

Detection of methane depletion associated with stratospheric intrusion by atmospheric infrared sounder (AIRS)

Xiaozhen Xiong,¹ Chris Barnet,² Eric Maddy,^{2,3} S.C. Wofsy,⁴ Liangfu Chen,⁵ Anna Karion,⁶ and Colm Sweeney⁶

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[1] Atmospheric methane (CH₄) concentration in the mid-to-upper troposphere has been retrieved using atmospheric infrared sounder (AIRS) data on NASA EOS/AQUA. By selecting the AIRS strong CH₄ absorption channels near 1306 cm⁻¹, severe CH₄ depletion was mapped during a stratospheric intrusion event on 27 March 2010. The areas with depleted CH₄ mixing ratio are collocated with enhanced ozone (O₃) and low tropopause height. Aircraft measurements observed the depleted CH₄ and enhanced O₃ down to 550 hPa. An estimate of the depleted CH₄ amount which resulted from stratospheric intrusion is -54 to -67 Tg yr⁻¹. This study suggests that the AIRS and/or other thermal infrared sounders can provide an observation of CH₄ variation associated with stratospheric intrusion, a key unknown in CH₄ budget, and this data set will be also useful for studying the stratosphere-troposphere exchange (STE). **Citation:** Xiong, X., C. Barnet, E. Maddy, S. C. Wofsy, L. Chen, A. Karion, and C. Sweeney (2013), Detection of methane depletion associated with stratospheric intrusion by atmospheric infrared sounder (AIRS), *Geophys. Res. Lett.*, 40, 2455–2459, doi:10.1002/grl.50476.

1. Introduction

[2] Extensive research on stratosphere-troposphere exchange (STE) points out that the STE in the extratropics is a key factor impacting the ozone budget in the upper troposphere and water vapor variability in the lower stratosphere [e.g., *Stohl et al.*, 2003; *Meloen et al.*, 2003; *Sprenger et al.*, 2007; *Pan et al.*, 2010]. In general, the STE includes the exchange in both directions: troposphere-to-stratosphere transport (TST) and stratosphere-to-troposphere transport (STT), and the latter one is also referred to as stratospheric intrusion.

[3] It has been observed that the STT in the extratropics can occur in tropopause folds, cutoff lows, mesoscale convective complexes, thunderstorms, and breaking gravity waves [*Stohl et al.*, 2003]. STT is also associated with the Brewer-Dobson circulation in the stratosphere, which attains maximum strength in late winter and early spring [*Appenzeller et al.*, 1996]. A lot of studies on the ozone transport due to stratospheric intrusion and its impact on tropospheric ozone have been made. For example, *Hauglustaine et al.* [1998] reported that the global stratosphere-to-troposphere ozone flux ranges between 391 and 846 Tg yr⁻¹ based on different chemical transport models. *Kentarchos and Roelofs* [2003] estimated the net annual Northern Hemisphere (NH) stratosphere-to-troposphere ozone flux to be 437 Tg yr⁻¹.

[4] As estimated by *Kentarchos and Roelofs* [2003], ozone from stratospheric origin contributes about 15% to the average oxidation capacity in the NH. The impact to the mean concentrations of the hydroxyl (OH) radical by STT can affect the budget of CH₄ and many other trace gases [*Esler et al.*, 2001]. It was believed that the observed decrease of CH₄ in the marine boundary layer in March–April in the high NH can be linked with the air descending from the lower stratosphere in the downward branch of the Brewer-Dobson circulation [e.g., *Dlugokencky et al.*, 1995], and this process can also result in a CO₂ minimum in spring [*Shia et al.*, 2006] and impact the seasonal cycle of nitrous oxide [*Nevison et al.*, 2011]. *Schauffler and Daniel* [1994] found that the decline of CH₄ increasing trend in 1992 can be explained by an increase in stratospheric circulation and tropospheric/stratospheric exchange rate, driven by the heating from the Pinatubo volcanic aerosols. *Butchart and Scaife* [2001] predicted an increase of STE of 3% per decade due to enhanced greenhouse gas concentrations. This increase of STE will result in more CH₄ depletion, thus will impact CH₄ budget.

[5] Compared to the study of the impact of STT on the ozone budget, quantification of the impact of STT on CH₄ budget has not been well studied, and such study has been hampered by the lack of observations of CH₄ in the mid-upper troposphere. CH₄ in the stratosphere and near-tropopause region can be observed by the Halogen Occultation Experiment (HALOE) using solar occultation measurements [e.g., *Mote et al.*, 1993], but was limited to 60°S to 60°N. Most recent global measurements of mid-upper tropospheric CH₄ have been made using the hyperspectral thermal infrared sensors, which include the Tropospheric Emission Spectrometer (TES) on NASA Earth Observing System (EOS) Aura mission [e.g., *Worden et al.*, 2012], the AIRS on EOS Aqua mission [*Xiong et al.*, 2008, 2010], the Infrared Atmospheric Sounding Interferometer (IASI) on European polar Meteorological Operational Platform (METOP-1) [*Crevoisier et al.*, 2009; *Razavi et al.*, 2009], and the Greenhouse gases Observation SATellite (GOSAT) [*Yokota et al.*, 2008].

¹IMSG at NOAA Center for Satellite Applications and Research, College Park, Maryland, USA.

²National Environmental Satellite, Data, and Information Service, NOAA, Silver Spring, Maryland, USA.

³Science and Technology Corporation, Hampton, Virginia, USA.

⁴Department of Earth and Planetary Sciences, Harvard University, Cambridge, Massachusetts, USA.

⁵State Key Laboratory of Remote Sensing Science, Institute of Remote Sensing Applications, Chinese Academy of Sciences, Beijing, China.

⁶NOAA/ESRL, Global Monitoring Division, Boulder, Colorado, USA.

Corresponding author: X. Xiong, NOAA/NESDIS/STAR, 5830 University Research Ct, University of Maryland, College Park, MD 20740, USA. (xiaozhen.xiong@noaa.gov)

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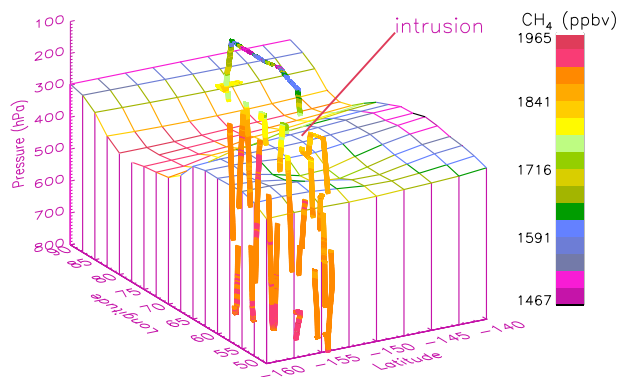


Figure 1. 3-D distribution of tropopause from NCEP/NCAR reanalysis data and the CH₄ mixing ratio on 27 March 2010 from HIPPO aircraft measurements. A vertical flight leg near (150°W, 60°N) with low CH₄ mixing ratio below 1700 hPa at altitude between 200 and 400 hPa marks the intrusion of stratosphere.

[6] On the basis of the CH₄ retrieval from AIRS [Xiong *et al.*, 2008] but with some modification in channel selection and retrieval layers, this paper presents observations of severe CH₄ depletion in the mid-upper troposphere during a stratospheric intrusion event. Some detailed 3-D structure of CH₄ change during this stratospheric intrusion event is investigated through the comparison of CH₄ and ozone distributions and their relationship with the tropopause heights and wind fields data. In situ aircraft measurements from HIPPO are also used to illustrate this stratospheric intrusion and CH₄ depletion. Based on the ratio of the change of CH₄ relative to ozone (both observed by AIRS) in this case, and the ozone flux impacted by STT from other literature [e.g., Kentarchos and Roelofs, 2003], we provided an approximate estimation of the depleted CH₄ amount from the stratosphere to troposphere intrusion.

2. CH₄ and O₃ Data from AIRS Retrievals and Aircraft Measurements

[7] AIRS is a 2378 channel nadir cross-track scanning infrared spectrometer on the EOS/Aqua polar orbit satellite, and a number of trace gas species, including CO₂, CH₄, O₃, and CO, can be retrieved [e.g., Suskind *et al.*, 2003]. Previous validation of the AIRS-retrieved ozone indicated that it reasonably captures the upper tropospheric to lower stratospheric ozone variability or total ozone column variability [Divakarla *et al.*, 2008; Wei *et al.*, 2010]. He *et al.* [2011] showed that a number of significant tropospheric ozone enhancements resulted from stratospheric intrusion events can be observed from AIRS. The AIRS ozone data retrieved using an algorithm similar to AIRS-V5 is used for analysis in this paper.

[8] Using a similar algorithm as detailed by Xiong *et al.* [2008], we selected 27 strong CH₄ absorption channels between 1252.96 and 1306.68 cm⁻¹, and 11 vertically overlapping trapezoidal functions layers for CH₄ retrieval. Use of these peak channels makes the retrievals more sensitive to CH₄ in higher altitudes, where the depletion of CH₄ under the stratospheric intrusion is more significant. As a result, the most sensitive region of the retrieval is in an altitude between 300 and 600 hPa.

[9] Two days of aircraft measurements of CH₄ vertical profiles close in time and location to the stratospheric intrusion event studied in this paper are used. One is the

measurements by the HIAPER Pole-to-Pole Observations (HIPPO) program over the Pacific Ocean on 27 March 2010 [Wofsy and Hippon Science, 2011], in which CH₄ was measured with a Quantum Cascade Laser Spectrometer (QCLS) at 1 Hz frequency with accuracy of 1.0 ppb and precision of 0.5 ppb. Another is the measurement by a NOAA/ESRL Alaska Coast Guard (ACG) flight on 30 March 2010, in which CH₄ was measured with a Cavity Ringdown Spectroscopy (CRDS) analyzer at 0.4 Hz frequency with an overall measurement uncertainty of 2 ppb [Karion *et al.*, 2012 and the references therein]. Data of the wind fields and thermal tropopause will also be used, and they both are downloaded from NCEP/NCAR Reanalysis data (<http://www.esrl.noaa.gov/psd/data/reanalysis/reanalysis.shtml>).

3. Results and Discussion

[10] Stratospheric intrusion brings air depleted CH₄ down to the troposphere, resulting in CH₄ decline in the mid-upper troposphere where AIRS has a good sensitivity. One case for which we happened to have HIPPO aircraft measurements on 27 March 2010 will be presented. Figure 1 first shows the CH₄ profiles from HIPPO (phase 3) aircraft measurements overlaid with the tropopause from NCEP/NCAR reanalysis data. The flat flight leg above 200 hPa near (150–140°W, 66–84°N) crossed a trough of the tropopause, in which the lowest tropopause is ~410 hPa at 80°N, and the mean CH₄ mixing ratio for this flight leg is 1596 ± 62 ppbv (mean ± one sigma). The mean ozone and water vapor concentrations for these samples with CH₄ mixing ratio less than 1700 ppb were 750 ± 396 ppb and 7.1 ± 2.8 ppm, respectively. Such a high ozone concentration and low humidity indicate that this air mass originated from the stratosphere. The samples with low CH₄ mixing ratio impacted by the stratospheric intrusion is evident from other flight legs at lower levels, for example, on the flight leg near (70.8–72.6°N, 148.8–148.7°W), where the tropopause is about 350 hPa, enhancement of ozone and depletion of CH₄ were measured down to 550 hPa (Figure 3 will show this again).

[11] Five days' data of CH₄ around 27 March 2010 from AIRS are used to show the impact of stratospheric intrusion. Because the sensitivity of AIRS to CH₄ is mostly at 300–600 hPa, we used the retrieved CH₄ at 407 hPa for analysis. From the snapshot of CH₄ distribution from AIRS at 407 hPa (the left panels of Figure 2), we can see that, compared to the CH₄ distribution 2 days ago (i.e., 25 March), there is a severe CH₄ depletion in a large area over the Arctic Ocean on 27 March 2010. In the area with low CH₄ mixing ratio, the tropopause height is as low as 400 hPa. Two days later (29 March 2010), the CH₄ mixing ratio over the Arctic Ocean increases but is still smaller than that on 25 March 2010. CH₄ mixing ratio remains low along the isotropic line of tropopause height at 350 hPa.

[12] Since the ozone is mainly in the stratosphere and upper troposphere, enhancement of the total ozone is commonly used to characterize the stratospheric intrusion. The right panels of Figure 2 show the total ozone distributions retrieved from AIRS on these 3 days overlaid with wind fields at 400 hPa. It is easy to see that the regions with enhanced ozone are collocated with the regions with low CH₄ mixing ratios and low tropopause. In regions with high ozone and low CH₄, the thermal tropopause heights are

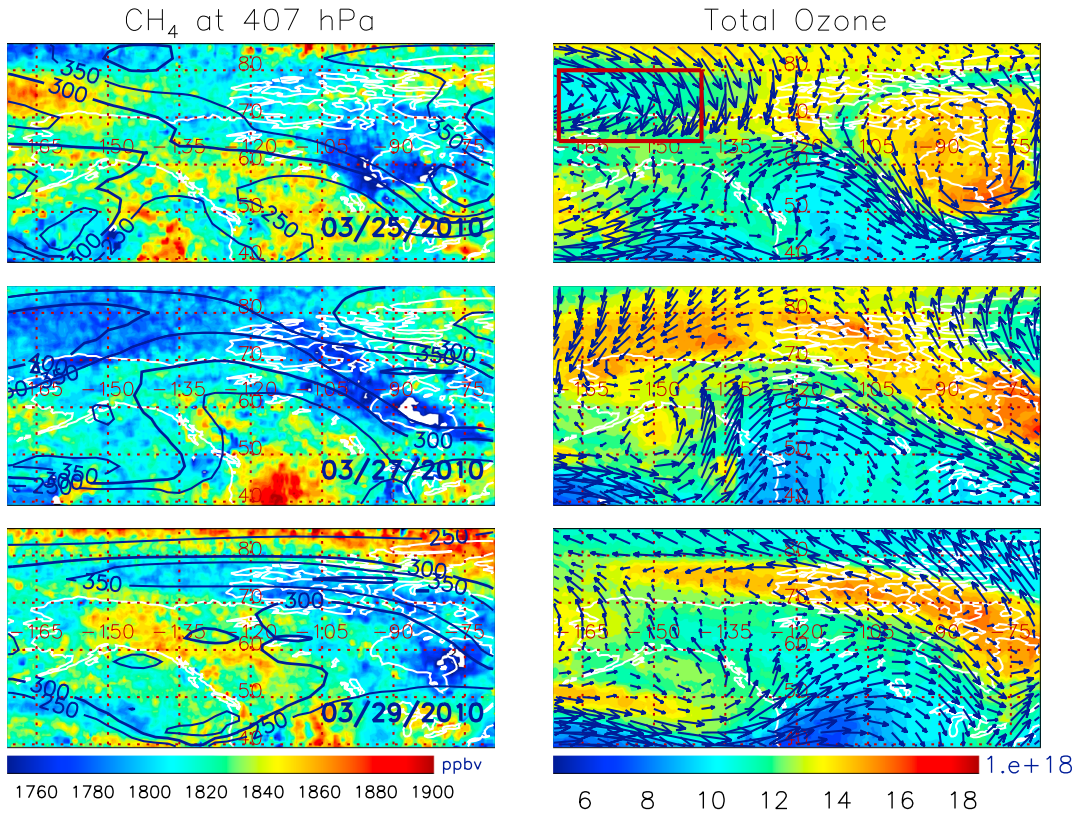


Figure 2. Distribution of CH₄ from AIRS at 407 hPa and the contour of tropopause (left panels) and total ozone amount from AIRS (overlaid with wind vectors at 400 hPa, right panels) for 3 days on 25, 27, and 29 March 2010. Dark blue regions in the left panels are air masses with low CH₄ impacted by stratospheric intrusion, and the enhancement of ozone is evident in the corresponding regions in the right panels. The red box in the upper right panel marks the region where the air masses with low CH₄ mixing ratio were detected and HIPPO aircraft measurement was taken in the middle of this box.

mostly below 350 hPa. Such a low tropopause and an enhancement of total ozone provided evidence for stratospheric intrusion, and its location is also the region where CH₄ decline is evident.

[13] In addition to the impact of stratospheric intrusion, the distribution of CH₄ and ozone is also impacted by zonal transport (Figure 2). At 400 hPa, the south wind was the dominant flow over the Arctic Ocean on 25 March and there is a big ridge across the United States. An anticyclone was developing near the southwest coast of Alaska on 25 March, which further developed on 27 and 29 March and moved slightly to the southeast. Over the northwest of Canada, there was an anticyclone on 25 March 2010, which was broken into two parts on 27 March, one formed a cyclone in the upper right corner on Figure 2, blocking the transport of air mass to the east, and the other part was merged with ridge in the south. The southeastward flow along the ridge on 27 and 29 March forced the air masses with low CH₄ and enriched ozone to move across Canada. A stationary center of low CH₄ and high ozone over Canada persisted until 29 March 2010.

[14] To better understand the CH₄ depletion in the vertical direction during the stratospheric intrusion, we chose an area [see the box of (60–75°N, 179–150°W) in Figure 2] and computed the mean profiles for these 3 days. As illustrated in Figure 3, from 25 March to 27 March 2010, the CH₄ mixing ratio at 300–500 hPa decreases by ~50 ppbv. In 2 days after

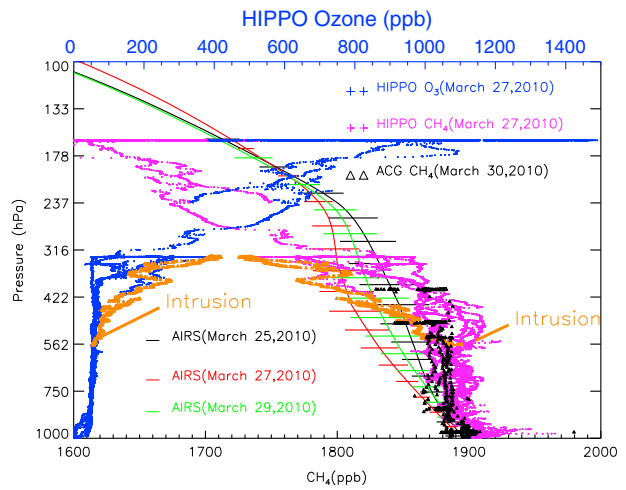


Figure 3. Mean vertical profiles of AIRS retrieved CH₄ averaged in the box (60–75°N, 179–150°W) of Figure 2 for 3 days on 25, 27, and 29 March 2010 with the bars representing the standard deviation. All aircraft measurements of CH₄ and ozone (x axis in the upper) mixing ratios by HIPPO on 27 March 2010 and ACG on 30 March 2010 are plotted. The orange triangles are the samples in one flight leg with CH₄ depletion and ozone enhancement detected down to 550 hPa and its location is (70.8–72.6°N, 148.8–148.7°W).

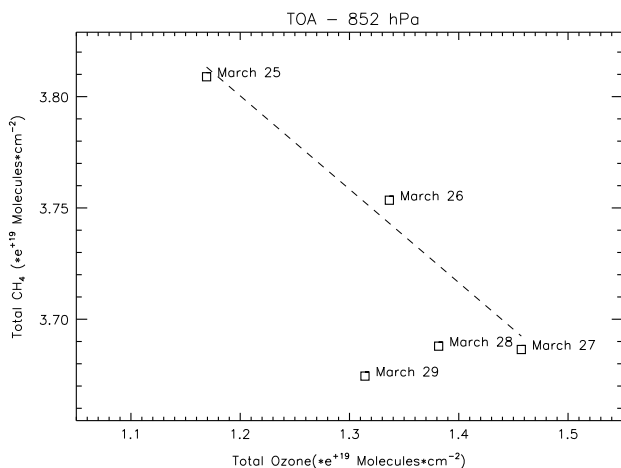


Figure 4. Variation of the AIRS retrieved column amount of CH₄ relative to ozone above 852 hPa, averaged in the blue box (60–75°N, 179–150°W) of Figure 2 for 5 days from 25 to 29 March 2010. Dash line is the linear fitting using 3 days' of data on 25–27 March. In the fitted linear equation, the values in the *parentheses* are standard deviation.

the stratospheric intrusion, i.e., 29 March, the CH₄ mixing ratio is getting back, but it is still ~10 ppb smaller than that before the intrusion on 25 March, suggesting the slow recovery of the depleted CH₄.

[15] All aircraft measurement data at different altitudes from HIPPO on 27 March, and ACG on 30 March 2010 [in the region (58–71°N, 165°W–152°W)] are plotted in Figure 3. From one flight leg of HIPPO, plotted with orange color, it is evident that the CH₄ depletion and ozone enhancement occurred down to 550 hPa. Although the heights sampled by ACG flights cannot give any information about the CH₄ at levels above 300 hPa on 30 March, we noticed that at layers below 500 hPa, the CH₄ from ACG is, on average, about 7.0 ppbv lower than the HIPPO measurements on 27 March, 2010. The median distance between the HIPPO and ACG measurements is 772 km, and the movement of air masses from the south of Alaska along the anticyclone system may be the major reasons for the lower mixing ratios measured by ACG on 30 March, 2010. Due to the large difference between AIRS and aircraft measurements in time and space, it is not our purpose to validate the AIRS retrievals, which requires us to select the collocated profiles and take into account the averaging kernels in the retrievals [e.g., *Maddy and Barnett, 2008* and *Xiong et al., 2008*].

[16] The intrusion of stratospheric air into the Arctic troposphere down to 700 hPa was ever detected by aircraft measurements made during the Arctic Gas and Aerosol Program (AGASP) before [*Schnell, 1984*]. In this case, on 27 March 2010, the CH₄ depletion is evident at 600–700 hPa from AIRS retrievals, which is lower than the level of ~550 hPa, as detected by the limited HIPPO aircraft sampling in this big area (Figure 3). We noted that as the degree of freedom in AIRS retrieval is around 1.0 or less in the Arctic, and the information in different vertical retrieval layers is correlated, the retrieved CH₄ in different layers are not independent [*Xiong et al., 2008*]. So, we cannot use AIRS retrieval data to quantitatively define the level impacted by the stratospheric intrusion.

[17] From 25 to 27 March, the air movement over Arctic Ocean and north Alaska is dominant by the flow from the Arctic towards the south, as evident from the wind fields (Figure 2), so we assume the change of CH₄ and O₃ is mainly from the stratospheric intrusion. We used the column amounts of CH₄ and ozone above 596, 753, and 852 hPa from AIRS retrievals to estimate the relative change of CH₄ to ozone during this stratospheric intrusion event. In this computation, the impact of emission and transport in the lower boundary layer is excluded. By fitting the column amounts of CH₄ relative to ozone from 25 to 27 March (Figure 4), we got the ratio of the change of total CH₄ relative to the total ozone ($\Delta\text{CH}_4/\Delta\text{O}_3$) for using the column amounts above 596, 753, and 852 hPa, respectively, and they are -0.123 , -0.146 , and -0.153 , respectively. According to the estimation of *Kentarchos and Roelofs [2003]*, the net annual Northern Hemisphere stratosphere-to troposphere ozone flux is 437 Tg yr^{-1} . By multiplying the ratio of $\Delta\text{CH}_4/\Delta\text{O}_3$ from AIRS with the ozone flux of 437 Tg yr^{-1} , we can estimate the net annual Northern Hemisphere stratosphere-to troposphere CH₄ loss as -54 , -64 , and -67 Tg yr^{-1} , respectively. Please note that as the vertical sensitivities of AIRS to CH₄ and O₃ are different, the above computation is just an approximation and a more accurate computation needs to use a model. Even so, it is promising that this amount is close in magnitude to the estimated stratospheric loss of $40 \text{ Tg CH}_4 \text{ yr}^{-1}$ in IPCC [2007]. The recovery of CH₄ after this intrusion on 28 and 29 March is also evident.

4. Summary and Conclusions

[18] A severe CH₄ depletion was observed in a large area during a strong stratospheric intrusion event on 27 March 2010, which is in consistent with ozone enhancement and low tropopause height, and in situ HIPPO aircraft measurements. The impact to ozone and CH₄ mixing ratio during this intrusion is detected down to ~550 hPa from HIPPO aircraft measurements on 27 March 2010. The decline of AIRS-CH₄ is as large as ~50 ppb on average, however, we cannot use AIRS data to quantify the pressure level impacted by this intrusion due to the limited information content in AIRS retrieval.

[19] Based on the change of the column amounts of ozone and CH₄ above 852 hPa or higher from 25 to 27 March 2010, we estimated the ratio of CH₄ change relative to ozone. Using this ratio multiplied by ozone flux due to STE from literature, we estimated the CH₄ loss of $54\text{--}67 \text{ Tg yr}^{-1}$. Rather than to provide a good estimate of the CH₄ depletion, this study just demonstrates the capability to utilize AIRS to observe the CH₄ depletion associated with stratospheric intrusion, and the potential to use this data set in conjunction with a model to better quantify the CH₄ exchange during the STE, which is very important for studying the CH₄ budget and its feedback with global warming. The AIRS retrieved CH₄ data may be also used as tracer to study the STE processes.

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