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Information-based mid-upper tropospheric methane derived from Atmospheric Infrared Sounder (AIRS) and its validation

X. Xiong[1,2], **C. Barnet**[1], **J. Wei**[1,2], **and E. Maddy**[1,2]

[1]NOAA/NESDIS/Center for Satellite Applications and Research, Camp Springs, Maryland, USA
[2]Perot Systems Government Services, Fairfax, Virginia, USA

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Correspondence to: X. Xiong (xiaozhen.xiong@noaa.gov)

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Abstract

Atmospheric Infrared Sounder (AIRS) measurements of methane (CH_4) generally contain about 1.0 degree of freedom and are therefore dependent on a priori assumptions about the vertical methane distribution as well as the temperature lapse rate and the amount of moisture. Thus it requires that interpretation and/or analysis of the CH_4 spatial and temporal variation based on the AIRS retrievals need to use the averaging kernels (AK). To simplify the use of satellite retrieved products for scientific analysis, a method based on the information content of the retrievals is developed, in which the AIRS retrieved CH_4 in the layer from 50 to 250 hPa below the tropopause is used to characterize the mid-upper tropospheric CH_4 in the mid-high latitude regions. The basis of this method is that in the mid-high latitude regions the maximum sensitive layers of AIRS to CH_4 have a good correlation with the tropopause heights, and these layers are usually between 50 and 250 hPa below the tropopause. Validation using the aircraft measurements from NOAA/ESRL/GMD and the campaigns INTEX-A and -B indicated that the correlation of AIRS mid-upper tropospheric CH_4 with aircraft measurements is ~0.6–0.7, and its the bias and rms difference are less than ±1% and 1.2%, respectively. Further comparison of the CH_4 seasonal cycle indicated that the cycle from AIRS mid-upper tropospheric CH_4 is in a reasonable agreement with NOAA aircraft measurements. This method provides a simple way to use the thermal infrared sounders data to approximately analyze the spatial and temporal variation CH_4 in the upper free tropospere without referring the AK. This method is applicable to derive tropospheric CH_4 as well as other trace gases for any thermal infrared sensors.

1 Introduction

As one of the most important greenhouse gases in the atmosphere, CH_4 has been observed to rise dramatically since the preindustrial era. The increase rate was observed to slow down after 2000 (Simpson et al., 2002; Dlugokencky et al., 2003), but a sig-

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nificant increase was found from 2006 to 2007 (for example, Rigby et al., 2008). The quantification of global methane emissions still has large uncertainties (Zhuang et al., 2009), as the methane emissions have a large spatial and temporal variation and our observations are limited. For example, large CH_4 emission from Arctic tundra during the onset of freezing was found recently (Mastepanov et al., 2008); Arctic lakes have recently been recognized as a major source (e.g. Walter et al., 2006); in the tropical region there are possibly some large emission from terrestrial plants (do Carmo et al., 2006; Keppler et al., 2006).

Space-borne measurements may be able to help us better understand the relative strengths of the methane sources and sinks as they provide the large spatial and temporal coverage. For example, from the comparison of model simulations with space-born observation using the SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY (SCIAMACHY) instrument onboard ENVISAT, Frankenberg et al. (2005, 2008) pointed out the exist of some unknown CH_4 sources in the tropical region. The Asian CH_4 plume was observed during the monsoon season using AIRS, and through the comparison of AIRS with model simulation, Xiong et al. (2009) suggested that the CH_4 emissions from rice paddies in Southeast Asia is possibly overestimated.

Recent missions of space-borne observations of CH_4 include the observation near the tropopause to higher altitudes based on a limb viewing geometry, the observation of CH_4 in the middle troposphere using thermal infrared spectrum, and the measurement of total column using near infrared spectrum. The space-borne observations of CH_4 based on a limb viewing geometry include the Atmospheric Chemistry Experiment-Fourier Transform Spectrometer (ACE-FTS) solar occultation instrument onboard the Canadian SCISAT-1 satellite (Nassar et al., 2005; DeMazière et al., 2008), Halogen Occultation Experiment (HALOE) measurement on the Upper Atmosphere Research Satellite (UARS) (Park et al., 2004 and references therein), the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) on ENVISAT (Payan et al., 2007). The observation of CH_4 in the middle troposphere using thermal infrared spectrum include

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the Interferometric Monitor for Greenhouse Gases (IMG) on board the Japanese Advanced Earth Observing Satellite (ADEOS) (Clerbaux et al., 2003), Tropospheric Emission Spectrometer (TES) on NASA/Aura (Payne et al., 2009 and references therein), AIRS on NASA/AQUA (Xiong et al., 2008), Infrared Atmospheric Sounding Interferometer (IASI) space-borne instrument on METEOP-1 (Razavi et al., 2009; Crevoisier et al., 2009). The measurement of total column using near infrared spectrum include SCIMACHY (Frankenberg et al., 2005 and 2008), and Greenhouse gases Observation SATellite (GOSAT), which carries both the Thermal And Near infrared Sensor for carbon Observation (TANSO) (http://www.gosat.nies.go.jp/) and was launched by Japan Aerospace Exploration Agency (JAXA) on 23 January 2009.

However, due to limit of the information content of thermal infrared sounder, use of satellite retrieved products is complicated as we need to consider the sensitivity of the retrieval, which strongly depend on the temperature lapse rate and the amount of moisture in the atmosphere (Xiong et al., 2008). The degrees of freedom for CH_4 from thermal infrared sounders are 0.5–2.0 (Payne et al., 2009). Because these sounder measurements are usually sensitive to a single piece of information (i.e., a vertically integrated column weighted toward the middle-to-upper troposphere), the satellite retrieved CH_4 mixing ratio is not exactly equivalent to the CH_4 mixing ratio in real atmosphere. To better characterize the CH_4 mixing ratio in the atmosphere, we need to use the averaging kernels and the a priori (or first-guess) information in the comparison of AIRS retrievals with CH_4 mixing ratio in the real atmosphere. For scientific analyses of the satellite retrieved products it is desirable to use a representation in which measurement information is dominant and the influence of a priori information is minimal (Payne et al., 2009). To take advantage of the sensitivity characterization allowed by the retrieval on a fine grid and reduce the influence of the a priori information, Payne et al. (2009) used an approach to transform each retrieved profile to an appropriate geographically varying coarse grid. Their results are promising but have not been validated.

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The aim of this paper is to develop another approach that uses the sensitivity information of retrieval but without transforming the retrieved profile to a coarse grid (Payne et al., 2009). This approach is based on the correlation of tropopause height with the maximum sensitive layer of AIRS retrieval, and uses the retrieval of AIRS in its most sensitive layer to characterize the mid-upper tropospheric CH_4 in the mid-high latitude regions. Since atmospheric CH_4 has a long life time of 8.7±1.3 years (IPCC, 2007) and is, in general, well-mixed in the troposphere, it should be possible to use the mid-upper tropospheric CH_4 from AIRS to represent the variation of CH_4 in the free troposphere atmosphere. We evaluate this method by comparing the derived tropospheric CH_4 from AIRS with in-situ aircraft measurements at 20 sites of the NOAA Earth System Research Laboratory, Global Monitoring Division (NOAA/ESRL/GMD), and data from recent aircraft campaigns, i.e. Intercontinental Chemical Transport Experiment (INTEX)-A and -B. More evidence is provided by comparing the seasonal variation of AIRS derived tropospheric CH_4 with that from aircraft measurements, in which aircraft data of multiple years in one site and data of multiple sites between 40–50° N in North America in the same year are binned separately to increase the number of samples to derive the seasonal cycle. This method provides a simple way to use the satellite retrieved products for scientific analysis, and is applicable to other long life time trace gases and for other thermal sounder instruments.

Section 2 is a brief introduction to the AIRS instrument and the retrieval of CH_4. A description of the method and the data used are present in Sect. 3. The validation to the AIRS mid-upper tropospheric CH_4 with aircraft measurements as well as the comparison of seasonal cycle are given in Sect. 4. A summary and conclusion are given in Sect. 5.

2 The AIRS instrument and its retrieval of atmospheric methane

AIRS was launched in the polar orbit (1:30 p.m., ascending node) on National Aeronautics and Space Administration (NASA) EOS/Aqua platform in May 2002. It has 2378

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channels covering from 649–1136, 1217–1613 and 2169–2674 cm^{-1} at high spectral resolution ($\lambda/\Delta\lambda$=1200) (Aumann et al., 2003), and the noise equivalent change in temperature (NEΔT), at 250 K reference temperature, ranges from 0.14 K in the critical 4.2 μm lower tropospheric sounding wavelengths to 0.35 K in the 15 μm upper tropospheric sounding region. The spatial resolution of AIRS is 13.5 km at nadir, and in a 24-h period AIRS nominally acquires measurements over the complete globe twice daily. In order to retrieve CH_4 in both clear and partially cloudy scenes, 9 AIRS pixels in the footprint of an Advanced Microwave Sounding Unit (AMSU) pixel are used to derive the cloud-cleared radiance (CCR) in this field of regard. The CCR product is then used for sounding retrievals with the spatial resolution of about 45 km. The version 5 of AIRS product retrieval software has been put into operation at NASA Goddard Earth Sciences Data and Information Services Center (DISC), and these data are available at the Goddard DISC (http://disc.gsfc.nasa.gov/AIRS/index.shtml/). An "off-line" version of the AIRS product is run at NOAA National Environmental Satellite, Data, and Information Service (NESDIS), Center for Satellite Application and Research, where the data are thinned to a 3°×3° spatial grid, and these data are used in this paper.

The "off-line" version of the AIRS product differs from the operational version in that the "off-line" version does not use AMSU channel 4 in any of its microwave retrieval steps, regressions, or in product quality assurance, whereas the DAAC version uses a synthetic regression based on other AMSU channels to predict AMSU channel 4. Due to the differences in retrieval state initialization regression operators, slight differences between the DAAC operational version and the "off-line" version will be apparent for all data after October 2007.

Different from SCIAMACHY which uses the absorption spectra of solar radiation in the near-infrared and is sensitive to the total methane column amount, AIRS is a nadir cross-track scanning infrared spectrometer, and is most sensitive to CH_4 in the middle to upper troposphere. As detailed by Xiong et al. (2008), 71 AIRS channels near 7.6 μm are used for the CH_4 retrieval in version 5, and the atmospheric temperature profile, water profile, surface temperature and surface emissivity required as inputs are derived

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from other AIRS channels. In the CH_4 retrieval, the change of CH_4 is derived using a singular value decomposition (SVD) on the weighted covariance of the sensitivity matrix and damping the least significant eigenfunctions of the SVD to constrain the solution (Susskind et al., 2003). This retrieval methodology minimizes the dependence of the solution to the first-guess field and the first-guess error characteristics, and relies exclusively on the signal to noise of the observation to indicate the degree to which the information contained in the radiances should be believed. The first-guess CH_4 profile is a smooth function of latitude and altitude, but does not varies with time and longitude. It was generated by using a non-linear polynomial fitting to various data, including aircraft measurements and model data (Xiong et al., 2008).

3 Method and data

3.1 Relationship between the AIRS maximum sensitive layer and the tropopause in the mid-high latitude regions

AIRS is a thermal infrared sounder sensitive to CH_4 in the middle to upper troposphere. From both the weighting function and the averaging kernels it is evident that the retrieved CH_4 in different layers are not independent, and the sensitivity of the AIRS CH_4 retrieval varies with time and location due to the variation of atmospheric state variables (Xiong et al., 2008). An appropriate validation to the retrieval requires the use of the averaging kernels. However, it is no doubt that the retrieved CH_4 in the maximum sensitive layer of AIRS will better characterize the CH_4 in the atmosphere than any others layers. Therefore when performing a comparison of the AIRS retrieved CH_4 profiles with measurements, it is better to compare CH_4 measurements with AIRS retrievals at altitudes that correspond with the maximum sensitivity for AIRS. The problem is that the most sensitive layer, which is usually determined from the averaging kernels, varies with atmospheric temperature and moisture profiles. The main reason that the most sensitive layer for our CH_4 retrieval varies significantly with geophysical state is the fact

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that our algorithm relies on the vertical sensitivity of AIRS to CH_4 (i.e., the sensitivity covariance) to provide a smoothing constraint. Our retrieval product therefore moves relative to the CH_4 a priori in linear combinations of the eigenvectors of the sensitivity covariance (Maddy and Barnet, 2008). On the other hand, use of a priori information (Rodgers, 2000) to constrain the inversion minimizes the vertical displacement of the retrieval's most sensitive layer by constraining the retrieval product to move in linear combinations of eigenvectors of the a priori covariance (see for instance, Pan et al., 1998).

Another difficulty is that the most sensitive layer is a quantity derived from the averaging kernels and currently not distributed within the V5 output data stream. This complicates the use of this diagnostic level to compare retrieved data with in-situ measurement or model simulations in that both averaging kernels and retrieval products are required.

As an example, Fig. 1 shows the averaging kernels average of 3 days (1–3 August 2004) in Alaska (longitude=150° W, latitude=66° N). 11 trapezoid functions were used in this retrieval. For convenience, the maximum sensitive level is used in this paper, which is defined as the pressure level where the maximum of averaging kernels area is located at the 100 level grid of the forward radiative transfer model (Strow et al., 2003). To obtain this, the area of averaging kernels (marked in black dash line in Fig. 1) in the effective pressure levels of the 11 trapezoid functions was interpolated to the 100 level radiative transfer model grid. The tropopause for each profile was computed using temperature profile, which is retrieved from AIRS using 91 channels independent of the CH_4 retrieval channels (Susskind et al., 2003), and the surface pressure, which was from the NCEP GFS model (Kanamitsu et al., 1991). The details of the algorithm to compute the thermal tropopause can be refereed to Reichler et al. (2003). In this example, the tropopause is at 234.0 hPa, and the most sensitive level, 390.9 hPa, is 156.9 hPa below the tropopause.

Analysis of more cases shows that in the middle to high latitude regions the most sensitive layer of AIRS to CH_4, which is mostly in the mid-upper troposphere, has a

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good correlation with the tropopause. As an example, Fig. 2 illustrates the variation of tropopause and the maximum sensitive level with latitude using one day global data in 4 March 2006 (Fig. 2). In this example, the tropopause decreases from ~120 hPa at near tropics to ~300 hPa at high Northern Hemisphere with latitudes greater 45° N. From the difference between the maximum sensitive level minus the tropopause (lower panel of Fig. 2), we can see the maximum sensitive levels are mostly located between 50 and 250 hPa below tropopause.

3.2 The mid-upper tropospheric CH_4 derived from AIRS retrievals based on tropopause

The finding of the correlation between AIRS' retrieved tropopause and the maximum sensitive level enables the use of the tropopause as an indicator of the most sensitive layer of the retrieval. Since the layer from 50 to 250 hPa below tropopause is a good approximation for the maximum sensitive layer of AIRS, in this paper we use the average of AIRS retrieved CH_4 in the layer starting from 50 to 250 hPa below tropopause to represent the CH_4 in the corresponding layer in the real atmosphere. In the mid-high latitude regions, this layer is mostly at 300–500 hPa, and 400–600 hPa in the high latitude in the winter. In the summer the maximum sensitive level is higher due to the increase of water amount in the atmosphere, thus it is closer to the tropopause. For simplification, we referred CH_4 at this layer as mid-upper tropospheric CH_4.

3.3 AIRS CH_4 and aircraft measurement data

AIRS retrieval products at 3°×3° spatial grid, generated at NOAA/NESDIS/STAR, are used in this paper. To minimize the sampling bias requires enough match-up profiles from AIRS corresponding to each aircraft profile, so the match-up AIRS profiles are picked up if they are within the same day and 800 km of the measurement time and site location. The mean of all these match-up profiles will be compared to the corresponding individual aircraft measurement.

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Aircraft measurements of CH_4 from NOAA /ESRL/GMD Carbon Cycle Group were used for a preliminary validation to AIRS CH_4 (Xiong et al., 2008) for up to ~300 hPa. In this paper we limit the comparison to data above 500–600 hPa. Aircraft measurements of CH_4 from INTEX -A and -B provide more profiles with measurements up to 200 hPa. These campaign data can be better used to validate AIRS CH_4 in the level where AIRS has the largest sensitivity in the middle to upper troposphere. To convolve the "truth" of aircraft measurement using the averaging kernels and a priori profiles (Xiong et al., 2008), we need to extrapolate the in-situ aircraft data to higher altitudes, and this was done by using the monthly average of model simulated data from the TM3 (Houwelling et al., 2006).

Both the aircraft measurement and the convolved "truth" derived from aircraft measurement are used for comparison with the match-up data from AIRS. Only aircraft measurements whose latitudes are larger than 25° N and have valid measurements in the layer of 50 to 250 hPa below the tropopause are used in this paper.

4 Validation of AIRS mid-upper tropospheric CH_4 using aircraft measurements

4.1 NOAA/ESRL/GMD aircraft measurements

These aircraft measurements of CH_4 are made by routinely collecting the air samples using 0.7 L silicate glass flasks on biweekly to monthly aircraft flights at 22 sites, operated by the NOAA/ESRL/GMD Carbon Cycle Group. Air samples were collected using turboprop aircraft with maximum altitude limits of 300–350 hPa. Individual flights required about 1.5 h to complete. Measurements were made by collecting samples of air (approximately 0.7 liter volume at 40 psa) in glass containers. Twelve to twenty flasks are held in a suitcase-sized container, and collection of air in a single flask at a unique altitude allows a sampling vertical resolution up to 400 m in the boundary layer. After each flight the flask packages are shipped to the NOAA laboratory in Boulder, Colorado for trace gas analysis. The locations and profiles for 22 sites used can be referred to

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Xiong et al. (2008). However, data in Rarotonga, Cook Islands (RTA) and Molokai Island, Hawaii, United States (HAA) are not used as they are in the tropics. For the rest 20 sites, the mean of the measured CH_4 mixing ratio in the layer of 50–250 hPa below the tropopause is compared with AIRS.

Figure 3 shows the comparison of the mid-upper tropospheric CH_4 from AIRS with the aircraft measurements operated by NOAA/ESRL/GMD from August 2003 to February 2006. Note that only those profiles that have measurement data at the layer of 50–250 hPa below the tropopause were used. Along the x-axis is the aircraft measurement not convolved (triangles) and the convolved one (diamonds) using averaging kernels (AK), and along the y-axis is the mean of AIRS retrieved profiles within 24 h and 800 km of the aircraft measurement time and site location. The correlation coefficient using the convolved truth, 0.64, is a little larger than using real aircraft measurement, 0.57. Compared to the previous validation on the layer 358–459 and 459–596 hPa by Xiong et al. (2008) (lower left and upper right panels of Fig. 9b), if we zoom in the mid-high latitude regions, the correlation between AIRS and aircraft measurements has been significantly improved. The bias and rms error compared with the aircraft measurement (non-convolved) is −0.57% and 1.0%, respectively. However, the bias and rms errors using the convolved truth are a little larger than using the aircraft measurements directly. This is out of expectation but understandable as NOAA/ESRL/GMD aircraft data are mostly below 350 hPa. While applying to AK to convolve the aircraft measurements, we need to extrapolate the aircraft data to upper altitudes using the monthly mean model data, which brings some uncertainties to the "convolved" truth.

4.2 INTEX-A

The INTEX-A field mission was conducted in the summer of 2004 (1 July to 15 August 2004) over North America (NA) and the Atlantic. This effort had a broad scope to investigate the transport and chemistry of long-lived greenhouse gases, oxidants and their precursors, aerosols and their precursors, as well their relationship with radiation and climate. NASA's DC-8 and J-31 were joined by aircraft from a large number of Euro-

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pean and North American partners to explore the composition of the troposphere over NA and the Atlantic as well as radiative properties and effects of clouds and aerosols in a coordinated manner (Singh et al., 2006). Air sample is collected into a conditioned, evacuated 2-L stainless steel canister equipped with a bellows valve, and is returned to our UC-Irvine laboratory for CH_4 analysis using gas chromatography (GC, HP-5890A) with flame ionization detection (FID). The use of primary CH_4 calibration standards dating back to late 1977 ensures that our measurements are internally consistent. The measurement accuracy is ±1% and the analytical precision at atmospheric mixing ratios is about 1 ppbv (Simpson et al., 2002, 2006).

Comparison of the mid-upper tropospheric CH_4 derived AIRS with aircraft measurement from INTEX-A is illustrated in Fig. 4. The number of aircraft profiles used is N=78. As expected, the correlation coefficient, bias and rms errors using the convolved truth is smaller than using non-convolved truth. The bias and rms error compared with the aircraft measurement (non-convolved) is 0.80% and 1.06%, respectively, and the correlation coefficient of the mid-upper tropospheric CH_4 derived AIRS with aircraft measurement (non-convolved) is 0.68.

4.3 INTEX-B

Intercontinental Chemical Transport Experiment-B (INTEX-B) was a major NASA led multi-partner atmospheric field campaign completed in the spring of 2006 (http://cloud1.arc.nasa.gov/intex-b/). INTEX-B was performed in two phases. In its first phase (1–21 March), INTEX-B operated as part of the MILAGRO campaign with a focus on observations over Mexico and the Gulf of Mexico. In the second phase (17 April–15 May), the main INTEX-B focus was on trans-Pacific Asian pollution transport. Multiple airborne platforms carrying state of the art chemistry and radiation payloads were flown in concert with satellites and ground stations during the two phases of INTEX-B (Singh et al., 2009).

Compared to the results from INTEX-A, most measurements are near tropics, so only 26 profiles with the latitude larger than 25° N is picked. Both the bias and rms error

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of AIRS CH_4 compared with aircraft measurements in INTEX-B (Fig. 5) are larger than INTEX-A, and the bias of AIRS mid-upper tropospheric CH_4 is −0.49%, and the rms error is 1.10%. The large negative bias is mainly from the profiles near the tropical regions, which may be associated with the tuning of 2% increase in CH_4 absorption coefficients for strong absorption channels (Xiong et al., 2008), indicating some adjustment of the turning will be required for the future improvement of CH_4 retrievals from AIRS. The correlation coefficient of the mid-upper tropospheric CH_4 derived AIRS with aircraft measurement (non-convolved) is 0.63, which is just a little smaller than in INTEX-A.

Further comparison of the biases using three different aircraft data sets indicates that INTEX-A shows a positive bias while NOAA and INTEX-B aircrafts show negative bias (Fig. 6). One reason is that measurements in INTEX-A were taken in the summer in the mid-high Northern Hemisphere. If we selected the profiles in July and August, there are 89 profiles among all the NOAA/ESRL/GMD profiles from August 2003 to February 2006, and retrieval bias of AIRS compared with the aircraft measurement (convolved) becomes −0.11%. This indicates some dependence of the retrieval error with the season. From the biases for different months estimated from NOAA/ESRL/GMD data (Fig. 6), we can see that the bias in the summer is smaller than in other seasons. As discussed before, the moist air during the summer pushes the maximum sensitive layer of AIRS to a higher altitude region than other seasons, and the lapse rate and the degree of freedom in the retrievals the summer is relatively larger (Xiong et al., 2008). In Fig. 6 we plotted the biases from INTEX-A and -B in the time corresponding to their own aircraft measurements. From Fig. 6 we can see that the bias using INTEX-B data is very close to that using NOAA aircraft data, but the bias using INTEX-A data is about 0.5% higher than that using NOAA aircraft data. We found that in INTEX-A, there are 9 profiles with a positive bias larger than 1.0%, and the CH_4 mixing ratios of these profiles are less than 1780.0 hPa. Possibly the intrusion of air from stratosphere to troposphere were sampled for these cases, however, AIRS retrievals is from a large area within 800 km, so some of the large positive bias is from the inappropriate match-

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up. We noted that there is a large forest fires in Alaska during the campaign of INTEX-A, this unusual event may bring some additional uncertainty.

4.4 Examples of seasonal variation of mid-upper tropospheric CH_4 from AIRS and its comparison with aircraft measurements

Another way to evaluate whether the mid-upper tropospheric CH_4 derived from AIRS can characterize the CH_4 in the real atmosphere is to check whether its seasonal cycle is consistent with aircraft measurements of NOAA/ESRL/GMD. As an example, data from Poker Flat, Alaska is plotted in Fig. 7. Each point for the AIRS data is an average of the retrievals within 800 km around the Poker Flat, while the blue squares denote the average of aircraft measurements in layer from 50 to 250 hPa below the mean tropopause of the corresponding AIRS match-up profiles. The solid line is the running mean in 30 days for AIRS derived tropospheric CH_4.

Comparison of AIRS CH_4 with aircraft measurements shows that overall their seasonal cycles are in a good agreement (Fig. 7). Specifically, they both show the decrease of CH_4 in April–May with the minimum occurring in late May to early June. It is evident from AIRS that the CH_4 increases significantly in the early summer from late June to July. Although the large difference in the sampling interval between AIRS and aircraft measurement makes this comparison difficult, the early summer increase was sampled successfully by aircraft measurements. We found that for all the aircraft profiles from May to July in 2004 and 2005, and the one with the largest CH_4 mixing ratio occurs in 10 July 2004 and 29 June 2005, respectively. A slight decrease is evident in late July to August from both AIRS and aircraft measurement, but the decrease is more significant from aircraft measurement. The time of this decrease of CH_4 is consistent with the occurring of the minimum of CH_4 at the marine boundary layer, and is attributed to the photochemical loss of CH_4. Increase of CH_4 in late August to September is also evident from both AIRS and aircraft measurement, which has been linked with wetland emissions (Dlugokencky et al., 1995).

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Since the aircraft measurements are made only 1 or 2 times a month, a monthly climatology of all aircraft measurements from 1999 to February 2006 with the annual mean in each year removed have been plotted in the lower panel. The number of aircraft sampling is increased significantly, so it is better to view the seasonal cycle from the running means in 30 days (blue dash line). Two years' AIRS data within an area of 800 km in 2004 and 2005 are used to derive the seasonal cycle (dark solid line). One obvious difference between them is the magnitudes of CH_4 increase in the early summer and the decrease in the August from aircraft, which are larger from aircraft measurements than from AIRS. The spread of CH_4 from aircraft measurement among different year is large, so the difference of the seasonal cycle from AIRS with that from aircraft measurements is associated with the difference in the sampling time.

Another way to add the sampling of aircraft measurements is to combine measurements from different sites that are close to each other. This can be done by mixing all the measurements from 13 sites in the region from 40° N to 50° N and 70° W to 127° W in the North America. The seasonal cycle illustrated from the 30 days' running mean of all these aircraft data is compared with the seasonal cycle of AIRS averaged in this region (Fig. 8). Considering the correlation between AIRS retrieval with aircraft measurements is mostly less than 0.7, which implies that the AIRS can detect the variation of CH_4 in the real atmosphere less than ~50% (R^2=0.49), the seasonal cycle from aircraft is in reasonable agreement with AIRS. The agreement of the seasonal cycle in 2005 is better than in 2004 as more aircraft measurements were available in 2005. The spread of aircraft measurements is partially due to the large latitudinal gradient between 40 and 50° N. From Fig. 8 we can also see that AIRS CH_4 is lower than aircraft measurements, and the mean bias in these two years is – 16.5 ppbv, which is consistent with the bias of –0.9% (Fig. 3).

Due to the large difference in the sampling time and location, comparison between AIRS and aircraft measurement is difficult. Since the first guess of CH_4 is a smooth function of latitude and pressure and has no seasonal variation in the retrieval of CH_4 from AIRS (Xiong et al., 2008), the seasonal variation of CH_4 and its reasonable agree-

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ment with aircraft measurement, as illustrated in Figs. 7 and 8, indicate that the mid-upper tropospheric CH_4 from AIRS provides valuable information of CH_4 in the atmosphere.

5 Summary and conclusions

Our analysis shows that the tropopause correlates well with the maximum sensitive level in the middle to high latitude regions. As the most sensitive levels is usually located between 50 to 250 hPa below the tropopause, a new approach was developed to use the AIRS CH_4 in this layer to characterize the variation of CH_4 in the mid-upper troposphere. This method is only applicable in the mid-high latitude. The mean CH_4 between 50 to 250 hPa below the tropopause using AIRS retrievals will be derived as a new product of CH_4 from AIRS. This new product together with the current products will be available to the users.

Validation to this product has been made using aircraft data from NOAA/ESRL/GMD and the campaigns INTEX-A and -B. Comparison results show the rms error is less than 1.2%. The accuracy is less than ±1%, depending on different seasons. The correlation coefficients are about 0.6–0.7. Another way to evaluate AIRS CH_4 was made by comparing the season cycle from AIRS mid-upper tropospheric CH_4 with NOAA/ESRL/GMD aircraft measurements. Due to the fact that the aircraft measurements are sparse, we binned the aircraft samples to derive the seasonal cycle. In one approach the samples of multiple years from 1999–2006 at Poker Flat, Alaska were binned into a single year, and in another approach the samples from multiple sites between 40–50° N in North America were combined. The comparison results indicated that the seasonal cycle of AIRS mid-upper tropospheric CH_4 is in a reasonable agreement with that from aircraft measurement, suggesting that this new information-based product provides some valuable information of CH_4 in the mid-upper troposphere. This new product is particularly valuable for the study of the seasonal variation of CH_4 in the high Northern Hemisphere, where the measurements are sparse and our estima-

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tions of CH_4 emissions from northern wetlands/permafrost in these regions have large uncertainties (Zhuang et al., 2009). However, due to the limit of aircraft data, further validations using the upcoming aircraft measurements and ground-based remote sensing will be on-going.

The uncertainties in AIRS retrievals include (1) the errors in atmospheric temperature and moisture profiles, surface temperature and emissivity derived from other channels of AIRS, (2) the errors in cloud/clearing, and (3) the noise of the sensor. The errors in cloud/clearing could be the largest source of uncertainty and is hard to estimate, but no systematic bias has been found, and on average the difference between the AIRS retrieved CH_4 between clear cases (with the cloud fraction less than 0.1) and cloud cases (with the cloud fraction over 0.8) is usually less than 1.0%. Uncertainty in the temperature and surface pressure will impact the computation of tropopause, but the error in the tropopause height to this new product should be small because the layer of 50 to 250 hPa below the tropopause used is a broad layer with a thickness of about 3–3.5 km. However, the changing the trapezoid functions has some influence on the maximum sensitive level, so for other thermal infrared sounders using different trapezoid functions, an optimized layer below the tropopause may be used. Difference in the calibration of aircraft measurement among these three sets will also contribute to some error in these validations, but this impact should be a small factor. On the contrary, a larger uncertainty can be from the aircraft sampling interval, altitude and speed of flight, as well as the preprocessing of these in-situ data to obtain the profiles (as a mean profile corresponding to one time and location). The aircraft measurements from NOAA/ESRL/GMD have mostly not covered the whole altitude range of the layer from 50 to 250 hPa below the tropopause used by AIRS, while data from INTEX-A and -B have better coverage of these altitude ranges. This difference could be one reason why the validation results from INTEX-A and -B are a little different from using NOAA data. Even though CH_4 is well-mixed in the troposphere, its temporal variation from the aircraft measurement to AIRS measurement in a different time of the same day can be significant as compared to the uncertainty in our validation. However, all the aircraft

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measurements were not carried out for the purpose to validate AIRS trace products, so we have not other choices.

As an new product following the AIRS retrievals, this method to derive mid-upper tropospheric CH_4 based on the tropopause height makes it easy to analyze the seasonal variation of CH_4 in the mid-upper troposphere without referring the averaging kernels. However, the averaging kernels still need to be used in many situations, for example, if we want to assimilate the AIRS products into the models to improve the estimation of CH_4 sources/sinks.

This method as well as the validation to the satellite retrieved CH_4 will be applied to retrievals from the EUMETSAT Infrared Atmospheric Sounding Interferometer (IASI) at NOAA/NESDIS/STAR. As more data becomes available, comparisons between AIRS and IASI will be made to extend our validation of these satellite products. This method is also applicable to other thermal infrared sounders, such as Cross-track Infrared Sounder (CrIS). It is also possible to use our method for other trace gas retrievals that use the NOAA AIRS/IASI/CrIS algorithm and thermal infrared sounder measurements, such as CO_2, N_2O and possibly CO.

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References

Aumann, H. H., Chahine, M. T., Gautier, C., Goldberg, M. D., Kalnay, E., McMillin, L. M., Revercomb, H., Rosenkranz, P. W., Smith, W. L., Staelin, D. H., Strow, L. L., and Susskind, J.: AIRS/AMSU/HSB on the aqua mission: Design, science objectives, data products, and processing systems, IEEE T. Geosci. Remote, 41, 253–264, 2003.

Clerbaux, C., Hadji-Lazaro, J., Turquety, S., Mégie, G., and Coheur, P.-F.: Trace gas measurements from infrared satellite for chemistry and climate applications, Atmos. Chem. Phys., 3, 1495–1508, 2003, http://www.atmos-chem-phys.net/3/1495/2003/.

Crevoisier, C., Nobileau, D., Fiore, A. M., Armante, R., Chédin, A., and Scott, N. A.: A new insight on tropospheric methane in the Tropics - first year from IASI hyperspectral infrared observations, Atmos. Chem. Phys. Discuss., 9, 6855–6887, 2009, http://www.atmos-chem-phys-discuss.net/9/6855/2009/.

De Mazière, M., Vigouroux, C., Bernath, P. F., Baron, P., Blumenstock, T., Boone, C., Brogniez, C., Catoire, V., Coffey, M., Duchatelet, P., Griffith, D., Hannigan, J., Kasai, Y., Kramer, I., Jones, N., Mahieu, E., Manney, G. L., Piccolo, C., Randall, C., Robert, C., Senten, C., Strong, K., Taylor, J., Tétard, C., Walker, K. A., and Wood, S.: Validation of ACE-FTS v2.2 methane profiles from the upper troposphere to the lower mesosphere, Atmos. Chem. Phys., 8, 2421–2435, 2008, http://www.atmos-chem-phys.net/8/2421/2008/.

Dlugokencky, E. J., Houweling, S., Bruhwiler, L., Masarie, K. A., Lang, P. M., Miller, J. B., and Tans, P. P.: Atmospheric methane levels off: Temporary pause or a new steady-state?, Geophys. Res. Lett., 30(19), 1992, doi:10.1029/2003GL018126, 2003.

do Carmo, J. B., Keller, M., Dias, J. D., de Camargo, P. B., and Crill, P.: A source of methane from upland forests in the Brazilian Amazon, Geophys. Res. Lett., 33, L04809, doi:10.1029/2005GL025436, 2006.

Frankenberg, C., Meirink, J. F., van Weele, M., Platt, U., and Wagner, T.: Assessing methane emissions from global space-borne observations, Science, 308, 1010–1014, 2005.

Frankenberg, C., Bergamaschi, P., Butz, A., Houweling, S., Meirink, J. F., Notholt, J., Petersen, A. K., Schrijver, H., Warneke, T., and Aben, I.: Tropical methane emissions: A revised view from SCIAMACHY onboard ENVISAT, Geophys. Res. Lett., 35, L15811, doi:10.1029/2008GL034300, 2008.

ACPD

9, 16331–16360, 2009

Houweling, S., Röckmann, T., Aben, I., Keppler, F., Krol, M., Meirink, J. F., Dlugokencky, E. J., and Frankenberg, C. : Atmospheric constraints on global emissions of methane from plants, Geophys. Res. Lett., 33, L15821, doi:10.1029/2006GL026162, 2006.

IPCC: Intergovernmental Panel on Climate Change, The Physical Science Basis, edited by: Solomon, S., Qin, D., Manning, M., Chen, Z., Marquis, M., Averyt, K. B., Tignor, M., and Miller, H. L., Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 500–590, 2007.

Kanamitsu, M., Alpert, J. C., Campana, K. A., Caplan, P. M., Deaven, D. G., Iredell, M., Katz, B., Pan, H.-L., Sela, J., and White, G. H.: Recent changes implemented into the global forecast system at NMC, Weather Forecast., 6, 425–435, 1991.

Keppler, F., Hamilton, J. T. G., Braß, M., and Röckmann, T.: Methane emissions from terrestrial plants under aerobic conditions, Nature, 439, 187–191, 2006.

Maddy, E. S. and Barnet, C. D.: Vertical resolution estimates in Version 5 of AIRS operational retrievals, IEEE T. Geosci. Remote, 46(8), 2375–2384, 2008.

Mastepanov, M. and Christensen, T. R.: Large tundra methane burst during onset of freezing, Nature, 456, p. 628, 2008.

Nassar, R., Bernath, P. F., Boone, C. D., Manney, G. L., McLeod, S. D., Rinsland, C. P., Skelton, R., and Walker, K. A.: Stratospheric abundances of water and methane based on ACE-FTS measurements, Geophys. Res. Lett., 32, L15S04, doi:10.1029/2005GL022383, 2005.

Pan, L., Gille, J. C., Edwards, D. P., Bailey, P. L., and Rodgers, C. D.: Retrieval of tropospheric carbon monoxide for the MOPITT experiment, J. Geophys. Res., 103(D24), 32 277–32 290, 1998.

Payan, S., Camy-Peyret, C., Oelhaf, H., Wetzel, G., Maucher, G., Keim, C., Pirre, M., Huret, N., Engel, A., Volk, M. C., Kuellmann, H., Kuttippurath, J., Cortesi, U., Bianchini, G., Mencaraglia, F., Raspollini, P., Redaelli, G., Vigouroux, C., De Mazière, M., Mikuteit, S., Blumenstock, T., Velazco, V., Notholt, J., Mahieu, E., Duchatelet, P., Smale, D., Wood, S., Jones, N., Piccolo, C., Payne, V., Bracher, A., Glatthor, N., Stiller, G., Grunow, K., Jeseck, P., Te, Y., and Butz, A.: Validation of version-4.61 methane and nitrous oxide observed by MIPAS, Atmos. Chem. Phys., 9, 413–442, 2009, http://www.atmos-chem-phys.net/9/413/2009/.

Payne, V. H., Clough, S. A., Shephard, M. W., Nassar, R., and Logan, J. A.: Information-centered representation of retrievals with limited degrees of freedom for signal: Application to methane from the Tropospheric Emission Spectrometer, J. Geophys. Res., 114, D10307,

doi:10.1029/2008JD010155, 2009.
Razavi, A., Clerbaux, C., Wespes, C., Clarisse, L., Hurtmans, D., Payan, S., Camy-Peyret, C., and Coheur, P. F.: Characterization of methane retrievals from the IASI space-borne sounder, Atmos. Chem. Phys. Discuss., 9, 7615–7643, 2009, http://www.atmos-chem-phys-discuss.net/9/7615/2009/.
Rigby, M., Prinn, R. G., Fraser, P. J., et al.: Renewed growth of atmospheric methane, Geophys. Res. Lett., 35, L22805, doi:10.1029/2008GL036037, 2008.
Park, M., Randel, W. J., Kinnison, D. E., Garcia, R. R., and Choi, W.: Seasonal variation of methane, water vapor, and nitrogen oxides near the tropopause: Satellite observations and model simulations, J. Geophys. Res.-Atmos., 109, D03302, doi:10.1029/2003JD003706, 2004.
Rodgers, C. D.: Inverse Methods for Atmospheric Sounding, edited by: Hackensack, N. J., World Scientific Publishing Company, 2000.
Simpson, I. J., Chen, T.-Y., Blake, D. R., and Rowland, F. S.: Implications of the recent fluctuations in the growth rate of tropospheric methane, Geophys. Res. Lett., 29(10), 1479, doi:10.1029/2001GL014521, 2002.
Simpson, I. J., Rowland, F. S., Meinardi, S., and Blake, D. R.: Influence of biomass burning during recent fluctuations in the slow growth of global tropospheric methane, Geophys. Res. Lett., 33, L22808, doi:10.1029/2006GL027330, 2006.
Singh, H. B., Brune, W. H., Crawford, J. H., Flocke, F., and Jacob, D. J.: Chemistry and transport of pollution over the Gulf of Mexico and the Pacific: spring 2006 INTEX-B campaign overview and first results, Atmos. Chem. Phys., 9, 2301–2318, 2009, http://www.atmos-chem-phys.net/9/2301/2009/.
Singh, H. B., Brune, W. H., Crawford, J. H., Jacob, D. J., and Russell, P. B.: Overview of the summer 2004 Intercontinental Chemical Transport Experiment – North America (INTEX-A), J. Geophys. Res., 111, D24S01, doi:10.1029/2006JD007905, 2006.
Strow, L. L., Hannon, S. E., De Souza-Machado, S., Motteler, H. E., and Tobin, D.: An overview of the airs radiative transfer model, IEEE T. Geosci. Remote, 41, 303–313, 2003.
Susskind, J., Barnet, C. D., and Blaisdell, J. M.: Retrieval of atmospheric and surface parameters from AIRS/AMSU/HSB data in the presence of clouds, IEEE T. Geosci. Remote, 41, 390–409, 2003.
Walter, K. M., Zimov, S. A., Chanton, J. P., et al.: Methane bubbling from Siberian thaw lakes as a positive feedback to climate warming, Nature, 443, 71–75, doi:10.1038/nature05040,

2006.
Walter, K. M., Smith, L. C., and Chapin III, F. S.: Methane bubbling from northern lakes: present and future contributions to the global methane budget, Philos. T. R. Soc. A, 365, 1856, 1657–1676, 2007.
Xiong, X., Barnet, C. D., Maddy, E., Sweeney, C., Liu, X., Zhou, L., and Goldberg, M.: Characterization and Validation of Methane Products from the Atmospheric Infrared Sounder (AIRS), J. Geophys. Res., 113, G00A01, doi:10.1029/2007JG000500, 2008.
Xiong, X., Houweling, S., Wei, J., Maddy, E., Sun, F., and Barnet, C.: Methane plume over south Asia during the monsoon season: satellite observation and model simulation, Atmos. Chem. Phys., 9, 783–794, 2009, http://www.atmos-chem-phys.net/9/783/2009/.
Zhuang, Q., Melack, J. M., Zimov, S., Walter, K. M., Butenhoff, C. L., and Khalil, M. A. K.: Global methane emissions from wetlands, rice paddies, and lakes, EOS T. Am. Geophys. Un., 90(5), 37–38, 2009.

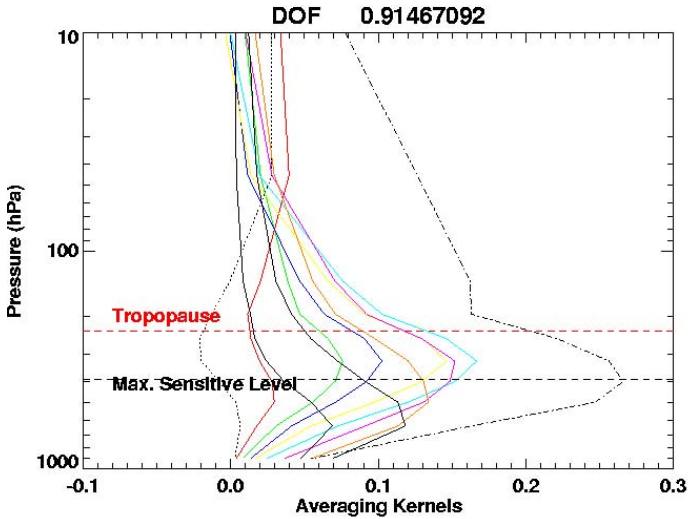

Fig. 1. The mean averaging kernels, the maximum sensitive level and the tropopause of averaged in 3 days from 1–3 August 2004 in Alaska (longitude=150° W, latitude=66° N).

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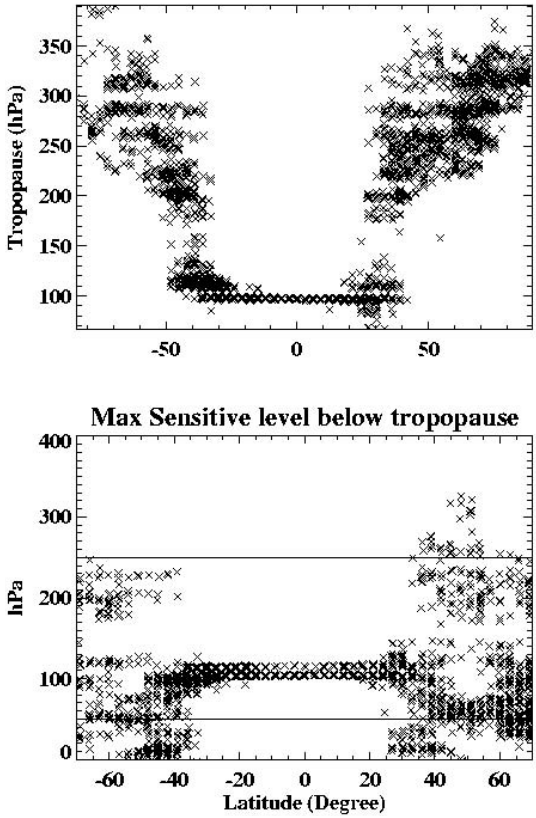

Fig. 2. Variation of tropopause with latitude (upper panel) and the difference of the pressure in the maximum sensitive level minus the tropopause with latitude(lower panel). The global data on 4 March 2006 are used.

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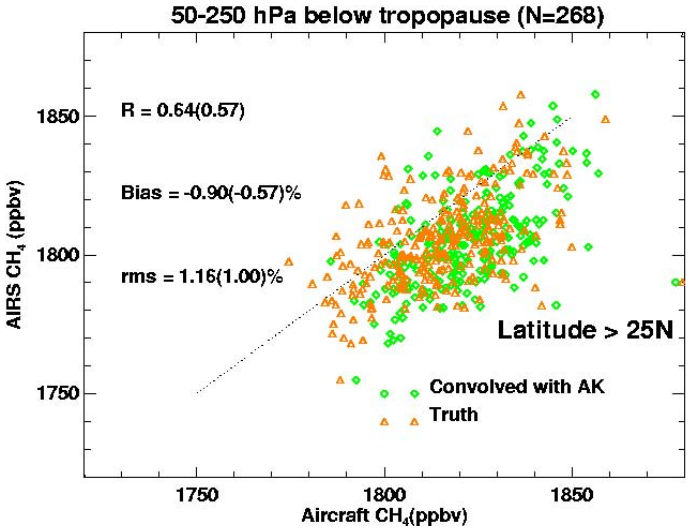

Fig. 3. Scatter plot of AIRS mid-upper tropospheric CH_4 vs. in-situ aircraft observations at 20 sites (N=268) run by NOAA/ESRL from August 2003 to February 2006. AIRS data is from the grid run of 3×3 degrees in latitude and longitude, and the mean of AIRS retrievals within 24 h and 800 km of the measurement time and site location are used. Triangles are the mean of aircraft profiles from 50 to 250 below the tropopause and squares are the convolved mean using averaging kernels. Non-convolved statistics are shown in parenthesis.

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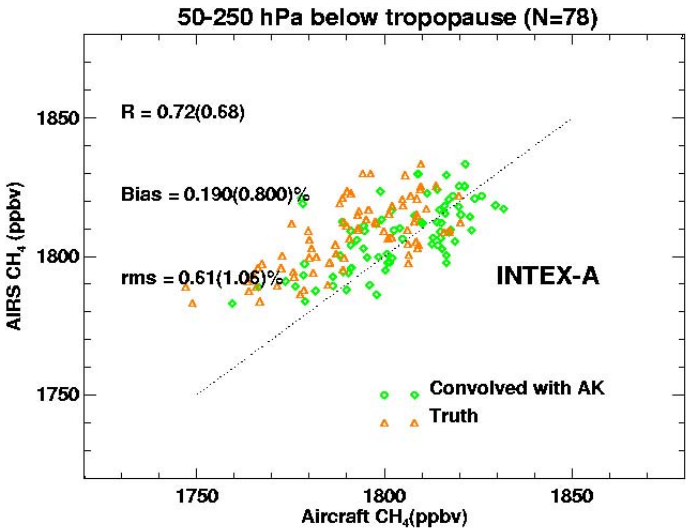

Fig. 4. Same as Fig. 3 but using the DC-8 in situ aircraft profiles from the campaign INTEX-A (1 July to 15 August 2004). The number of the aircraft profiles used is N=78.

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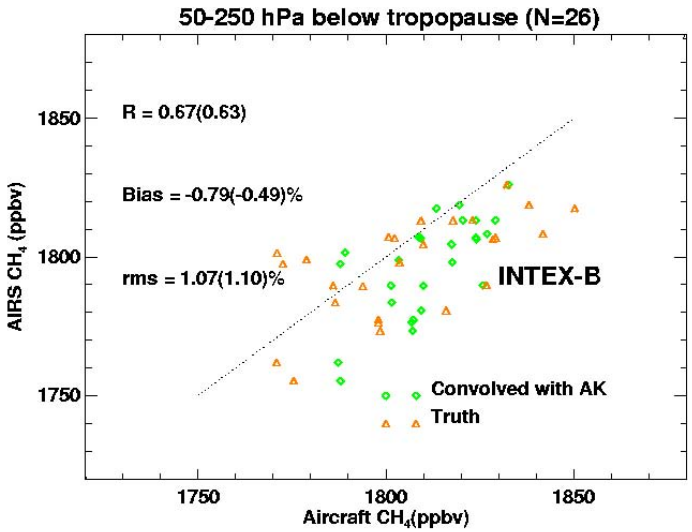

Fig. 5. Same as Fig. 4 but using data from the campaign INTEX-B (spring of 2006) with 26 aircraft profiles in latitude above 25° N.

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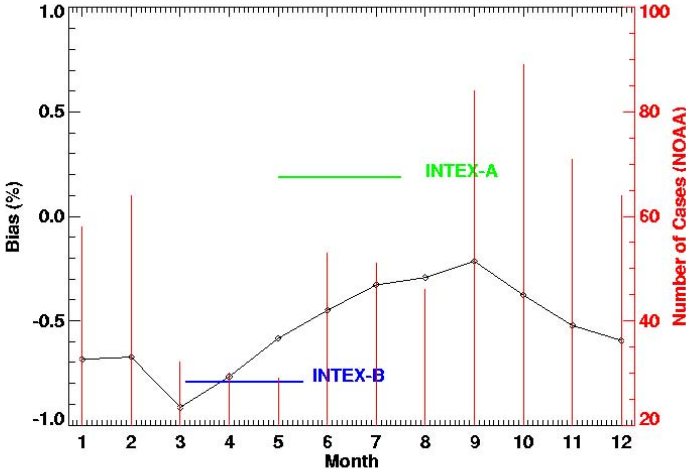

Fig. 6. Biases of AIRS CH_4 for different months using NOAA/ESRL/GMD data, and it comparison with data from the campaign INTEX-A and -B. Aircraft data have been convolved using the averaging kernels.

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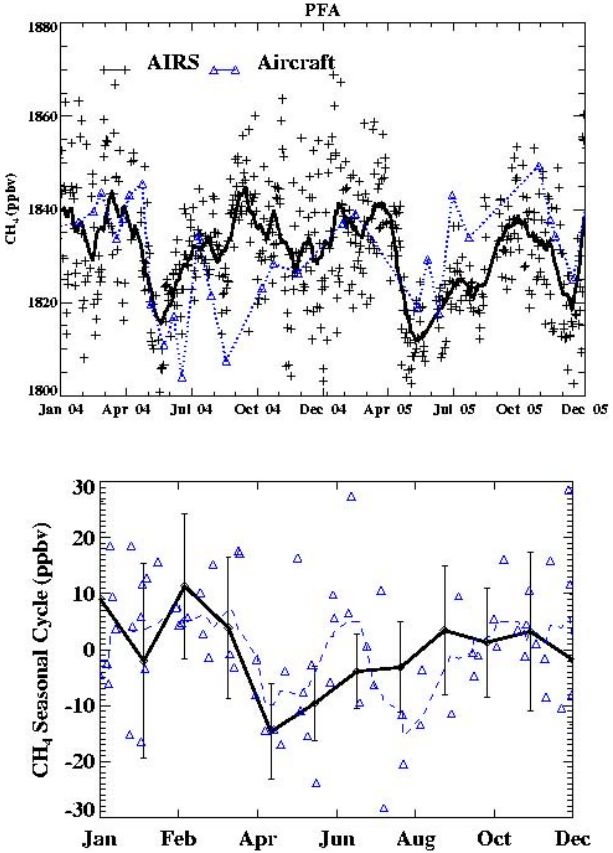

Fig. 7. Seasonal variation of mid-upper tropospheric CH_4 from AIRS and its comparison with NOAA/ESRL/GMD aircraft measurements at Poker Flat, Alaska (PFA). Black solid line is the running means of AIRS CH_4 over a 30 day window, triangles are aircraft measurements. Lower panel is their seasonal cycle after removing the annual mean of each year using aircraft data from 1999 to February 2006. Black line is the mean from AIRS retrievals with standard deviation as error bar.

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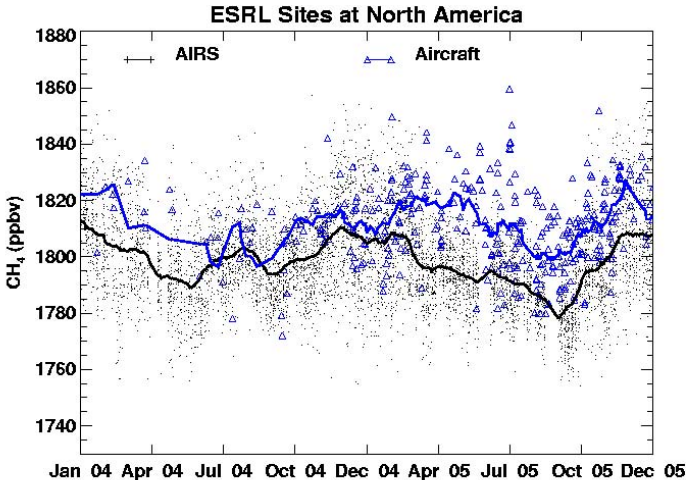

Fig. 8. Variation of mid-upper tropospheric CH_4 from AIRS from 2004 to 2005, and its comparison with NOAA/ESRL/GMD aircraft measurements at 13 sites in region 40° N–50° N, 70° W–127° W in the North America. Solid lines are the running means in 30 days (Black is AIRS, and Blue is aircraft measurements).

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