Spatial and Temporal Variations of Lower Tropospheric Methane During 2010–2011 in China

Yuyue Xu, Juanle Wang, Jiulin Sun, Yong Xu, and Warwick Harris

Abstract—Estimates of methane (CH₄) concentrations in the lower troposphere over the land surfaces of the world, and in more detail for China, were derived from data from the Atmospheric Infrared Sounder (AIRS). Derivation of CH₄ estimates is described, and these were validated with reference to CH₄ concentrations analyzed for air samples in three regions of China and the observations from three global baseline sites. The values of sample points and three global baseline sites were extracted from remotely sensed images. The correlation coefficients between remotely sensed geographical point estimates of CH₄ and observations were equal to 0.779 and 0.763. Seasonal and monthly variation of CH₄ concentrations for the 2010–2011 year and the spatial variation of these concentrations for the troposphere over China derived from remotely sensed data were mapped and interpreted with reference to possible sources of CH₄ emission. Methane concentrations were about 15 ppb higher in winter than summer. The changes in concentrations were about 50 ppb in Inner Mongolian Plateau between September and October 2010, northwest China between February and March 2011. Variations of CH₄ concentration are considered with reference to mixing of atmospheric gases by oceanic influences and to sources of CH₄ emissions including vegetation cover particularly wetland, crop, and pastoral land use.

Index Terms—AIRS, China, greenhouse gas, in-suit sensing, lower troposphere, methane.

I. INTRODUCTION

GREENHOUSE gases are considered to be a cause of global warming [1], [2]. Methane is the second most important anthropogenic greenhouse gas after carbon dioxide (CO₂) [3]–[6]. It has an indirect effect on climate through chemical feedbacks [3], [7], and its concentration has significantly exceeded the concentration of other organic compounds in the atmosphere [8]. It is at least 20 times more potent on a molecular basis in its greenhouse effect than CO₂ [9]. The atmospheric concentration of CH₄ has more than doubled over the past century [10], [11]. Recent studies of CH₄’s radiative impact over its entire atmospheric chemical lifetime suggest that it contributes 30%–38% of greenhouse warming [12]. Thus, increased knowledge of methane distribution and emissions is indispensable for a correct assessment of its impact on global temperature change [3]. Therefore, the study of methane distribution and contributions in the atmosphere is meaningful and necessary for the management of greenhouse gas emissions and impact evaluation. It is especially important for China which has higher levels of methane emission from agricultural (paddy fields) and industrial sources than any other country in the World [13].

In recent years, space-borne remote sensing providing large spatial and temporal coverage has been employed for the measurement of CH₄ [14]. The two major spectrums widely used in CH₄ measurement are near-infrared (NIR) and thermal infrared (TIR). The NIR measurements in operation include the SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY (SCIAMACHY) instrument onboard ENVISAT [15], [16], and the Greenhouse gases Observation SATellite (GOSAT). The GOSAT carries the Thermal And Near-infrared Sensor for carbon Observation (TANSO) [17]. The TIR measurements in operation include the Tropospheric Emission Spectrometer (TES) on NASA Earth Observing System (EOS) Aura mission [7], [18], the Infrared Atmospheric Sounding Interferometer (IASI) on European polar METeorological Operational Platform (METOP-1) [19], [20], and the Atmospheric Infrared Sounder (AIRS) on EOS Aqua mission [21].

This paper is based on the AIRS product. This is appropriate as SCIAMACHY provides remote sensing images from 2003 to 2009, and there are no data for 2010 and 2011 online, while the data can be obtained from AIRS. Another advantage of using the TIR (i.e., AIRS) rather than the NIR instrument is its capability to measure the CH₄ profile [22], thus providing some information about CH₄ distribution in the lower troposphere.

Current ground-based measurements of CH₄ in China are spatially sparse and not representative at large spatial scales. Observations from space now allow for the global detection of spatial and temporal variations in atmospheric CH₄ concentrations, thereby enabling identification of known sources and discovery of new ones. This is particularly the case for regions that...
are poorly sampled by existing surface measurement networks [23]. In order to do this research, we conducted three field surveys in three contrasting regions of China to collect gas samples.

The objectives of the research were to study CH$_4$ concentration in the lower atmosphere over China based on AIRS and to analyze the CH$_4$ spatiotemporal distribution. Section II defines the data sources used in this research. Section III describes the method of generating monthly average concentrations of CH$_4$. Section IV presents the results, and validates the results by sampling data and observations from global baseline sites. Finally, Section V draws conclusions.

II. DATA SOURCES

A. Field Measurement

China has a vast territory, and its regional differences are obvious. China’s topography is high in the northwest and low in the southeast. From 4 May to 12 July, 2011, sampling for near surface atmospheric CH$_4$ for the validation of data derived from the AIRS product was carried out in three regions of China. These regions were the Qinba mountain area in Shaanxi provinces, Poyang-Dongting Lakes of Jiangxi and Hunan provinces, and the Qinghai-Tibet Plateau in Qinghai province (Fig. 1). The three different regions are representative regions of the natural environment in China. Qinba mountain area lies in the center of China, while the other regions are in the southeast and northwest of China. The details of the three regional sampling programs are shown in Table I.

Equipment consisting of a 5-m-long iron pipe 8 cm in diameter, a plastic hose with 4-mm diameter bounded to the iron pipe, a three-way valve, a syringe, and a sealing bag was used to obtain air samples (Fig. 2). The height of the iron pipe and other dimensions of the equipment were determined by referring to “Greenhouse Gases sampling by Pyrex Flask” issued by the China Meteorological Administration [24]. The field methodologies and processes that yielded the data sets used in this work are reviewed here. The syringe and one end of the plastic hose were linked to the three-way valve, and then the plastic hose was sealed off by the syringe. Next, the sealing bag was linked to the three-way valve, and the air in the 5-m-high pipe was withdrawn and injected into the sealing bag through the pliable material by the syringe. After the bag was full of gas, it was sealed up and transported to the Institute of Geographical Sciences and Natural Resources Research, Chinese Academy of Sciences, Beijing, for trace gas analysis. After that, the CH$_4$ concentration ($X_{CH_4}$) of samples were measured by the mass spectrometer (Agilent 7890A gas chromatography (GC)). The CH$_4$ mixing ratios were derived from the samples to an accuracy of about 4 ppb against standard gases. This accuracy is a little larger than others’ measurements that have been made [25], and this is related to the quality of the inspection equipment used.

B. Remote Sensing Data

The AIRS is a nadir cross-track scanning infrared spectrometer on EOS (Earth Observing System)/AQUA with 2378 channels covering from 649 to 2674 cm$^{-1}$ at high spectral resolution [22], [26]. The spatial extent of the ground pixels considered in this study is 45 km (east–west) $\times$ 45 km (north–south). Global coverage is achieved twice per day. As detailed by Xiong et al., over 70 AIRS channels near 7.6 $\mu$m are used for CH$_4$ retrieval in version 5, and validation using in situ aircraft observations showed the bias of the retrieved CH$_4$ profiles is $-1.4\%\pm0.1\%$ and its rms difference is about 0.5%–1.6% depending on altitude [22].
The AIRS CH$_4$ Level 2 product extending from 1 July, 2010, to 30 June, 2011, was obtained from the Goddard Earth Sciences (GES) Data and Information Services Center (DISC), available at http://disc.sci.gsfc.nasa.gov/AIRS/data-holdings. In this research, we used the AIRX2RET product. As one type of AIRX2RET’s fields, CH$_4$ VMR$_{eff}$ was used for our analysis, as it relates most directly to the concentration of methane near to the ground surface. A series of seven vertically overlapping trapezoidal functions by dividing the tropospheric CH$_4$ profile: 0.016–32 (1), 32–160 (2), 160–260 (3), 260–359 (4), 359–460 (5), 460–596 (6), and 596–1100 hPa (7) [22]. The seventh trapezoid function of 596–1100 hPa, whose altitude is near the ground surface, was chosen for our analysis. The fields of “sat_lon” and “sat_lat” representing satellite geodetic longitude and latitude, respectively, are also needed for further processing except for CH$_4$ VMR$_{eff}$.

III. METHODS

AIRS data is divided into 240 scenes per day, each of 6-minutes duration. In order to derive the monthly average CH$_4$ concentration, firstly the three types of data mentioned in Section II-B were selected from *.hdf files of 240 scenes, then the selected images were geo-corrected (Section III-A), and finally the 240 scenes were integrated into one whole panoramic image and clipped by China’s administrative boundary (Section III-B). The CH$_4$ concentration of a day was validated by field surface air sample data observed on the same day (Section IV). The monthly average of the methane concentration was calculated and then interpolated by ArcGIS which is a complete system for designing and managing solutions through the application of geographic knowledge (Section III-C).

A. Processes of Image Geometric Correction

Remote sensing images cannot be directly used in practical application, because the geometric shape, size, and direction of image objects are often not identical with the definition of the reference system. There are some geometric distortion problems in raw data, for example, the sizes of image row and column are asymmetrical. Furthermore, pixel spatial resolution does not correspond with practical ground range or terrain surfaces irregularities [27]. Image geometric correction can rectify image distortions caused by system and nonsystem factors [28], [29].

A geographic lookup table (GLT) can be used to place each image pixel in its proper location in a georeferenced output image created by ENVI. This is the premier software solution used by GIS, and image analysts, scientists, and researchers for processing and analyzing geospatial imagery. The GLT file is a file of 2-D images, which contains two bands: rows and columns of the geographical correction images. After the GLT is created, the georeference of each scene can be built using GLT in ENVI.

B. Global Mosaic and Regional Extraction of CH$_4$ Data

After each scene was geo-corrected, the image mosaic method was used to join the 240 scenes of each day into an image in ENVI (Fig. 3). Then the image was clipped by China’s administrative boundary in ENVI (Fig. 4). All of the images were transformed to the WGS84 (World Geodetic System 1984) coordinate system.

From the distribution of CH$_4$ concentration on 1 May, 2011, it is obvious that the CH$_4$ concentration is higher in the Northern than in the Southern Hemisphere.

The concentration of CH$_4$ on 1 May, 2011 was between 1800 and 1900 ppb for most of the area of China. As there are few values in the remote sensing images corresponding to the southwest of China on 1 May, 2011, we obtained only a few values for CH$_4$ in this region after processing.
C. Calculation of Monthly Average CH\textsubscript{4} Concentration

After getting the CH\textsubscript{4} concentration distribution of each day, the monthly average CH\textsubscript{4} concentration was calculated using an IDL program (Fig. 5).

There are no values in some pixels within a small area in the map of monthly average CH\textsubscript{4} concentration. The function of Nibble which can replace cells of a raster corresponding to a mask with the values of the nearest neighbors was used to interpolate in ArcGIS. In order to use the function of Nibble, a mask was built at first by means of the RegionGroup function and SetNull function. For each cell in the output, Regiongroup records the identity of the connected region to which that cell belongs. A unique number is assigned to each region. And the SetNull function returns NoData if a conditional evaluation is true and returns the value specified by another raster if it is false, on a cell-by-cell basis. The process was established by ModelBuilder that is an application by which you create, edit, and manage models [30].

IV. RESULTS AND DISCUSSION

Using the procedures described in Section III, the CH\textsubscript{4} concentration of 365 days of the year from 1 July, 2010, to 30 June, 2011, were calculated, and then the changes of CH\textsubscript{4} concentrations from month to month were considered (Fig. 6).
A. Comparison of Remotely Sensed CH$_4$ Concentration With Ground-Sampled Air Data

To verify the estimations of CH$_4$ concentrations derived from remotely sensed images, we compared the results calculated from AIRS product with the CH$_4$ concentrations we measured for 67 points on the ground (Table I) using the device described in Section II-B (Fig. 2). In order to extract the CH$_4$ concentration from the results calculated from AIRS, a vector file containing sample points was created in ArcGIS, then the function of Sample which creates a table that shows the values of cells from a raster for defined locations was used to extract CH$_4$ concentration.

Ground data for eight points was measured on concrete surfaces near YangLing city and Hanzhong city that lie in Qinba mountain area. The CH$_4$ concentrations for these were between 2063 and 2302 ppb, and they were obviously higher than the other 59 measured on grassland, forest, or farmland (Fig. 7). As these eight points were near cities, these eight values may related to urban and industrial activity. These eight values from over concrete were not included in the validation regression.
Values for \( \text{CH}_4 \) concentrations obtained for Poyang-Dongting Lakes were higher than those obtained for Qinba mountain and the Qinghai-Tibet Plateau. The higher \( \text{CH}_4 \) concentrations for Poyang-Dongting Lakes can be attributed to high levels of the \( \text{CH}_4 \) emissions from wetland [31]. There are six observations from Poyang-Dongting Lakes that are larger than the other 12 observations. These six observations were collected at the edge of the lakes covered by wild Artemisia selengensis (a wild plant growing in wet soil whose tender stem is edible). The higher \( \text{CH}_4 \) concentration is related to this plant. The other samples were obtained in the lakes by boat.

A rough estimate of AIRS \( \text{CH}_4 \) quality was based on the validation error and correlation coefficient (Fig. 7). Standard deviation of the observation-calculated results difference is 43.9 ppb, the correlation coefficient is 0.779 (\( P = 0.001 \)), rms is 93.4, and bias is 82.6. Thus, the quality of AIRS \( \text{CH}_4 \) product is relatively stable, and the relationship showed agreement for most of the results. It can be seen that the \( X_{\text{CH}_4} \) of the ground observation is higher than the \( X_{\text{CH}_4} \) calculated from AIRS (Fig. 7). This is because the ground air samples were collected from the 5 m high layer of the atmosphere above ground level (Section II-A), while the \( X_{\text{CH}_4} \) of AIRS was extracted from the effective \( \text{CH}_4 \) volume mixing ratio profile, and it has value in seven trapezoids. Although we used the seventh trapezoid, which is the closest to the ground surface for our estimations, this trapezoid includes a higher layer of the atmosphere above ground level than the 5-m ground-sampled layer. The second reason for higher value of the ground sample is that the spatial resolution of AIRS images is 45 km, while the distance between some sampling points is less than 45 km. Therefore, the values of these sampling points in the remote sensing images will be the same. It can be seen in Fig. 7 that the ground \( X_{\text{CH}_4} \) observations varied between 1846 and 2152 ppb, while the \( X_{\text{CH}_4} \) calculated from AIRS product had a smaller variation range from 1809 to 1966 ppb.

### Table II

<table>
<thead>
<tr>
<th>Station</th>
<th>Longitude</th>
<th>Latitude</th>
<th>Time (yy/mm/dd)</th>
<th>Number of observations</th>
<th>Types of data</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mt. Waliguan</td>
<td>100.90</td>
<td>36.28</td>
<td>2010/7/1 - 2011/12/31</td>
<td>144</td>
<td>daily</td>
</tr>
<tr>
<td>Shangdianzi</td>
<td>117.12</td>
<td>40.65</td>
<td>2011/7/7 - 2011/12/2</td>
<td>20</td>
<td>weekly</td>
</tr>
<tr>
<td>Lulin</td>
<td>120.87</td>
<td>23.47</td>
<td>2011/7/7 - 2011/12/28</td>
<td>25</td>
<td>weekly</td>
</tr>
</tbody>
</table>

#### B. Comparison of Remotely Sensed \( \text{CH}_4 \) Concentration With Observations From Global Baseline Sites

The World Data Centre for Greenhouse Gases (WDCGG) is one of the WDCs under the Global Atmosphere Watch (GAW) programmer. It serves to gather, archive and provide data on greenhouse gases (e.g., \( \text{CO}_2 \), \( \text{CH}_4 \), CFCs, \( \text{N}_2\text{O} \), and surface ozone) and related gases (e.g., \( \text{CO}, \text{NOx}, \text{SO}_2 \), and VOC) in the atmosphere and ocean, as observed under GAW and other programmers.\(^1\) It aims to provide a critically maintained, long-time-scale record to identify temporal drifts and spatial biases in the calibration of space-based sensors [32]. The WDCGG has 336 stations distributed throughout the world. Mt. Waliguan, Lulin, and Shangdianzi are the three stations located in China. The latest data available for these stations is about December 2010. This was updated in November, 2011. We compared the results we calculated from the AIRS product with the \( \text{CH}_4 \) concentrations that were measured at the three stations between 1 July, 2010, and 29 December, 2010 (Fig. 8). For Lulin and Shangdianzi station data is for weekly intervals, and it is daily at Mt. Waliguan (Table II).

\(^1\)[Online]. Available: http://gaw.kishou.go.jp/wdcgg/
Fig. 9. Differences between consecutive months (Δ) of CH₄ concentrations estimated by AIRS in the lower troposphere over China for the year July 2010 to June 2011. Results are displayed in ppb, ranging in most pixels from −50 to 50 ppb (±3% relative difference).

(Fig. 7), rms, SDV, and bias is smaller for the WDCGG stations (Fig. 8) arising from the higher accuracy of the observations at these stations. As the global baseline sites, these station have appropriate observation environments and better observation equipment. It can be seen from Fig. 8 that the observations from Shangdianzi are larger than those from Mt. Waliguan and Lulin. The reason is that the station of Shangdianzi is near from Beijing which is one of largest cites in China.
C. Spatial Distribution Feature of CH\textsubscript{4} Concentration

Monthly spatial variations of the CH\textsubscript{4} concentration of the lower troposphere over China calculated from AIRS are mapped in Fig. 6 and given here.

1) XCH\textsubscript{4} in southwest China was discontinuous and distinguishable from XCH\textsubscript{4} in the adjacent areas. This is an artifact of the inadequate number of images for this region, and the area is not considered further.

2) XCH\textsubscript{4} was higher in parts of north of China and particularly northeast China. Some previous studies of satellite-retrieved methane [20], [33], [34] see a higher concentration in parts of the south of China. One reason is that our study is based on surface measurements, while they focused on total column. Another reason is that AIRS exhibits a lack of sensitivity near the ground [23]. There are large differences in XCH\textsubscript{4} between some regions for particular months, e.g., over northeast China, XCH\textsubscript{4} is 80 ppb larger than that over the south in October 2010. The decrease of XCH\textsubscript{4} from northwest China to south China was about 100 ppb in January 2011.

One explanation of these regional differences is that there are large areas of forest and wetland in north China. Wetlands have been indicated to be the source of high levels of CH\textsubscript{4} observed in northeast China [23]. Another explanation is that some areas in south China are covered by the subtropical oceanic air mass, which has a much lower CH\textsubscript{4} concentration than north China [35].

3) Apart from these broad regional differences, higher concentrations of CH\textsubscript{4} enhancement are observed in smaller and more specifically defined areas of China. The highest concentrations are found in the Inner Mongolia Autonomous Region and northeast China. These arise from anthropogenic activities. More than 50% of present-day global methane emissions are anthropogenic. The largest contributors are fossil fuel production, ruminants, rice cultivation, and waste handling [36]. There are many ruminant livestock (i.e., sheep, goats, and cattle) that are related to emission of CH\textsubscript{4} in the Inner Mongolia Autonomous Region. While there is intensive and extensive rice cultivation in northeast China and wetland conditions of paddy fields are conducive to CH\textsubscript{4} emission. The main natural source of CH\textsubscript{4} is considered to be from wetlands, although there is some uncertainty about the magnitude of the wetlands source [37]–[39].

D. Temporal Changes of CH\textsubscript{4} Concentrations

The change in XCH\textsubscript{4} was not obvious from September 2010 to December 2010 and from January 2011 to April 2011. The extent of change between consecutive months of CH\textsubscript{4} was remarkable for several areas (Fig. 9). The larger changes of CH\textsubscript{4} between months were for: 1) South China between July and August 2010; 2) Inner Mongolian Plateau and Tarim Basin between September and October 2010; 3) Tarim Basin between October and November 2010; 4) Tarim Basin and central China between November and December 2010; 5) Southwest China and northeast China between January and February 2011; 6) Northwest China between February and March 2011; and 7) south China between March and April 2011.

For each month, the mean monthly CH\textsubscript{4} concentrations for the entire area of China were extracted from the table of layer properties in ArcGIS, and then the twelve monthly values were plotted in Fig. 10. The seasonal cycle of the CH\textsubscript{4} lower troposphere concentrations in China are clearly shown with the lowest concentrations in summer and the highest in winter. The high levels of incoming solar radiation in summer results in vigorous vertical mixing of the lower and upper air masses of the atmosphere. Accordingly, CH\textsubscript{4} concentration of the lower troposphere is usually lowest in August. In contrast, in winter, atmospheric mixing is often stabilized by cooling near the ground surface by radiation of heat into the atmosphere, especially at night and causing frosts [40]. As a result, the CH\textsubscript{4} concentrations became higher in winter, especially in December 2010 and January 2011. This finding of lower CH\textsubscript{4} in summer and lower concentration in lower troposphere is consistent with many in situ observations, e.g., Dlugokenchy et al. [14], [25], [41].

V. Conclusion

Remote sensing from space has been proven to be feasible for detection of CH\textsubscript{4} concentrations in the atmosphere with some definition of concentrations in different layers of the troposphere. The ability of satellites to sense sources globally is unique and opens a new window for the analysis of the biogeochemical cycle of CH\textsubscript{4} and anthropogenic impacts on this [23]. In this paper, monthly methane concentrations were estimated and their spatiotemporal variation over the surface of China (excluding the southeast) for one year was plotted.

The seasonal cycle in the lower troposphere layer shows that CH\textsubscript{4} concentrations were lowest in summer and highest in winter. The average monthly CH\textsubscript{4} concentration in the lower troposphere of China increased from September to October and decreased from March to April.

Compared with values calculated from AIRS product images, the near-surface measurements of CH\textsubscript{4} mixing ratios in the lower troposphere were higher by about 20–130 ppb. This
is explained by different depths of the lower troposphere sampled by the two methods. However, the correlation between the remotely sensed and air analyzed values was significant.

This study shows the techniques with which we can monitor the spatiotemporal distribution of CH₄ in the lower troposphere above China, and validates the results by field measurements. While rapid changes of CH₄ were observed for specific areas of China, information is not immediately available to explain the causes of these changes. Therefore, with AIRS in operation for several years, we should be able to examine the temporal and spatial variations of CH₄ over longer time periods and give more detailed attention to smaller areas. The integration of these measurements with precise ground-based observations located to cover a range of vegetation covers and land uses should greatly reduce uncertainties in the methane source strength, helping to draw a more consistent picture of the global methane budget. This should be a top priority for future climate research [23].

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