

# Method and results of retrieval of the methane content in the atmosphere of Western Siberia from AIRS data

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The paper discusses the method of determination of the methane content in the atmosphere from spectra of AIRS sensor installed aboard AQUA satellite and the method testing with data of 2005–2006. Maps of the horizontal methane distribution and seasonal variations of CH<sub>4</sub> in the atmosphere over wetland of Western Siberia are presented. Sets of clear sky AIRS spectral data selected for the period from spring of 2005 to fall of 2006 and direct/inverse FIRE-ARMS model were used. The methane content in the atmospheric column was determined, and seasonal maps of the horizontal methane distribution in the atmosphere of Western Siberia (58–67°N; 58–90°E) were constructed. Seasonal mean methane variations in the atmosphere of the studied region were revealed.

## Introduction

Key problems of the XXI century are accumulation of greenhouse gases in the atmosphere and global warming. The subarctic zone of the Western Siberia wetland ecosystem is the most sensitive to the increase of the surface air temperature, which initiates the permafrost thawing process.<sup>1</sup> The investigation of the global warming effect on the carbon balance of the arctic and subarctic wetland ecosystems is very important because of the expected large-scale defrosting of peat layers, in which the giant amounts of organic carbon are deposited. Peatbog defrosting may lead to a significant increase in methane (CH<sub>4</sub>) emissions from bogs of the extended subarctic zone of Western Siberia, to methane accumulation in the atmosphere, and, consequently, to the growing greenhouse effect.

The most promising method for global monitoring of accumulation of greenhouse gases in the atmosphere and for revealing the main sources of their emission is the thermal sensing of the Earth from space.<sup>2</sup> Various methods for determination of vertical profiles of atmospheric gases from emission spectra of the atmosphere, recorded from space, are actively developed for a long time.<sup>3–8</sup> The data of new-generation sensors, such as IMG aboard the ADEOS satellite,<sup>9,10</sup> AIRS aboard AQUA,<sup>11</sup> TES aboard AURA,<sup>12</sup> and IASI aboard METOP,<sup>13</sup> having a rather high spectral resolution (0.1–0.5 cm<sup>-1</sup>) in the thermal IR region 600–2000 cm<sup>-1</sup> allow reconstruction of vertical profiles and columnar contents of various greenhouse gases, such as H<sub>2</sub>O, O<sub>3</sub>, CO, CH<sub>4</sub>, CO<sub>2</sub> (Refs. 10, 14–19) and even their isotopic modifications, for example, HDO and the HDO/H<sub>2</sub>O

in the atmosphere.<sup>20</sup> With the aid of SCIAMACHY sensor aboard Envisat, first measurements of the methane distribution in the lower atmosphere from space have been obtained recently.<sup>21,22</sup> However, no data on seasonal methane variations over the permafrost zone of the wetland ecosystem in Western Siberia were obtained.

We have undertaken an attempt to retrieve seasonal methane variations in the atmosphere over the wetland ecosystem of Western Siberia (58–67°N; 58–90°E) including a part of the subarctic permafrost zone from AIRS spectra for 2005–2006.

Among sensors orbiting in 2005–2006, AIRS (NASA) is the most suitable for monitoring of the methane content over large territories. It is represented by a grid spectrometer of rather high resolution (~0.5 cm<sup>-1</sup>) with 2378 spectral channels in the thermal IR region 3.7–15.4 μm [Ref. 11]. It scans the atmosphere in the transversal direction in the angle range ±49.5° relative to nadir. Each scanning line consists of 90 pixels with a size of 13.5 km in nadir and 41 × 21.4 km at the maximal scanning angle from the 705.3-km orbit. The AIRS ensures the complete coverage of the globe every 12 hours, which allows accumulation of sufficient number of spectra obtained under clear-sky conditions for coverage of the entire territory of Western Siberia for a season.

The objectives of this work were to determine the horizontal distribution of the methane content in the atmosphere and to reveal average seasonal methane variations over the vast wetland ecosystem of Western Siberia using AIRS/AQUA spectra obtained under clear-sky conditions.

The direct/inverse FIRE-ARMS model tested earlier with IMG spectra<sup>14,23</sup> was adapted to the

problem of determination of the methane content in the atmospheric column from AIRS spectra. At the first stage, the vertical temperature profile is reconstructed from the spectral range 680–830  $\text{cm}^{-1}$  by the classical method of optimal statistical estimation,<sup>4,5,24</sup> and then the total methane content in atmospheric column is determined from the spectral range 1298–1308  $\text{cm}^{-1}$  using the restricted minimization method. Features of this inverse code and the results of retrieval of the horizontal methane distribution in the atmosphere of Western Siberia from AIRS spectra for 2005–2006 are under discussion.

## Method and results of retrieval of the $\text{CH}_4$ content in the atmosphere from AIRS spectra

The radiative transfer equation for the outgoing Earth's thermal radiation, observed in nadir from space in the cloudless slightly aerosol atmosphere has the form<sup>25</sup>:

$$\begin{aligned} W_v &= \varepsilon_v B_v(T_0) \exp\left(-\int_0^H k_v dh\right) + \\ &+ \int_0^H k_v B_v \exp\left(-\int_h^H k_v dh'\right) dh + \\ &+ (1 - \varepsilon_v) W_v^{\text{down}} \exp\left(-\int_0^H k_v dh\right), \quad (1) \\ W_v^{\text{down}} &= \int_0^H dh k_v B_v \exp\left(-\int_0^h k_v dh'\right), \end{aligned}$$

where  $W_v$  is the spectral brightness of the outgoing thermal radiation;  $W_v^{\text{down}}$  is the downward radiation,  $W/(\text{m}^2 \cdot \text{cm}^{-1} \cdot \text{sr})$ , at the frequency  $\nu$ ;  $B_v(T(h))$  is the Planck blackbody radiation function at the temperature  $T(h)$ ;  $\varepsilon_v$  is the surface emissivity;  $h$  is the height;  $H$  is the height of the atmospheric top;  $k_v(h)$  is the absorption coefficient of gaseous constituents of the atmosphere calculated by the *line-by-line* method using spectroscopic parameters from the HITRAN-2000 database.<sup>26</sup> The water vapor continuum absorption is taken into account within the CKD 2.4 model,<sup>27</sup> and the effect of line mixing in the  $\text{CO}_2$  band is calculated by the model from Ref. 28. For a slant path,  $dh$  in Eq. (1) is replaced with  $\sec\theta dh$ , where  $\theta(h)$  is the observation zenith angle as a function of height taking into account the atmospheric sphericity. The model described is used in the FIRE-ARMS software (<http://remotesensing.ru>) and applied in this paper to calculate the spectral brightness of the outgoing radiation recorded in AIRS spectral channels. The FIRE-ARMS software performs the *line-by-line* calculations of  $W_v$  with a step up to 0.0001  $\text{cm}^{-1}$  and its following convolution with the instrument function of the AIRS sensor. In addition, this

software allows the calculation of tables of absorption cross sections for individual gases, due to which it becomes possible to accelerate significantly the calculations of spectra and derivatives. The data on the emissivity of different types of surface collected on the HITRAN-96 CD-ROM<sup>26</sup> were used as values of the surface emissivity  $\varepsilon_v$  in calculations of  $W_v$  over Western Siberia.

The algorithm for processing of every recorded AIRS spectrum consisted of the following steps:

1) Determination of the vertical temperature profile using the iteration scheme of the optimal statistical estimation method,<sup>24</sup> with the use of nonoverlapping spectral intervals from the range 680–830  $\text{cm}^{-1}$  with a rather good density of temperature weighting functions from the surface to high atmospheric layer, in which the influence of water vapor variations on the atmosphere emission spectrum can be neglected<sup>29</sup>:

$$x_{k+1} = C_k(y - y_k) + (I - C_k K_k)(x_0 - x_k) - x_k, \quad (2)$$

$$C_k = (K_k^T S_e^{-1} K_k + S_a^{-1})^{-1} K_k^T S_a^{-1},$$

where  $x_{k+1}$ ,  $x_k$  is the sought vector (temperature profile) at the  $(k+1)$ st and  $k$ th iteration;  $x_0$  is the initial approximation;  $y$  and  $y_k$  are the recorded and calculated spectra (in selected spectral intervals);  $I$  is the unit matrix;  $K_k$  is the Jacobian of the direct model;  $S_e$  is the covariance matrix of errors of spectral measurements;  $S_a$  is the *a priori* covariance matrix of the sought vector.

The covariance matrices  $S_a$  of the vertical temperature profiles were constructed for different seasons from the data of balloon measurements of meteorological observatories in Western Siberia: Pechora (65.12°N, 57.10°E), Salekhard (66.53°N, 66.67°E), Khanty-Mansiisk (61.02°N, 69.03°E), Turukhansk (65.78°N, 87.93°E), available from the site of the British Atmospheric Data Center (<http://badc.nerc.ac.uk>).

2) For the water vapor vertical profile, the month average profile (corresponding to the month of measurement of the processed spectrum) of radiosonde measurements at the nearest meteorological observatories was used.

3) Determination of the effective vertical profile of methane by the method of minimization of the functional  $F$  [Ref. 14] with restrictions on possible methane variations tested earlier on IMG spectra<sup>23</sup>:

$$F(x) = \frac{1}{2} \sum_i^M [W_i^{\text{cal}}(x)/W_i^{\text{obs}} - W_i^{\text{obs}}/W_i^{\text{cal}}(x)]^2, \quad (3)$$

where  $M$  is the number of spectral channels of the AIRS sensor taken into account in this problem.

We used the range 1298–1308  $\text{cm}^{-1}$ , since water vapor variations of up to 300% about the monthly mean value in this range negligibly change the brightness spectrum. The essence of the method is the fitting of the model and experimental spectra with minimization of the target function of the form (3).

In this process, the following restrictions are imposed on the sought solution:

$$ax_i^{\text{ref}} \leq x_i \leq bx_i^{\text{ref}}, \quad (4)$$

where  $x_i$  and  $x_i^{\text{ref}}$  are the variable sought and the reference atmospheric parameters (concentration of  $\text{CH}_4$ ) at the  $i$ th height, while  $a$  and  $b$  meet the conditions  $0 < a < 1$  and  $b > 1$ . To minimize the target function (3), the Fletcher-Reeves algorithm modified for restrictions (4) was used. We took the values  $a = 0.9$  and  $b = 1.8$  at a model surface methane concentration of 1.7 ppm, and the acceptable variability range of the concentration was 1.5–3.0 ppm, which is in agreement with the airborne measurements in Western Siberia.<sup>30</sup> Significant methane variations were observed in the lower atmosphere between the surface and 7 km. That is why, the so-called effective profile was reconstructed in this method, that is, possible variations of the methane concentration were taken into account only in the following 10 atmospheric layers: 0–100 m, 100–500 m, 0.5–1 km, and then 1-km layers between 1 and 8 km. The methane profile above 8 km was approximated by the standard atmospheric model.

The methane content in the atmospheric column was calculated from the obtained vertical effective profiles of methane and temperature as:

$$\text{total CH}_4 = 10^6 N_A^{-1} \int_0^H \frac{p}{kT} N_{\text{CH}_4} dh,$$

where total  $\text{CH}_4$  is the total methane content in the atmospheric column, mol/m<sup>2</sup>;  $N_A$  is the Avogadro number;  $p$ ,  $T$  are the vertical pressure and temperature profiles;  $N_{\text{CH}_4}$  is the methane concentration, ppm.

The errors of this method were estimated by the scheme of closed model experiments with synthetic AIRS spectra. For the selected set of atmospheres (profiles), the FIRE-ARMS software was used to calculate fragments of AIRS-like IR spectra (with the corresponding type and width of the instrumental function specified by the device characteristics) including spectral intervals required for the method of reconstruction of the temperature and methane profiles. These spectra disturbed by the normally distributed noise with the level, characteristic of the used AIRS spectral channels. Then, the method described was used to reconstruct the effective methane profile and the columnar methane content from model spectra. The results obtained were compared with the initial (known) profiles and the total content; the rms deviation was calculated as well.

As a result, it was found that the relative accuracy of the method proposed for determination of the methane content in the atmospheric column from AIRS spectra is about 2.5%. This result is acceptable for detection of seasonal methane variations in the atmosphere. However, it should be noted that this value should be considered as an error of the solution scheme (algorithm) in the inverse problem of the atmospheric methane determination from AIRS spectra.

That is, it is the lower estimate of the error, neglecting possible uncertainties associated with variations of the surface emissivity for spring, summer, and fall seasons, with influence of aerosol provided its high concentrations are present in the observed atmosphere, and with possible inaccuracies in atmospheric gas absorption line intensities in the HITRAN database.

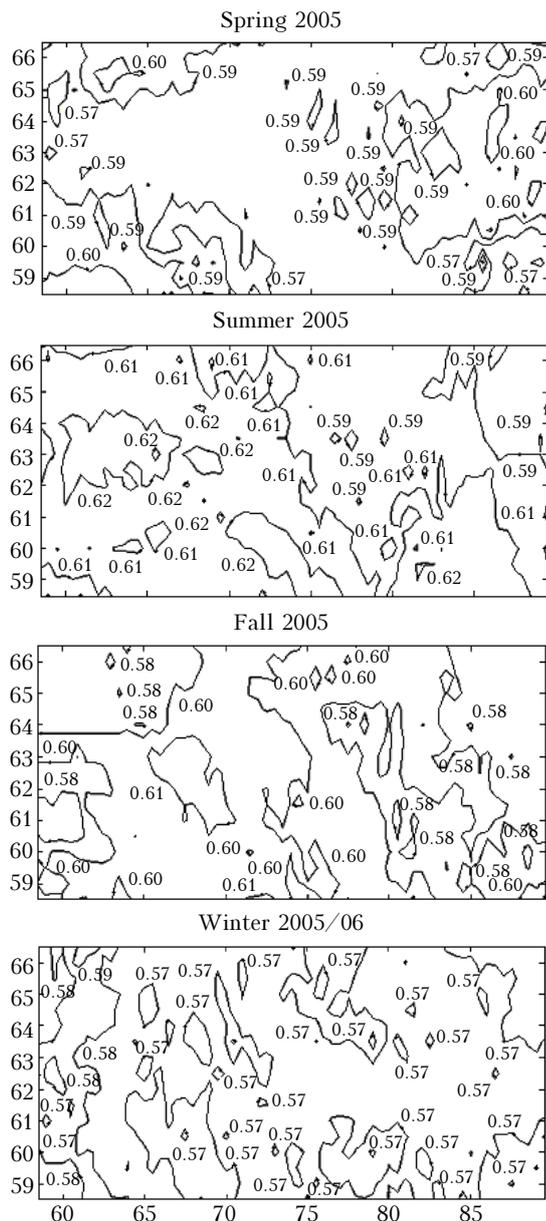
To assess independently the method reliability, we have compared the methane concentration in the surface layer (0–100 m) reconstructed from the AIRS spectrum with the time-synchronized direct (chromatographic) methane measurement in the key area of West-Siberian wetlands. The measurements were conducted within the framework of the INTAS Project CASUS 03-51-6294. The direct chromatographic measurements of methane were carried out occasionally in the region 60.58°N, 70.10°E from June 17, 2004, to September 12, 2004; air was sampled at a height of 2 m from the ground at different places of the key area. The measured methane concentrations mostly varied between 1.48 and 3.89 ppm. The clear-sky situation convenient for the comparative experiment was observed on June 18, 2004. The processing of the AIRS spectrum recorded this day over this area gave a methane concentration of 2.04 ppm for a surface layer of 0–100 m. The area-averaged direct measurements of methane in this area at a height of 2 m yielded 2.58 ppm. This discrepancy is quite acceptable for problems of this kind.

The method proposed was applied to processing of spectra recorded by the AIRS sensor over Western Siberia. The AIRS spectra recorded under the clear-sky conditions over the region 58–67°N, 58–90°E were sampled from spring of 2005 to winter of 2005/06. All spectra were processed by the method to determine and to map the methane content over the region under study. The maps of the monthly average methane content were drawn by averaging the total  $\text{CH}_4$  values at every 0.5° latitude × 0.5° longitude cell reconstructed from the sample of clear-sky AIRS spectra for a season.

The obtained seasonal maps of the horizontal methane distribution in the atmosphere of Western Siberia are shown in Fig. 1.

Relatively high spring values of methane in the atmosphere result from emission of methane, accumulated during winter in bogs and released as a result of snow and soil thawing, while the horizontal distribution is caused by both the dynamics of thawing processes and the characteristic wind rose in the troposphere in this period. Similar values of the methane content in the atmosphere of this region were earlier obtained from the IMG/ADEOS data for April of 1997 [Ref. 15].

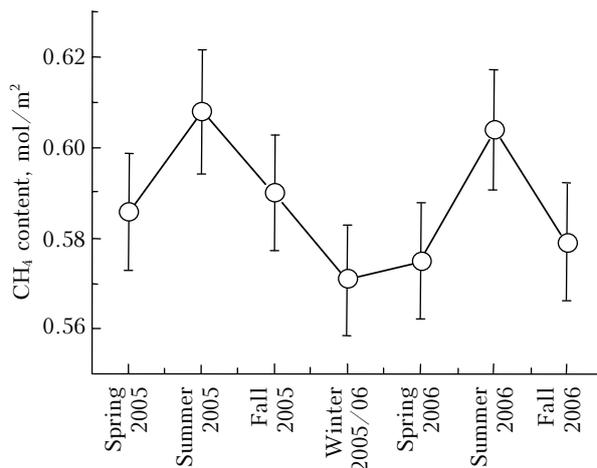
In summer, a more uniform methane distribution is observed in the atmosphere of Western Siberia, because in this period the entire region becomes a source of methane emission from wetlands. In winter (in the absence of natural methane emission from wetlands), the nearly uniform distribution of background atmospheric methane with an insignificant increase in the western part of the region is observed.



**Fig. 1.** Horizontal distribution of the methane content in the atmosphere of Western Siberia in spring, summer, and fall of 2005, as well as in winter of 2005/06 obtained from clear-sky AIRS/AQUA spectral data.

This may be connected with the activity of the oil and gas industry in this region and the presence of significant permanent anthropogenic methane emission sources, such as compressor houses for long-distance pipeline transportation of natural gas. A map of this kind drawn for a successful one-time AIRS image of a rather large cloudless area (about  $1000 \times 1000$  km) over the studied region in winter at a stable previous wind pattern can yield the horizontal methane distribution, reflecting the location of main sources of anthropogenic methane emission.

Figure 2 shows the seasonal dependence of atmospheric methane in Western Siberia averaged over the entire region under study.



**Fig. 2.** Seasonal variations of the methane content in atmospheric column averaged over the entire studied territory of Western Siberia ( $58\text{--}67^\circ\text{N}$ ;  $58\text{--}90^\circ\text{E}$ ).

At a given level of error (shown in the figure), it is possible to determine reliably only the presence of variations in the atmospheric methane content in winter and summer. The average value of the atmospheric methane content for the entire period shown in the figure is about  $0.59 \text{ mol/m}^2$ . The observed difference of about  $0.04 \text{ mol/m}^2$  between the summer 2005 and 2006 and winter (2005/06) methane content can be treated as a pure contribution of the natural methane emission from wetlands for the summer period.

## Conclusions

The increased methane content in the atmosphere of Western Siberia obtained in spring, summer, and fall relative to the winter season is obviously caused by the natural methane emission from wetlands (during the whole summer period and roughly the later half of the spring season and the earlier half of fall) at these latitudes ( $58\text{--}67^\circ\text{N}$ ,  $58\text{--}90^\circ\text{E}$ ). The methane content during the whole period under study is a sum of the background methane content in the atmosphere and an additional contribution due to emission from wetlands. In the winter season, the natural methane emission terminates due to the soil freezing and in this period it is mostly determined by the background component. The addition of anthropogenic methane due to its emission from objects of the oil and gas industry, situated in the western part of this region, is not as high as the natural emission from wetlands in summer and in warm periods of the spring and fall seasons.

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