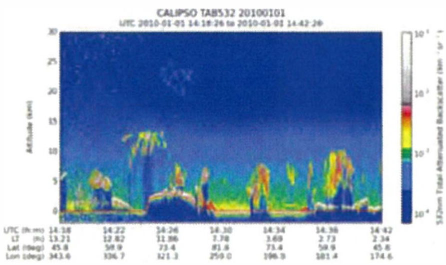


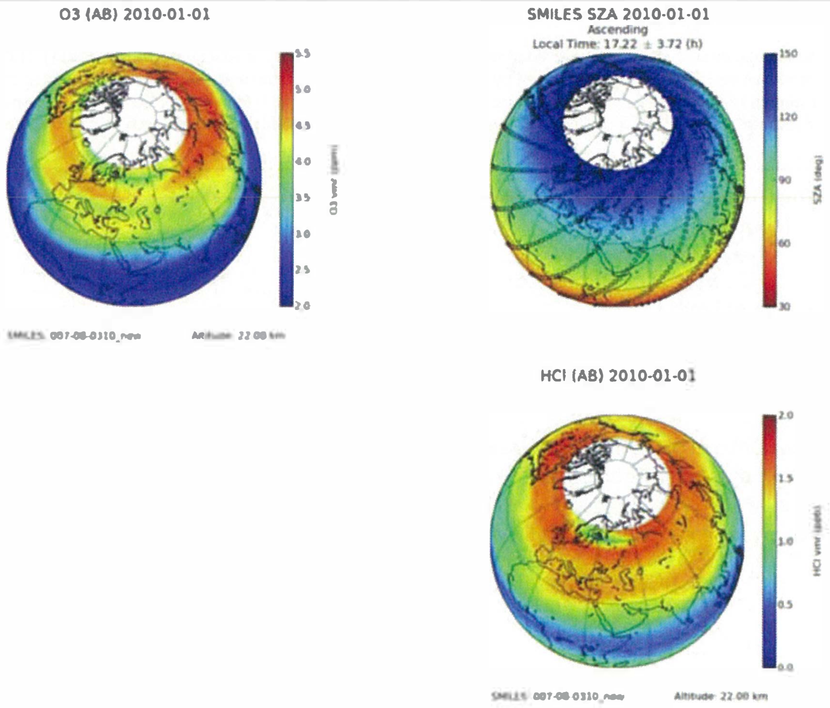


SMILES O3(LeftUpper), ClO(LD), Locations(RU). CALIPSO (RD)

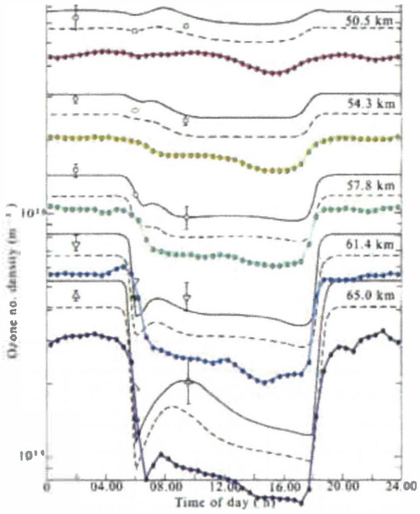








Diurnal variation of O3 in the mesosphere observed by SMILES



- mesospheric O_3 shows strong diurnal variation, but it was not observed clearly from ground and space.
- Daytime O_3 and HO_2 etc have relations to H_2O, which is now under evaluation. (Clancy et al, JGR 1994)

$$O_3 \propto (k5 \cdot k19)^{1/3}(k7 \cdot J3a)^{-1}(k9 \cdot k10)^{-1/2}[H_2O]^{-1/2} \quad (1)$$

$$HO_2 \propto (k5 \cdot k19)^{-1/2}(k7 \cdot J3a \cdot k9)^{1/2}(k10)^{-1/2}[H_2O]^{1/2} \quad (2)$$

SMILES and a model calculation (Vaughan, *Nature*, 1984)

Kovalenko et al (2007) have questioned the reaction rate of ClO + HO_2 → HOCl + O_2 (JPL206/2011), and it proposed to be nar the value reported by Stimpfle (1979). von Clarmann et al ら(2009) also supported this claiming from MIPAS-B balloon measurements. This could affect Total Ozone Loss at the lower stratosphere significantly. We checked the reaction rate by using steady state relations around [HOCl] at the 30-45 km.

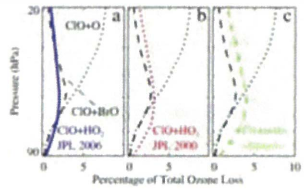


Figure 5. Calculated contribution of the three most important catalytic cycles involving ClO to 24-hour averaged ozone loss, for the 20 September 2005 flight, for model runs (a) $k_1^{JPL2006}$, (b) $k_1^{JPL2000}$, and (c) $k_1^{Stimpfle}$.

Zonal mean temperature = 249K.

k_1 (nominal)

k_1 = 2.7e-12 * exp(+220/*T*)

6.53 x10^{-12} cm^3/molecule/s (JPL2006)

k_1 (upper limit)

9.66x10^{-12} cm^3/molecule/s

k_1 (lower limit)

4.41x10^{-12} cm^3/molecule/s

Kovalenko et al (2007) [Balloon and Model] showed better agreement with k1 by Stimpfle et al (1979). von Clarmann et al (2009) also reported same results using ENVISAT/MIPAS.

k_1 = 3.3x 10^{-11} exp(*-850/T*) + 4.5x $10^{-12}$$(T/300)^{3.7}$

3.34479x 10^{-12} cm3/molecule/s (Stimpfle)

ClO + HO_2 → HOCl + O_2 (k_1)

At the middle atmosphere 40-50 km, chlorine spices are distributed among HCl, HOCl, and ClO. The chemical balance among these species is controlled by following chemical reactions;

ClO + HO_2 --> HOCl + O_2 … k_1 (1)

HOCl + OH --> H_2O + ClO … k_2 (2)

HOCl + hv--> OH + Cl … j_3 (4)

The daytime chemical equilibrium [ClO]/[HOCl] can be simplified at 40-50km, where [OH] is small, as following,

$$\frac{[\mathrm{ClO}]}{[\mathrm{HOCl}]} = \frac{j_3}{k_1[\mathrm{HO_2}]} \quad (5)$$

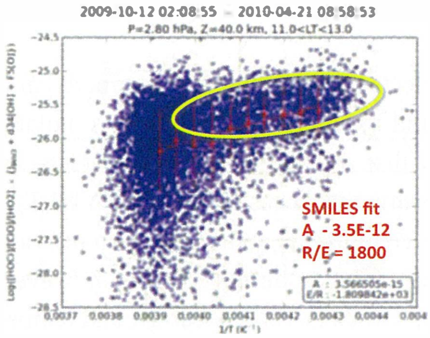
- SMILES ver. 2.0a, [ClO], [HO2], [HOCl]
- GEOS-5 meteorological data
- j_3: HOCl photolysis is calculated, from cross-section JPL2006, multiple scattering calculation using MODTRAN5 (DISORT, 16 streams).

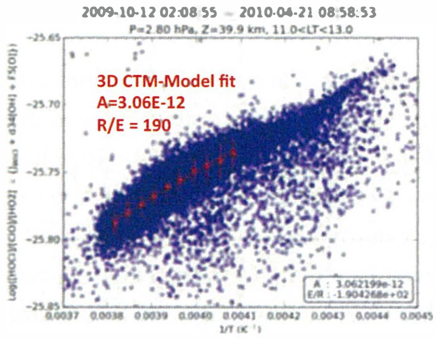
SMILES観測(ver. 2.0a)に基づくk_1の計算は30, 35, 40 kmにおいてJPL2006を支持している.
Oct. 12, 2009, 57.1±1.8°N, SZA=64.8±1.6°

H (km)	T (K)	k_1 (cm^3/molecules/s)			
		SMILES	WACCM	JPL2006	Stimpfle
30	223.0	5.91E-12	6.87E-12	7.24E-12	2.23E-12
35	228.3	9.75E-12	6.10E-12	7.08E-12	2.44E-12
40	239.8	7.25E-12	5.38E-12	6.76E-12	2.92E-12
45	249.2	3.72E-12	3.70E-12	6.53E-12	3.36E-12

Stratospheric ClO, HO_2, and HOCl measurements by SMILES can estimate k (d_{34}): ClO + HO_2 → HOCl + O_2
JPL2011 value: A=2.7E-12, R/E = 220

d_{34} = [HOCl]/[ClO]/[HO2]x{JHOCL + d34[OH] + f35[O]





SMILES data fit (left), was not successful but agree with JPL2011 at 250 K.
If we look at only the yellow circle region T<250 K (1/T > 0.004), it may agree with JPL2011.