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Comparisons of satellite (GOSAT) and ground-based spectroscopic measurements of CH<sub>4</sub> content near Saint Petersburg: Influence of data collocation.

Nikolai M. Gavrilov\*, Maria V. Makarova, Yuri M. Timofeev, and

Anatoly V. Poberovsky

Saint-Petersburg State University, Atmospheric Physics Department, Saint-Petersburg, Russia

\*Corresponding author. Email address: gavrilov@pobox.spbu.ru

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We compared atmospheric column average mole fractions of methane measured with ground-based Fourier-transform spectroscopy at the Physical Department of Saint-Petersburg State University (59.9° N, 29.8° E) in years 2009-2012 with similar data obtained from the Japanese GOSAT satellite. For the GOSAT data version V02.xx, average and median values of biases between satellite and ground-based methane mole fractions are -(1.7 - 4.1) ppb and their standard deviations are 10 - 13 ppb. These values are similar to biases between the GOSAT satellite and the ground based Total Carbon Column Observation Network and Network for the Detection of Atmospheric Composition Change making Fourier transform spectroscopic observations. Average and median biases for satellite data selected within  $\alpha = \pm 1^{\circ}$  latitude-longitude vicinity from the ground-based observations site are smaller than those for  $\alpha = \pm 3^{\circ}$  and  $\alpha = \pm 5^{\circ}$ .

# Comparisons of satellite (GOSAT) and ground-based spectroscopic measurements of CH<sub>4</sub> content near Saint Petersburg: Influence of data collocation.

Nikolai M. Gavrilov\*, Maria V. Makarova, Yury M. Timofeev, and

Anatoly V. Poberovsky

Atmospheric Physics Department, Saint-Petersburg State University, Saint-Petersburg, 198504, Russia

(Received ..., accepted ...)

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Key words: CH<sub>4</sub>, atmospheric methane, general content, ground-based measurements, FTIR spectroscopy, the satellite GOSAT, comparison, validation

\*Corresponding author. Email address: gavrilov@pobox.spbu.ru

#### 1. Introduction

Methane is an important anthropogenic greenhouse gas (e.g. Kondratyev and Varotsos 1995). Despite its low concentration in the Earth's atmosphere, methane contribution to the anthropogenic greenhouse effect is ~ 15%. For local methane monitoring, laboratory analyses of air samples are typically used to determine CH<sub>4</sub> mole fractions near the Earth's surface and in the troposphere using aircraft (Conway et al. 2003). Optical spectrometer methods of measurements, based on registration of IR absorption spectra of solar radiation are also applied to identify the total CH<sub>4</sub> contents and average mole fractions  $X_{CH4}$  in the atmospheric column.

Ground-based optical measurements of methane can be useful for validating satellite measurements, which also provide information about total CH<sub>4</sub> contents in the atmospheric columns. To test satellite observations of greenhouse gases, an international ground-based network TCCON (The Total Carbon Column Observing Network) was developed, which uses Fourier transform (FT) spectrometers measuring direct solar infra red (IR) radiation to determine total column contents of CO<sub>2</sub>, CH<sub>4</sub> and other climate-forming gases (Wunch et al. 2011). Similar FTIR measurements are also performed at the international ground-based network NDACC (Network for the Detection of Atmospheric Composition Change, see <a href="http://www.ndsc.ncep.noaa.gov/">http://www.ndsc.ncep.noaa.gov/</a>).

In Saint-Petersburg State University (SPbU), spectroscopic measurements of total methane were started in the year 1991 (Mironenkov et al. 1996). These measurements up to the year 2009 were carried out using a solar thermal spectrometer with resolution of 0.4 - 0.6 cm<sup>-1</sup>. From January 2009 onwards, Atmospheric Physics Department of SPbU conducts FTIR measurements using a Bruker IFS 125 HR interferometer with high spectral resolution up to 0.002 cm<sup>-1</sup>. Similar interferometers are used at the NDACC network sites. These measurements of atmospheric trace components are described, for example, by Poberovsky et al. (2010), Virolainen et al. (2011); Polyakov et al. (2011).

The first global satellite data on the total content of methane in the atmospheric column were obtained using the IMG/ADEOS device, which measured outgoing atmospheric thermal radiation with high spectral resolution (Kobayashi et al. 1999). Further studies were conducted with the SCIAMACHY, AIRS, IASI, TES instruments

(Sussmann et al. 2005; Razavi et al. 2009; Xiong et al. 2010; Wecht et al. 2012). Despite the extensive observational programme, geographical distributions of methane and its sources are not sufficiently clarified (Solomon et al. 2007). Regular global satellite measurements of  $CH_4$  can help us in studying global methane climatology.

In January 2009, the GOSAT (Greenhouse Gas Observation by Satellite) satellite was launched, which is a joint project of the Japan Aerospace Exploration Agency, Ministry of the Environment and the National Institute for Environmental Studies in Tsukuba, Japan (Kuze et al. 2009). The satellite is designed for space-based monitoring of the global distributions of total column  $CO_2$  and  $CH_4$  in the atmosphere. The average column mole fractions of carbon dioxide  $X_{CO2}$  and methane  $X_{CH4}$  are recovered from the data of the TANSO-FTS (Thermal And Near infrared Sensor for carbon Observations-Fourier Transform Spectrometer) instrument for measurements of greenhouse gases on board of GOSAT (Yoshida et al. 2011).

Morino et al. (2011) and Yoshida et al. (2013) have performed comparisons of  $X_{CH4}$  and  $X_{CO2}$  obtained from the GOSAT satellite with measurements at the groundbased FTIR spectroscopy TCCON network (see above). Some algorithms for  $X_{CH4}$  and  $X_{CO2}$  retrieval from the GOSAT and TCCON data gave good agreement between satellite and ground-based measurements (Notholt et al. 2012; Cogan et al. 2012). Because these comparisons were made for latitudes below 55°, it is interesting to compare satellite (GOSAT) and ground-based observations at higher latitudes using different recovery algorithm.

Gavrilov et al. (2013) made comparisons of the GOSAT satellite  $X_{CH4}$ measurements with respective ground-based FTIR observations near Saint-Petersburg. They use the GOSAT data obtained within  $\pm 3^{\circ}$  latitude and longitude vicinity of the site of ground-based observations. In this paper, we continue the comparisons of  $X_{CH4}$ measurements in years 2009-2012 in the vicinity of St. Petersburg at a latitude of about 60° N. We select the GOSAT data for three different latitude and longitude vicinities ( $\pm 5^{\circ}, \pm 3^{\circ}, \pm 1^{\circ}$ ) and study dependencies of the comparison results on the collocation of satellite and ground-based data.

## 2. Measurement and data processing

The FTIR measurements of solar radiation are performed near the Old Peterhof railway station (59.88° N, 29.82° E) at elevation about 20 m above sea level, which is located

about 35 km southwest from the centre of Saint Petersburg. Spectra registrations take place from cloudless sky or sufficiently large breaks in cloud covers. Measurements typically use the optical path difference of 180 cm, which corresponds to the spectral resolution of 0.005 cm<sup>-1</sup>. The time of accumulation and averaging of ten individual recordings (to get a single spectrum) is about 12 minutes.

Determination of total column contents of gas species in the atmosphere from high-resolution solar radiation spectra measured with Bruker IFS125 HR uses version v.3.92 of the standard international SFIT2 software (Pougatchev et al. 1995; Rinsland et al. 1998; Hase et al. 2004), which has been developed for the NDACC network. The SFIT2 algorithm determines total column contents of atmospheric gases using statistical regularization by the Newton iterative method. As a source of information about the fine structure of molecular absorption lines we used the HITRAN 2000 (with additions of 2001) database of spectroscopic line parameters (Rothman et al. 2003).

The main input parameters for SFIT2 are as follows: measured spectrums of solar radiation, optical path difference, the device aperture, solar zenith angle, signal to noise ratio, meteorological data (temperature and pressure profiles during the day of measurements), a priori information on the profiles of gas mole fractions. Weather information (temperature, pressure), required for spectral processing, comes from the upper air sounding station MGO-Voejkovo (e.g., Weather Web 2013), which is located about 50 miles from Peterhof. A priori profiles of gas concentrations in the atmosphere have been calculated using the Whole Atmosphere Community Climate Model WACCM (Garcia et al. 2007) for the latitude, longitude and altitude of the measurement station in Peterhof.

For retrieval of total column CH<sub>4</sub> in the atmosphere we use the three spectral intervals (2613.7-2615.4, 2835.5-2835.8 and 2921.0–2921.6 cm<sup>-1</sup>) recommended by Sussmann et al. (2011). Average values of the signal-to-noise ratio in the spectral bands are ~ 800.

Random relative errors of single  $X_{CH4}$  measurements do not exceed 0.5% (estimated by the error matrix calculated within the statistical regularization method implemented in the SFIT2 program). In stable and steady state conditions, variations of  $X_{CH4}$  from atmospheric spectra measured in the course of a day, usually do not exceed 1%. Gavrilov et al. (2013) described other details of the measurement technique.

#### 3. The results of comparisons

To compare  $X_{CH4}$  measured near St. Petersburg at the Earth's surface with values obtained with the GOSAT satellite, we choose intervals of simultaneous measurements in years 2009-2012. For these time intervals, we selected GOSAT version V02.xx values of  $X_{CH4}$  from the database of the National Institute for Environmental Studies in Tsukuba, Japan (NIES 2013), which were measured in the  $\pm \alpha$  latitude-longitude vicinity of the site of ground-based observations. In the present study, we use different widths of these vicinities:  $\alpha = 5^{\circ}$ ,  $\alpha = 3^{\circ}$  and  $\alpha = 1^{\circ}$ . Ground-based  $X_{CH4}$  values used for the comparisons were measured at lowest solar zenith angles (usually within  $\pm 3$  hrs from the local noon). In addition, we used only  $X_{CH4}$  values that fall in the 95% confidence intervals about the mean values for the corresponding periods of observations. Since the satellite  $X_{CH4}$  values are obtained for dry atmosphere (excluding water vapour), ground-based  $X_{CH4}$  were also calculated for the dry atmosphere, using the data of reanalysis of meteorological information from the ECMWF (Dee et al., 2011).

For comparisons, we selected pairs of ground-based  $X_{CH4\_SPB}$  and satellite  $X_{CH4\_GOS}$  values, for which differences in dates of their measurements do not exceed two days. When we had several ground-based measurements during a day, we used the daily mean value for  $X_{CH4\_SPB}$ . For  $X_{CH4\_GOS}$  we used individual satellite measurements selected for different latitude and longitude vicinities  $\alpha = 5^\circ$ ,  $\alpha = 3^\circ$  and  $\alpha = 1^\circ$ .

Figure 1 presents  $X_{CH4}$  values for ground-based and satellite measurements selected for different latitude-longitude vicinities. Figure 2 shows respective pairs of  $X_{CH4\_SPB}$  and  $X_{CH4\_GOS}$ . The solid line in Figure 2 corresponds to  $X_{CH4\_SPB} = X_{CH4\_GOS}$ . Table 1 shows the average and median values, as well as standard deviations for ground-based and satellite data presented in Figure 2. The dashed line in Figure 2 is shifted by – 3 ppb relative to the solid line, which correspond to the average values of deviations  $\delta X_{CH4} = X_{CH4\_GOS} - X_{CH4\_SPB}$  presented in Table 1 for GOSAT data selected for latitude-longitude vicinities with different widths  $\alpha$ .

Table 1 shows that for the GOSAT version V02.xx data the average and median values of  $\partial X_{CH4}$  for different latitude-longitude vicinities are -(1.6 - 2.8) ppb, or -(0.1 - 0.2)% and their standard deviations are (13 - 15) ppb, or less than 1%. Absolute values of  $\partial X_{CH4}$  and their standard deviations tend to be smaller at  $\alpha = 1^{\circ}$  compared to that at larger  $\alpha$ . Standard deviations of ground-based  $X_{CH4}_{SPB}$  values are ~ (10.2 - 12.1) ppb in

Table 1, which are larger than standard deviations of satellite  $X_{CH4\_GOS}$  values (8.4 – 11.6) ppb in Table 1.

Figure 3 shows histograms of differences  $\delta X_{CH4}$  between pairs of ground-based and satellite measurements for different latitude-longitude vicinities presented in Figure 2. One can see that histograms in Figures 3(*b*) and 3(*c*) for  $\alpha = 3^{\circ}$  and  $\alpha = 5^{\circ}$  are shifted to negative  $\delta X_{CH4}$ , while the histogram in Figure 3(*a*) for  $\alpha = 1^{\circ}$  is more symmetric. This corresponds to smaller absolute values of average and median  $\delta X_{CH4}$  in Table 1 for  $\alpha =$  $1^{\circ}$  compared to wider latitude-longitude vicinities with larger  $\alpha$ .

#### 4. Discussion

Gavrilov et al. (2013) made comparisons of  $X_{CH4}$  measurements near Saint Petersburg with the GOSAT satellite data. For latitude-longitude vicinity  $\alpha = 3^{\circ}$  they found average and median values of  $\partial X_{CH4}$  to be between -2.4 and 3 ppb depending on the selection of the GOSAT data for comparison. This shows that the results of comparisons of satellite and ground-based  $X_{CH4}$  may depend on the used methods of data selection and their statistical processing. Comparisons of average and median values of  $\partial X_{CH4}$  in Table 1 show their dependencies on the width of latitude-longitude vicinities  $\alpha$  for data selection for the comparisons; the difference may reach 0.1 %.

Yoshida et al. (2013) made comparisons of the GOSAT version V02.xx data with observations at TCCON ground-based network and found average biases of -5.9 ppb (-0.3%) and standard deviations of 12.6 ppb (0.7%). Our analysis (see Table 1 and Figure 3) shows average and median deviations between satellite and ground-based values in the range -(0.1 - 0.2)%, which is about 0.1% smaller than estimates by Yoshida et al. (2013). These differences may be partly caused by substantial statistical errors (because of the limited number of measurements obtained at Saint-Petersburg). Gavrilov et al. (2013) also obtained smaller absolute values of average deviations between GOSAT and Saint-Petersburg data.

The magnitudes of average and median values of differences  $\delta X_{CH4}$  in Table 1 as well as comparisons by <u>Gavrilov et al. (2013)</u> show that Saint-Petersburg FTIR observations using the retrieval algorithms from the NDACC network could be in reasonable agreement with GOSAT satellite data. Standard deviation of  $\delta X_{CH4}$  values in Table 1 is 13 - 15 ppb (about 0.7 - 0.9 %), which is compatible with compound errors of both types of measurements and corresponds to the value 12.6 ppb obtained by Yoshida et al. (2013). This may be indirect evidence that ground-based measurements in St. Petersburg are consistent with measurements of the methane mole fractions at the TCCON network.

Values of  $X_{CH4}$  obtained with GOSAT using several retrieval algorithms were compared with ground-based FTIR observations in various publications. Parker *et al* (2011) compared the  $X_{CH4}$  values measured by the GOSAT satellite with the groundbased TCCON network and with the results of numerical simulation. They found the relative differences of individual satellite and ground-based measurements to be within 0.1 - 0.9% depending on the latitude. Notholt et al. (2012) having analyzed various methane recovery algorithms obtained the standard deviations of differences between GOSAT and land values in the range from 0.8 - 4%. Schepers et al. (2012), when studying the effect of light scattering and Cirrus clouds on the FTIR spectroscopy, got average differences between GOSAT and ground-based  $X_{CH4}$  between -0.3 and -0.4%. Butz et al. (2011) also shows the presence of systematic errors ~ -0.3%. Results of Table 1 with average and median  $\partial X_{CH4}$  values of -(0.1 - 0.2) % correspond to these estimates. This may indicate that FTIRS observations near St. Petersburg are reasonably well aligned with the GOSAT satellite data.

Note that the standard deviations of ground-based  $X_{CH4}$  values are quite large ~ 10 - 12 ppb (see Table 1). One should take into account that our measurements are taken near the Saint-Peterburg metropolis, so the variability of total methane there could be higher than that for background measurements. Makarova et al. (2006) estimated that anthropogenic emissions of St. Petersburg could give contribution of up to 2% of the total CH<sub>4</sub> contents in the atmospheric column.

Comparisons of ground-based and satellite methane measurements conducted in this paper do not take into account some characteristics that may influence the results of measurement and data processing. For example, differences in the averaging kernels applied to remote sensing techniques (Parker et al. 2011), or uncertainty in the parameters of the fine structure of spectral lines (Chesnokova et al. 2011). In addition, relatively small amount of sunny days for FTIR measurements near St. Petersburg requires further data accumulation for more reliable comparisons of satellite and ground-based measurements of atmospheric methane.

### 5. Conclusion

This study analyzed data from satellite and ground-based FTIR instruments. We compared the average column mole fractions of atmospheric methane measured using FTIR spectrometer on board the Japanese satellite GOSAT (data version V02.xx) and at the Earth's surface at the Atmospheric Physics Department of Saint-Petersburg State University (59.9° N, 29.8° E) in years 2009-2012. Average and median relative differences are -(0.1 - 0.2)% and indicate that FTIR observations near St. Petersburg can provide an acceptable agreement with the GOSAT satellite data. Standard deviations of these differences 13 - 15 ppb (0.7 - 0.9%) match compound errors of ground-based and satellite measurements. Absolute average and median values of differences  $\delta X_{CH4}$  for satellite data selected within the  $\alpha = 1^{\circ}$  latitude-longitude vicinity of the ground-based observation site are smaller than those for  $\alpha = 3^{\circ}$  and  $\alpha = 5^{\circ}$ . More reliable comparisons of ground-based and satellite measurements require further accumulation of FTIR data.

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Vicinity	$\alpha = 5^{\circ}$			$\alpha = 3^{\circ}$			$\alpha = 1^{\circ}$		
Parameter	X <sub>CH4_SPB</sub>	$X_{CH4\_GOS}$	$\delta X_{CH4}$	X <sub>CH4_SPB</sub>	$X_{CH4\_GOS}$	$\delta X_{CH4}$	X <sub>CH4_SPB</sub>	$X_{CH4\_GOS}$	$\delta X_{CH4}$
Average	1782.4	1780.0	-2.4	1783.2	1781.3	-1.9	1783.6	1781.9	-1.7
Median	1783.0	1779.2	-2.8	1783.6	1779.2	-2.4	1785.2	1784.6	-1.6
St. Dev.	12.1	11.6	15.4	11.1	9.8	14.5	10.2	8.4	13.0
Ν	506	506	506	256	256	256	60	60	60

**Table 1.** Average characteristics (in ppb) for the data presented in Figure 2.

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**Figure 1.** Average atmospheric column mole fractions of methane measured from the Earth's surface near St. Petersburg and from the GOSAT satellite within  $\alpha = 5^{\circ}$ ,  $\alpha = 3^{\circ}$  and  $\alpha = 1^{\circ}$  latitude-longitude vicinities.



**Figure 2.** Comparisons of pairs of  $X_{CH4}$  values measured from the ground near St. Petersburg and from the GOSAT satellite within latitude-longitude vicinities  $\alpha = 5^{\circ} - 1$ ,  $\alpha = 3^{\circ} - 2$  and  $\alpha = 1^{\circ} - 3$ , for which differences between dates of measurement did not exceed two days. The line 4 corresponds to  $X_{CH4\_SPB} = X_{CH4\_GOS}$ , and the line 5 is shifted from the line 4 by -3 ppb in accordance with average values of  $\delta X_{CH4}$  in Table 1.



**Figure 3.** Histograms of differences  $\delta X_{CH4} = X_{CO2\_GOS} \cdot X_{CO2\_SPB}$  between pairs of measurements presented in Figure 2 for different latitude-longitude vicinities  $\alpha = 1^{\circ}(a)$ ,  $\alpha = 3^{\circ}(b)$  and  $\alpha = 5^{\circ}(c)$ .