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Spatiotemporal variations in mid-upper tropospheric methane over China from satellite observations

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Spaceborne measurements by the Atmospheric Infrared Sounder (AIRS) on the EOS/Aqua satellite provide a global view of methane (CH_4) distribution in the mid-upper troposphere (MUT-CH_4). The focus of this study is to analyze the spatiotemporal variations in MUT-CH_4 over China from 2003 to 2008. Validation of AIRS CH_4 products versus Fourier transform infrared profiles demonstrates that its RMS error is mostly less than 1.5%. A typical atmospheric methane profile is found that shows how concentrations decrease as height increases because of surface emissions. We found that an important feature in the seasonal variation in CH_4 is the two peaks that exist in summer and winter in most parts of China, which is also observed in *in-situ* measurements at Mt. Waliguan, Qinghai Province, China (36.2879°N 100.8964°E, 3810 m). Also, in the summer, only one peak existed in western and southern China since there are no more significant anthropogenic sources in winter than at any other time of the year. Further analysis of the deseasonalized time-series of AIRS CH_4 in three fixed pressure layers of AIRS from 2003 to 2008 indicates that CH_4 in the Northern Hemisphere has increased abruptly since 2007, with no significant increase occurring before 2007. The increase in China is generally more significant than in other areas around the world, which again correlates with *in-situ* measurements at Mt. Waliguan.

satellite remote sensing, mid-upper tropospheric methane, spatiotemporal variation

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Atmospheric methane (CH_4) over the globe has risen dramatically since the pre-industrial era [1,2]. However, in recent years the rate of increase has not been so significant and even decreased in 1992 [3]. As one of the most important greenhouse gases, changes in atmospheric methane are very important for atmospheric chemical processes and climate change. Atmospheric methane accounts for about 20% of the total of 2.66 W m^{-2} of the anthropogenically produced greenhouse gas [4]. The mixing rate of atmospheric methane is less than that of carbon dioxide, but 25 times more effective on a per unit mass basis than carbon dioxide in absorbing long-wave radiation and its increase will affect the earth's radiation budget [5]. Atmospheric methane is one of the most important carbon compounds; it can change the concentrations of OH and CO and has an important role in the chemistry of O_3 and H_xO_y [6,7]. However, much work is still needed to build on intermittent and scattered observations from the 1960s and a systematic study from the 1980s [7,8]. From 1983, the World Meteorological Organization (WMO) began to co-ordinate the global *in-situ* measurement of methane. Quantification of methane emissions still has large uncertainties, mainly due to undersampling over most regions of the globe by the surface observation network. Even the results of aircraft observations have low spatial and temporal distributions due to their collection method. Therefore, the information available for the precise measurement or modeling of global distributions of methane and its variability are still unclear [8–11].

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In recent years, spaceborne remote sensing has been employed to measure CH_4 with large spatial and temporal coverage, which can effectively compensate for the lack of surface observations [12]. Two major types of measurement are (1) the measurement of the total column using the near-infrared spectrum and (2) the measurement of the mid-to-upper troposphere methane (MUT-CH_4) using the thermal infrared spectrum with bands of 7.66, 3.3 and 2.3 μm [13–23]. One of the spaceborne thermal infrared sounders in operation, the Atmospheric Infrared Sounder (AIRS), was launched in polar orbit (13∶30, ascending node) on the EOS/Aqua satellite in May 2002; CH_4 is one of its research products [13,14].

In this study, we validate the AIRS CH_4 product versus Fourier transform infrared (FTIR) profiles and *in-situ* measurement in China and discuss the spatiotemporal variations in MUT-CH_4 over China during 2003–2008.

1 Data and methods

1.1 AIRS measurement of CH_4

AIRS was launched in polar orbit (at 13∶30 local time, ascending node) on the EOS/Aqua satellite on 4 May 2002. It has high spectral resolution ($\lambda\backslash\Delta\lambda$=1200), and low noise [17]. The spatial resolution of AIRS is 13.5 km at nadir, and the swath width is 1650 km. Scan angles range from –49.5° to 49.5°, IFOV (instantaneous field of view) is 1.10, and in a 24-h period AIRS normally completely observes the globe twice.

The mixing rate of CH_4 data used in this study was downloaded from NASA (http://disc.sci.gsfc.nasa.gov). It includes three layer products with the maximum sensitive layer of AIRS (407.25, 306.75 and 206.25 hPa). The singular value decomposition method was used for CH_4 retrieval based on the 7.66 μm band with high spectra resolution [13–18]. Validation using *in situ* aircraft observations from 2003 to 2006 showed that the bias of the retrieved CH_4 profiles is approximately –1.4% to 0.1% and its root mean square (RMS) difference is about 0.5%–1.6% [13]. Validation of MUT-CH_4 using aircraft measurements from NOAA/ESRL/GMD and NASA Intercontinental Chemical Transport Experiment phases A and B demonstrated that in the HNH (high Northern Hemisphere) the RMS error of AIRS MUT-CH_4 is less than 1.2%, and the correlation coefficient between the AIRS MUT-CH_4 and aircraft measurements is about 0.6–0.7 [16]. This product has been used for the analysis of the variability and transport of atmospheric methane [16,17].

1.2 Ground-based measurement of CH_4

In situ CH_4 mixing ratio data from the MBL (mixed binding layer) were collected at Mt. Waliguan, Qinghai Province, China (36.2879°N 100.8964°E). The data were measured based on the GAW (Global Atmosphere Watch) standard from 1994 [24–26] and were shown to be of high quality [27,28]. In this work the data from Mt. Waliguan was compared with the AIRS CH_4 data.

FTIR measurements were carried out at a ground-based hyperspectral remote sensing laboratory located at the National Satellite Meteorological Center. The Bruker FTIR instrument (IFS 120 M, which made in Ettlingen of German) was used for observations. The specifications for atmospheric spectrum measurements are based on the Network for the Detection of Atmospheric Composition Change [29,30]. Observation of greenhouse gases has been carried out for several years [31].

2 Results and discussion

2.1 Validation of AIRS CH_4 measurements

In situ measurements of CH_4 from Mt. Waliguan were used for comparison with AIRS CH_4, which is generated by the flask method twice a week based on the cooperation of NOAA-CMDL and CAMS. Figure 1 shows the comparison

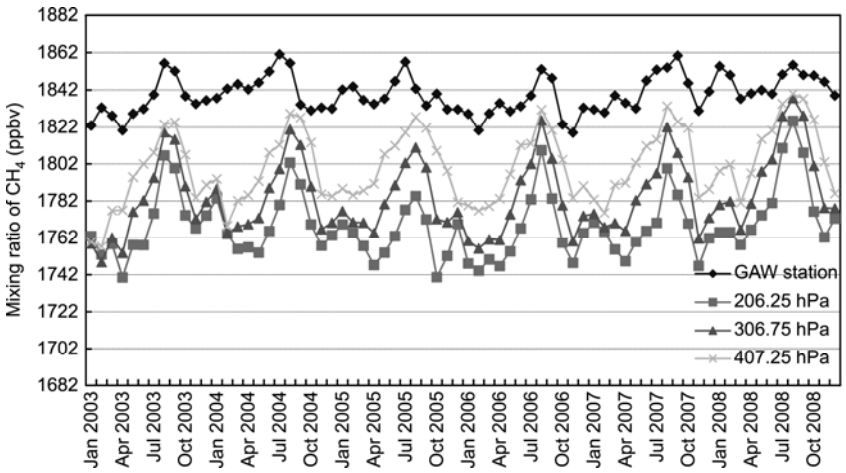

Figure 1 Comparison of AIRS CH_4 products and *in-situ* measurements from Mt. Waliguan.

of *in situ* data and AIRS products. It can be seen clearly that the seasonal cycle of AIRS is similar to that of *in-situ* measurements. CH_4 concentrations decreased with an increase in height.

The CH_4 profile data from ground-based remote sensing by FTIR also was used to quantitatively validate AIRS CH_4 measurements (Figure 2). FTIR measurements were made at the National Satellite Meteorological Center in Beijing from January to June, 2009. The results show that the AIRS retrieval products at three different heights is very close to the results from ground based measurement. The bias is 0.2%–1.1%, which is similar to aircraft validation results (0.5%–1.6%) [13].

2.2 Spatial distribution of CH_4 over China

Average distributions of CH_4 at a range of heights over China are shown in Figure 3. The CH_4 mixing ratio in the middle of the troposphere (407.25 hPa) is higher than that in the upper troposphere and at the top of the troposphere because of emission in the boundary layer. The highest CH_4 concentrations exist in the middle of the troposphere over the east, northeast, and south of China and cities in the northwest. This is associated with rice paddy and swamp emissions. The CH_4 mixing ratio is higher on the coast than that over the ocean because of the frequency of air exchange between the mainland and proximal marine environments. Low CH_4 concentrations are found on the Qinghai-Tibetan Plateau in the west of China as a result of the high altitude and minimal human activity. However, the highest CH_4 mixing ratios in the upper-middle and top of the troposphere are in areas of inland China, Inner Mongolia and Shenyang. Also, low CH_4 mixing ratios likely exist in southeastern China because of the effect of the marine environment.

Figure 4 shows the monthly variation in CH_4 over five different areas in China (Northeast: 42°–48°N, 120°–128°E; Northwest: 40°–44°N, 80°–92°E; East: 30°–40°N, 108°–118°E; West: 30°–38°N, 88°–100°E; South: 22°–26°N, 104°–116°E). CH_4 concentrations decrease with an increase in height. The lowest CH_4 mixing ratio at the top of the troposphere is in southern China. Also, in the upper troposphere, CH_4 mixes well in all regions. In the middle troposphere, a low CH_4 concentration center is located over western China due to the minimal occurrence of industrial and agricultural activities.

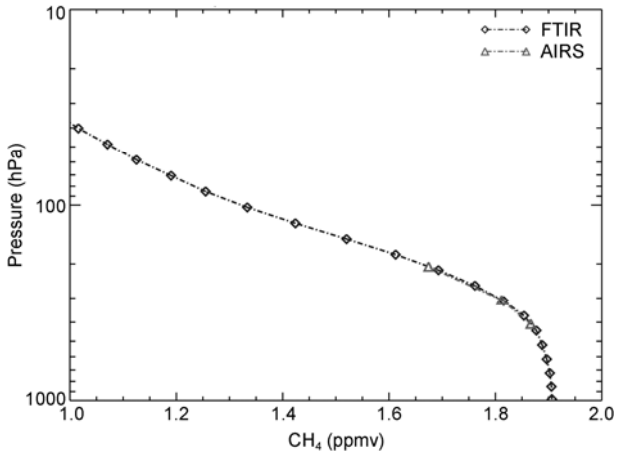

Figure 2 Averaged CH_4 products from FTIR and AIRS from January to June, 2009.

2.3 Seasonal cycle of CH_4 over China

The seasonal cycle of CH_4, averaged from 2003 to 2008 in different areas of China, is shown in Figure 5.

Two peaks are apparent in summer and winter over eastern, northeastern and northwestern China, which is consistent with the *in-situ* measurement for Mt. Waliguan [32]. Only one peak (in summer) occurs over southern and western China.

The reason for the two peaks of CH_4 over certain areas of China is related to CH_4 from biological and non-biological sources. The winter peak might be caused by CH_4 emission from fossil fuels burned for central heating in houses in the north of China. Also, the weather conditions in winter are conducive to the accumulation of pollutants. The summer peak may correspond to the most productive period of biological CH_4 emissions. Rice paddies, rivers and lakes are important CH_4 sources. Previous studies show that the

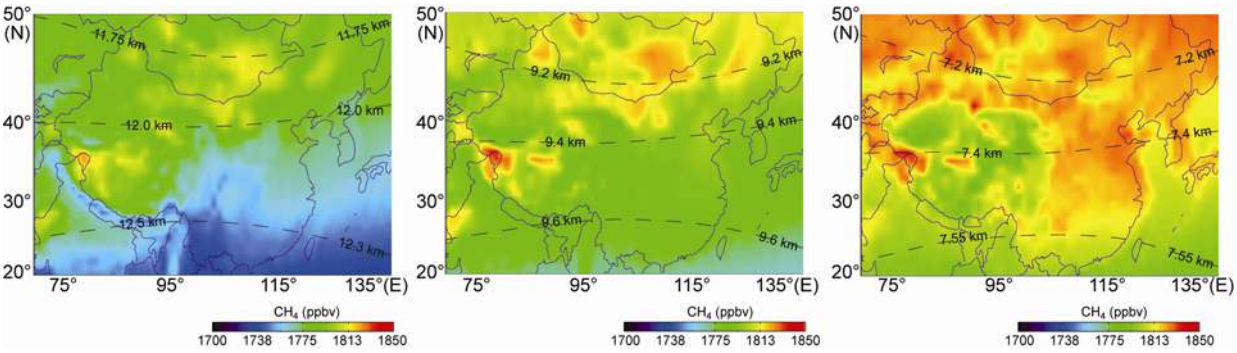

Figure 3 Distribution of average troposphere CH_4 over China from 2003 to 2008 (Left to right: 206.25, 306.75, 407.25 hPa).

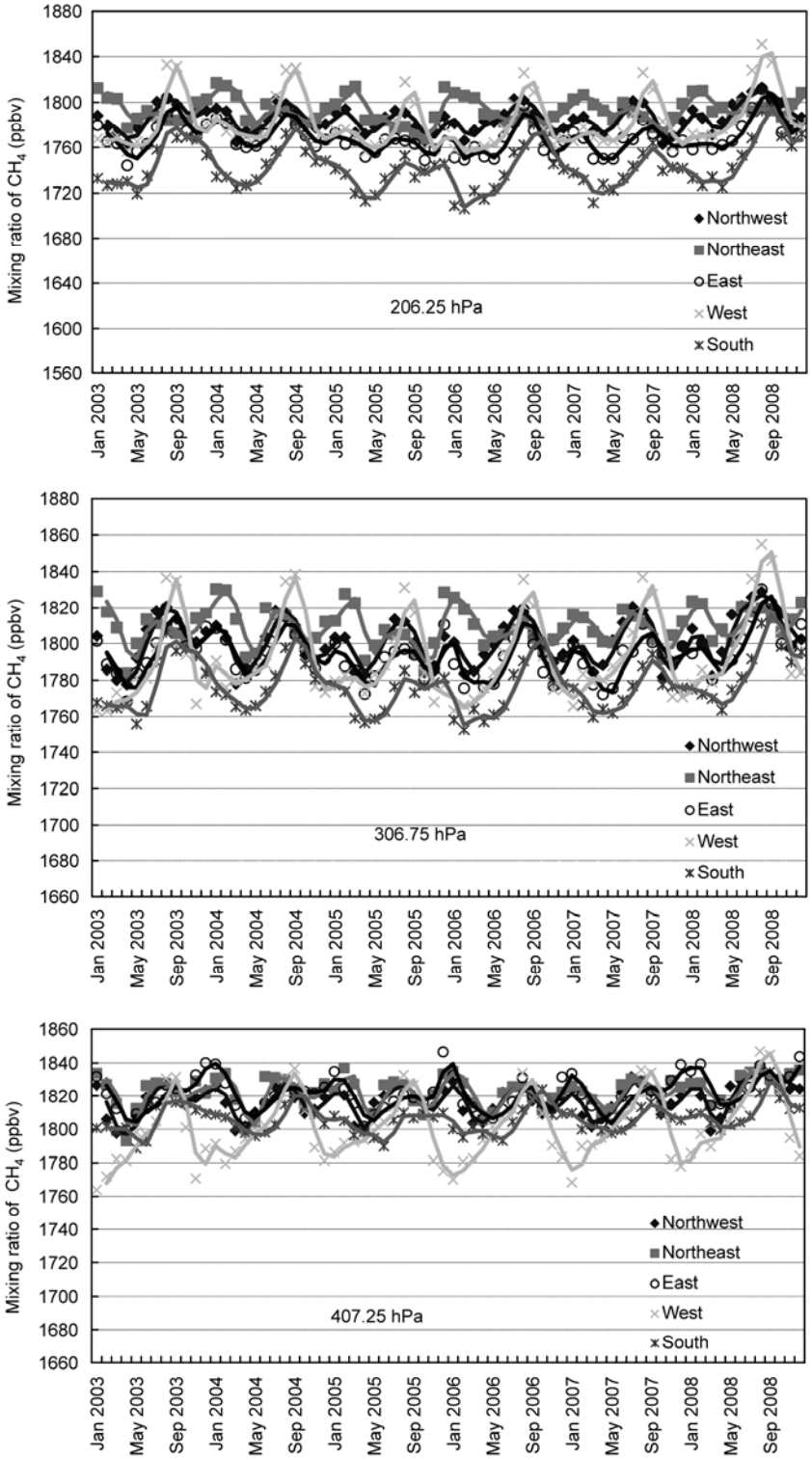

Figure 4 Monthly concentrations of CH_4 for different areas (as indicated) over China.

emission of CH_4 is closely related to temperature. There is little CH_4 emission from rice paddies, rivers and lakes when the temperature is lower than 15°C, whereas much more CH_4 is emitted at temperatures of 35°C than in 25°C [33–39]. Waste decomposition is another important biological source of methane. As cities expand and population grows, the organic waste in landfills decomposes and produces more and more CH_4. In the southern region of China, the significant anthropogenic sources of methane do not vary with season because there is not the same need for heating

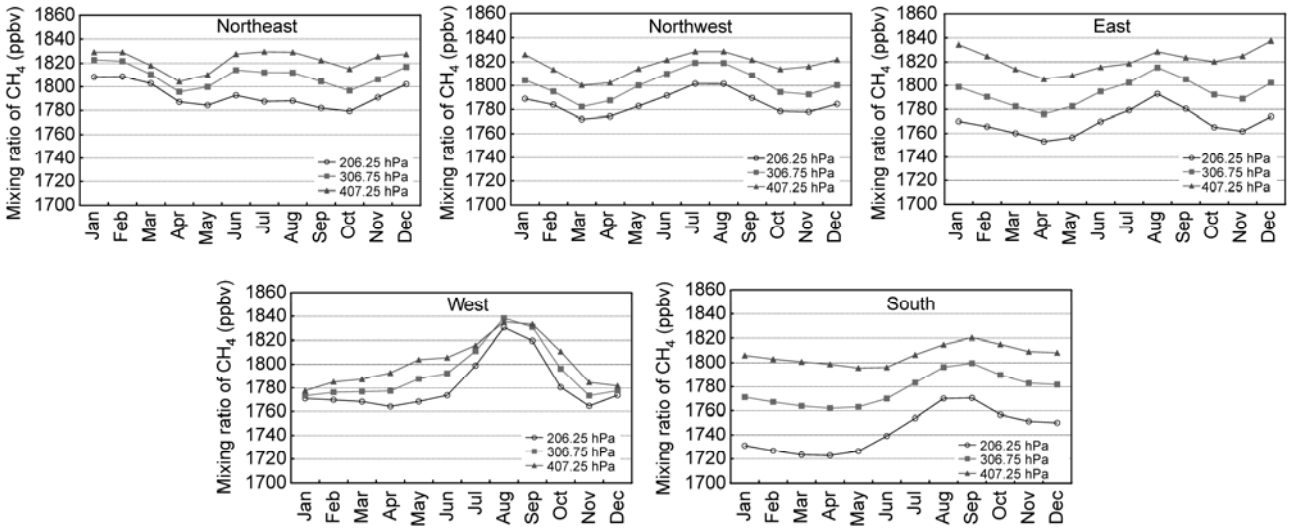

Figure 5 Seasonal variation in CH_4 averaged from 2003 to 2008 in different areas over China.

systems in winter. In the sparsely populated western region of China, there are few anthropogenic sources of CH_4 from industry and biological emissions are the main source of CH_4. In the summer, the high temperature and humid environment is conducive to CH_4 production, because abundant CH_4 comes from the decomposition of organic soil matter and can be released to the atmosphere. In colder periods, biologically sourced contributions to CH_4 emission are greatly decreased. Thus the lowest CH_4 mixing ratios occur in the winter in western China. Note that the seasonal cycle of methane based on AIRS observations is dependent on the methane emissions from sources and closely related to the ability of the atmosphere to transport them.

2.4 Long-term CH_4 trends over China

Figure 6 shows the variation in CH_4 mixing ratios in the troposphere over seven areas around the world from 2003 to 2008. A significant increase in CH_4 concentrations in the troposphere after 2007 can be found in most areas of the Northern Hemisphere. However, before 2007, the CH_4 mixing ratio is almost stable. That said, the CH_4 mixing ratio does show a slightly decreasing trend over Australia (in the southern hemisphere) before 2007 and then starts to grow. This conclusion is consistent with previous studies [15]. The average methane mixing ratio during the last 6 years over China is comparable with those of other countries in the Northern Hemisphere. Table 1 shows the difference in methane concentration between 2008 and the mean of 2003–2007 for six areas around the world. The most significant increases are found in China, followed by the United States and Canada. Note that CH_4 concentrations in Australia show a slight downward trend before 2007 and then start to growth in 2008. This conclusion is consistent with *in-situ* measurements [40–43]. Table 2 shows CH_4 growth rates in different layers of the troposphere over China from

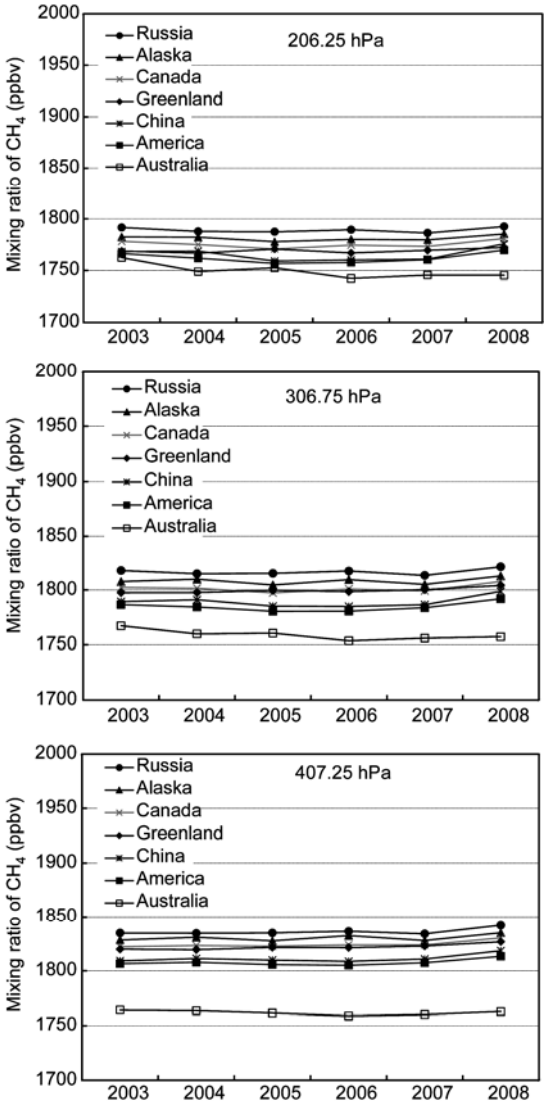

Figure 6 Tropospheric CH_4 growth rates for various sites around the world (2003–2008).

Table 1 Methane concentration differences between 2008 and the 2003–2007 average for typical areas

Area	Latitude	Longitude	Methane concentration differences (ppbv)		
			206.25 hPa	306.75 hPa	406.25 hPa
China	22°–42°N	90°–120°E	12.4	11.3	8.5
America	33°–48°N	124°–75°W	9.1	8.7	6.7
Canada	50°–65°N	130°–90°W	6.6	7.4	7.0
Alaska	60°–70°N	163°–142°W	4.9	5.5	5.6
Russia	55°–70°N	45°–135°E	4.0	5.4	7.0
Granada	65°–80°N	50°–20°W	3.8	5.5	5.7
Australia	30°–20°S	120°–150°E	–4.9	–2.1	1.2

Table 2 CH_4 mixing ratio growth rate over China from 2006 to 2008

Altitude (hPa)	Methane concentration (ppbv)			Growth rate (ppbv/a)
	2006	2007	2008	
206.25	1760.14	1760.81	1775.77	4.9
306.75	1785.04	1786.55	1798.77	6.9
407.25	1808.60	1810.77	1818.37	7.8
Mt. Waliguan	1832.67	1841.57	1846.22	6.8

2006 to 2008. The annual mean growth rate is 5–8 ppbv/a, which is similar to the Mt. Waliguan *in-situ* measurement (6.8 ppbv/a). However, *in-situ* measurements show that the CH_4 mixing ratio starts to growth significantly from 2006, which is a year earlier than the satellite result (i.e. starting in 2007). Note that the trend of CH_4 based on satellite observation still has some uncertainties with the 6-year average data. More data are needed for further trend analysis.

3 Conclusions

In this study, AIRS data were used to study the spatiotemporal distribution of CH_4 over China. The primary conclusions are as follows.

(1) Validation with ground-based measurements demonstrated that the retrieval RMS errors of AIRS CH_4 are mostly less than 1.5%, which proves that the AIRS product can be used for distribution analysis.

(2) Methane mixing ratios over China decrease with an increase in height because of the effects of human activities and natural emission of CH_4 from the boundary layer.

(3) Obvious bimodal seasonal variations in CH_4 concentrations exist in most areas of China, with the highest values in summer and the second highest in winter. Only the summer peak occurs in the western and southern areas of China.

(4) The mixing ratio of MUT-CH_4 is relatively stable before 2007 over several main regions of the Northern Hemisphere (including China). Significant growth is found after 2007, with China having the highest growth rate.

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