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Analysis of Methane Total Column Variations in the Atmosphere near St. Petersburg using Ground-Based Measurements and Simulations

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Abstract—We present a joint analysis of data obtained by Fourier transform infrared measurements of CH_4 and EMAC model calculations for Petergof station (St. Petersburg State University) in 2009–2012. The systematic differences between observed and calculated data are 1.3% and 0.3% for the values of total column and atmospheric column-averaged mole fraction of methane, respectively. The high correlation for experimental and model data of the total column ($r = 0.8$) indicates that EMAC reproduces the total variability of the methane total column in the atmosphere due to meteorological processes. Using model data, we have analyzed the effect of meteorological conditions typical for Fourier transform IR observations on the resulting estimates of the mean values of total column and column-averaged mole fraction of CH4. We have shown that there can be systematic shifts (up to ~0.4%) in the experimental estimates for the mean value relative to the "true" value. This fact should be taken into account in comparing the climatological or model data with the results of Fourier transform IR measurements, especially for stations with a relatively small number of observation days.

Keywords: atmospheric methane, atmospheric Fourier transform IR spectrometry, photochemical transport models

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INTRODUCTION

Methane is a key greenhouse gas in the Earth's atmosphere. This is conditioned by the specific features of its molecular structure and spectral ranges of methane absorption bands. Because methane is a long-lived gas and its temporal changes in the atmosphere are relatively small, the observation systems (both ground-based and satellite) must have a high measurement accuracy [1–3].

At present, basic data on atmospheric methane are provided by the global ground-based networks WMO Global Atmosphere Watch (GAW/NOAA) [11, 12], Network for the Detection of Atmospheric Composition Change (NDACC) [13], and Total Carbon Column Observing Network (TCCON) [14], as well as by the SCIAMACHY [15], AIRS [16], IASI [17], TES [18], and GOSAT satellite measurement programs [19].

Key activities on methane research in Russia were the ground-based measurements of the methane total column from spectra of solar radiation with low spectral resolution conducted over the past 20–40 years in the Institute of Atmospheric Physics of the Russian Academy of Sciences (IAP RAS) [20], Institute of Experimental Meteorology [21], and St. Petersburg State University [22].

Since 2009, the atmospheric monitoring station at St. Petersburg State University [23] has conducted comprehensive measurements of key climatically and chemically active trace gases of the atmosphere with the help of a Bruker IFS 125 HR high-resolution Fourier spectrometer [24–26]. The same year (2009) was characterized by the launching of the atmospheric Fourier station located in Kourovka Astronomical Observatory [27] (Urals Federal University), which is part of the TCCON international measurement network.

One should also mention the activities of the Institute of Atmospheric Optics of the Siberian Branch of the Russian Academy of Sciences, where the network of Siberian stations conducts measurements of methane concentrations in towers [28]. The Voeikov Main Geophysical Observatory (MGO) conducts regular measurements of surface concentrations of CH_4 at several stations of the network of the Federal Service for Hydrometeorology and Environmental Monitoring of Russia (Roshydromet) [29]. Moscow State University offers long-term series of measurements of sur-

face methane concentrations [30]. IAP RAS runs the TROICA project [31], which is a mobile observatory travelling along Russian railways and providing latitude and longitudinal sections of different atmospheric parameters (including surface concentrations of methane).

This paper analyzes the temporal changes in atmospheric methane at Petergof station for 2009–2013 according to measurement data and EMAC model results [32].

METHODS OF MEASUREMENT AND MODELING

Fourier Transform Infrared Measurements of Methane Total Column. Fourier transform infrared measurements have been conducted in Petergof (59.88° N, 29.82° E; 20 m above sea level), some 35 km southwest of central St. Petersburg. The interferograms are recorded under cloudless sky conditions or when there are cloudiness breaks sufficient for the recording of spectra. Normally, the measurements are conducted at an optical path difference of 180 cm, which corresponds to a spectral resolution of 0.005 cm^{-1}. The stability of the Fourier spectrometer and its characteristics are controlled by HBr cell measurements and LINEFIT analysis [33].

High-resolution spectra of direct solar radiation are interpreted (the total column of atmospheric gases are retrieved) using SFIT2 v. 3.92 [34], software developed for NDACC. The meteorological data required for processing spectra included radiosonde data of MGO at Voeikovo [35]. A priori profiles of gas concentrations in the atmosphere were taken to be the profiles calculated by the Whole Atmosphere Community Climate Model (WACCM) [36] for the measuring station at Petergof.

For retrievals of CH_4 total column in the atmosphere, we used the strategy described in [2]. In this case, three spectral microwindows were used: 2613.7–2615.4, 2835.5–2835.8, and 2921.0–2921.6 cm^{-1}. The random relative errors of a single measurement of methane total column did not exceed 0.5%. More details on the technique determining the total content of CH_4 can be found in [2, 37].

Modeling Tools. The total column and column-averaged mole fraction of CH_4 for 2009–2012 were calculated by the ECHAM/MESSy Atmospheric Chemistry (EMAC) model developed in the Max Planck Institute for Chemistry [32]. EMAC combines the ECHAM5 atmospheric general circulation model [38] with different submodels integrated by the Modular Earth Submodel System (MESSy) interface [39]. EMAC simulates the chemical and dynamical processes from the troposphere to the mesosphere (39 vertical layers) with a horizontal resolution of 2.8° × 2.8°. The EMAC-generated data on the total column and column-averaged mole fractions of methane are time series with an 11-h step.

RESULTS AND DISCUSSION

The results of ground-based Fourier transform infrared (FTIR) measurements of methane in the total atmospheric column (2009–2013) in comparison with EMAC-calculated data (2009–2012) are shown in Figs. 1 and 2. Figure 1 shows the daily means of the total column (TC) of methane and Fig. 2 shows the column-averaged mole fraction (MF) of methane.

The value of MF was obtained by dividing the TC by the total number of molecules in a dry atmosphere. The transition from the total column to the column-averaged mole fraction makes it possible to eliminate the influence of pressure variations on methane variations. The calculations of MF were based on meteorological sounding data obtained at Voeikovo station (for measurements) and on data obtained from EMAC meteorological module (for EMAC). It should be noted that currently this parameter (MF) is widely used in simulations, as well as in space and ground-based systems of remote sensing (such as the SCIAMACHY and GOSAT instruments and the TCCON network) [14, 15, 19].

The simulation results for 2009–2012 were agreed by date and time with FTIR measurements through the interpolation of EMAC data. A total of 233 daily average values of TC and MF of methane are available to compare with the model for 2009–2012. The bottoms of Figs. 1 and 2 show the difference between FTIR measurements and model data. The dotted line indicates the average value of this difference.

Hereafter, starting with Figs. 1 and 2, the following notations are used for the data arrays:

FTIR: data obtained using ground-based Fourier transform infrared measurements;

EMAC: full time series generated by the EMAC model;

EMAC/FTIR: a sample of simulation results agreed by date and time with FTIR measurements.

The experimental data given in Figs. 1 and 2 are considered by us as daily average TC and MF of methane, because each value is the result of averaging several methane measurements obtained during the day. Usually, the exception is provided by winter measurements when the reduced daylight hours and meteorological conditions prevent one from conducting series of observations. The diurnal variability of TC and MF of methane as estimated from our measurements is ~0.3% (standard deviation) on the average.

It should be noted that an earlier study [37] compared the results of satellite remote sensing (GOSAT) with our data on the MF of methane. The systematic difference between the two measuring systems was shown to be ~3 ppb (0.2%).

For FTIR, EMAC/FTIR, and EMAC data on the total column of methane, Table 1 presents the main statistical characteristics: mean value and median (with errors), standard deviation (σ), extreme values,

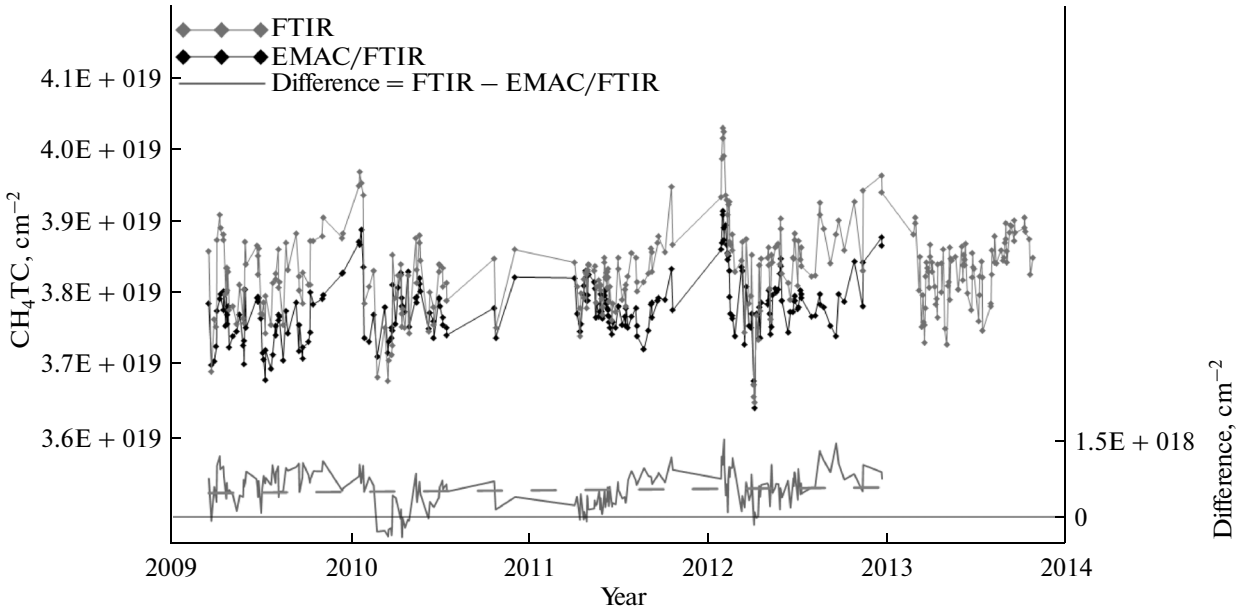

Fig. 1. Measured and calculated daily means of methane total column (TC) and the difference between them.

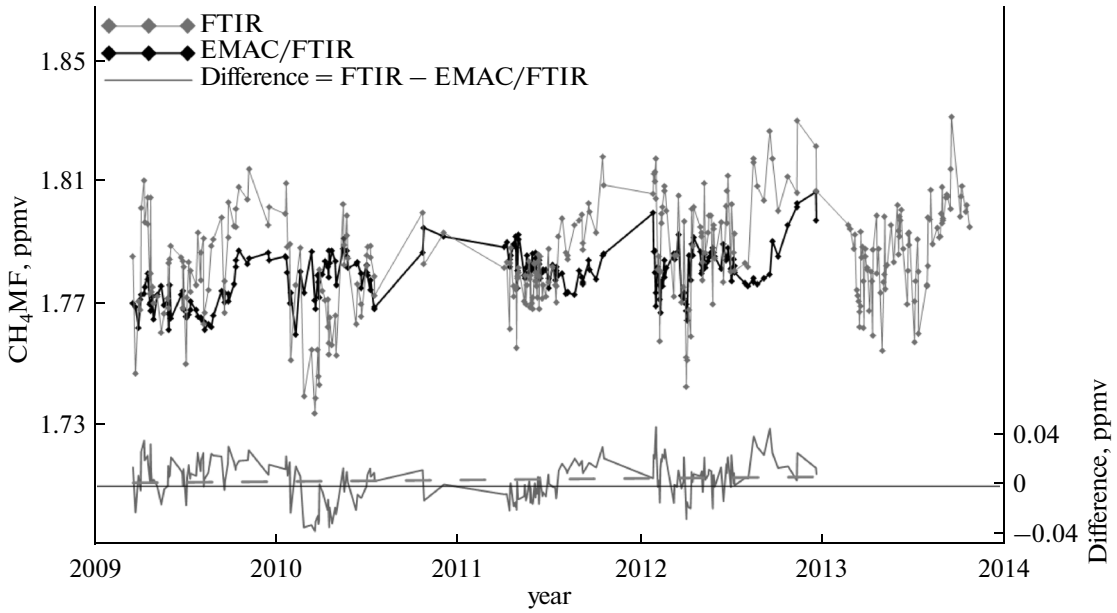

Fig. 2. Measured and calculated daily means of column-averaged CH_4 mole fraction (MF) and the difference between them.

and coefficients of skewness μa and kurtosis μe. Since the model data are limited to late 2012, all the results given in Table 1 also refer to the period from 2009 to 2012 (the measurement data obtained in 2013 were ignored). Table 2 is similar to Table 1 in its structure and meaning and shows the same statistical parameters, but for MF of CH_4.

As a vivid illustration of the statistical data shown in Tables 1 and 2, Figs. 3 and 4 show the histograms of frequency distribution of TC (Fig. 3) and MF (Fig. 4) for all series analyzed in the abovementioned tables (except for the difference between FTIR-EMAC/FTIR measurements and the model).

CH_4 Total Column. We consider the statistical parameters of the EMAC/FTIR series of TC in comparison with FTIR TC (see Table 1; Figs. 1, 3). It can be seen that the measurement data are systematically higher (by ~1.3%) than the simulation results. Indeed,

Table 1. Main statistical parameters for 2009–2012 methane total column data (FTIR, EMAC/FTIR, and EMAC)

TC	Mean, $\times 10^{19}$ cm^{-2}	Median, $\times 10^{19}$ cm^{-2}	σ, $\times 10^{19}$ cm^{-2}	Minimum, $\times 10^{19}$ cm^{-2}	Maximum, $\times 10^{19}$ cm^{-2}	μ*a*	μ*e*
FTIR	3.827 ± 0.004	3.827 ± 0.004	0.06 (1.6%)	3.644 (02.04.2012)	4.027 (29.01.2012)	0.2	1.2
EMAC (all days)	3.761 ± 0.001	3.757 ± 0.001	0.05 (1.3%)	3.627 (05.01.2012)	3.912 (28.01.2012)	0.3	0.2
EMAC/FTIR (measurement days)	3.777 ± 0.003	3.777 ± 0.003	0.04 (1.2%)	3.636 (02.04.2012)	3.912 (28.01.2012)	0.3	0.9
Difference (FTIR-EMAC/FTIR)	0.05 ± 0.002 (1.3%)	0.05 ± 0.002 (1.3%)	0.04 (1%)	–0.04 (–1.0%)	0.15 (3.9%)	–0.1	–0.2

Table 2. Main statistical parameters for 2009–2012 column-averaged CH_4 mole fraction data (FTIR, EMAC/FTIR, and EMAC)

MF	Mean, ppmv	Median, ppmv	σ, ppmv	Minimum, ppmv	Maximum, ppmv	μ*a*	μ*e*
FTIR	1.7842 ± 0.001	1.7837 ± 0.001	0.017 (1.0%)	1.7340 (16.03.2010)	1.8301 (10.11.2012)	–0.2	0.0
EMAC (all days)	1.7826 ± 0.0003	1.7820 ± 0.0003	0.010 (0.6%)	1.7537 (29.01.2009)	1.8133 (19.12.2010)	0.2	–0.3
EMAC/FTIR (measurement days)	1.7792 ± 0.0005	1.7799 ± 0.0005	0.008 (0.5%)	1.7599 (02.08.2010)	1.8067 (17.12.2012)	0.1	0.2
Difference (FTIR-EMAC/FTIR)	0.005 ± 0.001 (0.3%)	0.006 ± 0.001 (0.3%)	0.016 (0.9%)	–0.037 (–2%)	0.048 (3%)	–0.2	–0.2

the difference between experimental and model data is statistically significant and positive for all seasons except for spring (Table 1; Fig. 1, bottom).

The variability of experimental data on TC is higher by ~0.4% than the variability of model data (see the values σ in Table 1). The values of μ*a* and μ*e*, as well as the ratio of the median and mean values for EMAC/FTIR and FTIR values of TC, indicate that the distribution functions for both data arrays are similar in shape and symmetrical (see Fig. 3).

The consistency between temporal variations in TC in both ensembles is clearly seen in Fig. 1. The correlation coefficient r calculated for EMAC/FTIR TC and FTIR TC for 2009–2012 is statistically significant (with a probability of 95%): $r = 0.82 \pm 0.08$ (the corresponding scatter diagram is shown in Fig. 5). The model reproduces the observed periods of TC maxima and minima: the dates of minimum values for EMAC/FTIR TC and FTIR TC series coincide, and the maxims fall on neighboring days (see Table 1).

Column-Averaged CH_4 Mole Fraction. Table 2 shows that, for the data of column-averaged mole fractions FTIR MF, EMAC/FTIR MF, and EMAC MF (Table 2; Figs. 2, 4), the coefficients μ*a* and μ*e* are small and thus the distributions shown in Fig. 4 can be taken as symmetrical and close to the normal distribution. It should be noted that the FTIR MF measurement series is characterized by bimodality: 1.780 and 1.795 ppmv.

Like for the total column, the MF measurement data are an average of 0.3% (a statistically significant value) higher than the simulation results, with the only exception occurred in the first half of the year (see Fig. 2).

Unlike the total column, the temporal variations in FTIR MF and EMAC/FTIR MF are often different (Fig. 2). The minima of FTIR MF and EMAC/FTIR MF fall on different seasons: March and August, respectively (Table 2). This is confirmed by a lower (than TC) correlation coefficient of $r = 0.4 \pm 0.1$ (which is statistically significant at a confidence level of 95%). The scatter diagram for the EMAC/FTIR MF and FTIR MF series is shown in Fig. 6.

In [40], the simulation results (obtained by the ACTM photochemical transport model [41]) for MF of CO_2, N_2O, and CH_4 were compared with measurement data obtained at TCCON network stations. It should be noted that the correlations between the series of observations and model data for CO_2 and N_2O are sufficiently high (r varies in the range from 0.69 to 0.97), while no high values of r were obtained for MF of CH_4: for stations in the 45°–68°N latitudi-

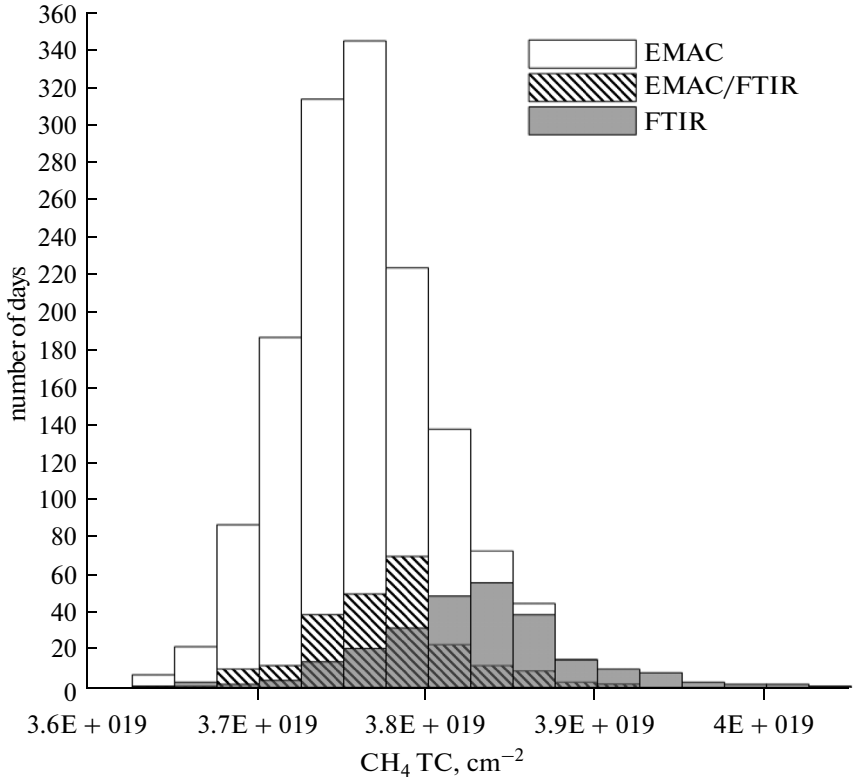

Fig. 3. Histograms of frequency distribution for experimental and model data on CH_4 total column.

nal belt, the correlation coefficient oscillates from 0.24 to 0.57 and is 0.41 (averaged over seven stations). Although [40] used another model, the results published are in good agreement with our data. The main reason for a relatively low reproducibility of observational data on MF of CH_4 by models is supposed to be the lack of data on methane sources (since different models often use the same databases) and the behavior of the main sink of methane—hydroxyl radical OH [40].

For Petergof station, we found that the transition from TC to MF units decreased the values of the following parameters (Tables 1, 2):

(i) the mean value of the difference between FTIR and EMAC/FTIR by 1%. This is caused by the fact that the surface pressure in EMAC is (on the average) 1% lower than the observed values (probably due to the fact that the model pixel size is large: 2.8° × 2.8°);

(ii) variability (σ) of experimental and model values by ~0.6–0.7% (since the influence of variations in atmospheric pressure during the transition from TC to MF is eliminated).

Using the value of $(1 - r^2)$, where r^2 is the coefficient of determination, we can estimate the relative contribution of variations in pressure (density) to the total variance of TC series:

(i) 36% ($(1 - r^2) = 0.36$) for FTIR MF and FTIR TC series;

(ii) 74% for EMAC/FTIR MF and EMAC/FTIR TC;

(iii) 80% for EMAC MF and EMAC TC.

Thus, the variance of TC determined by atmospheric pressure variations is approximately twice as high for the model series than for experimental data.

The estimate obtained by us for FTIR TC is close to the results of [8], where it was shown that almost 40% of variations in methane TC can be explained by variations in atmospheric pressure; then, the contributions decrease in the following order: the annual variation (the seasonality of CH_4 sources and sinks) contributes up to ~18%, the stratospheric anomalies contributes ~2–10%, and the arrival of polluted air masses contributes less than 5%.

Evaluation of the Impact of Meteorological Conditions during FTIR Measurements on Average Values of Methane TC and MF. The analysis of the EMAC model results makes it possible to assess the efficiency of FTIR measurements conducted only under cloudless sky conditions or sufficient cloudiness breaks in reflecting the actual mean values of methane TC and MF in the atmosphere.

For the values of TC and MF, we compare the complete EMAC model data with a sample data array of EMAC/FTIR. In this case, the complete array of EMAC is assumed to be "standard" or "true," and the

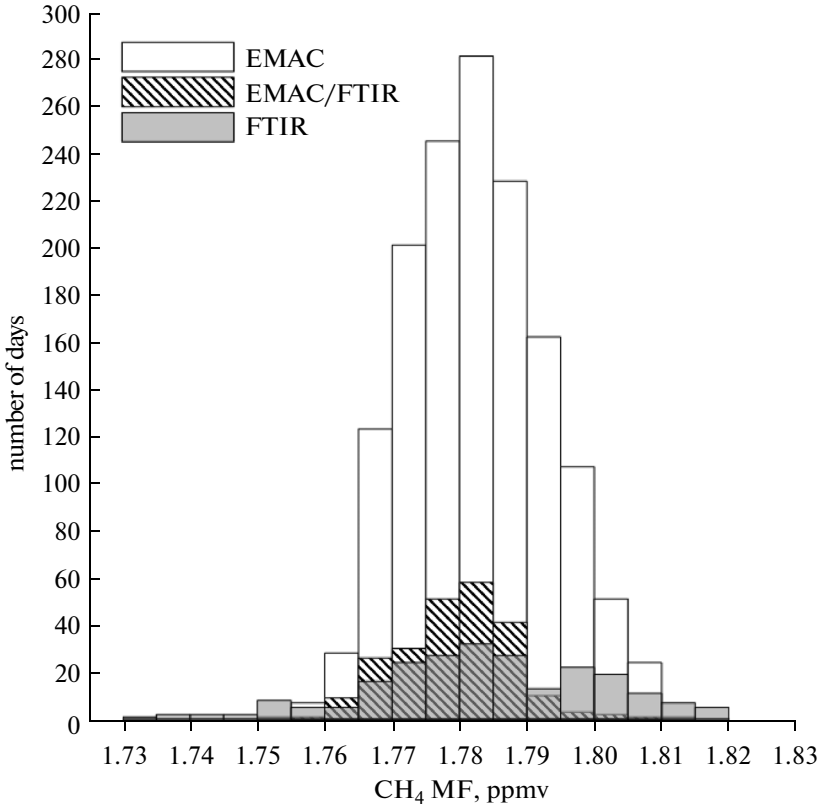

Fig. 4. Histograms of frequency distribution for experimental and model data on column-averaged CH_4 mole fraction.

sample array is assumed to be an analog of FTIR measurements.

The results of this comparison (for 2009–2012) indicated the following:

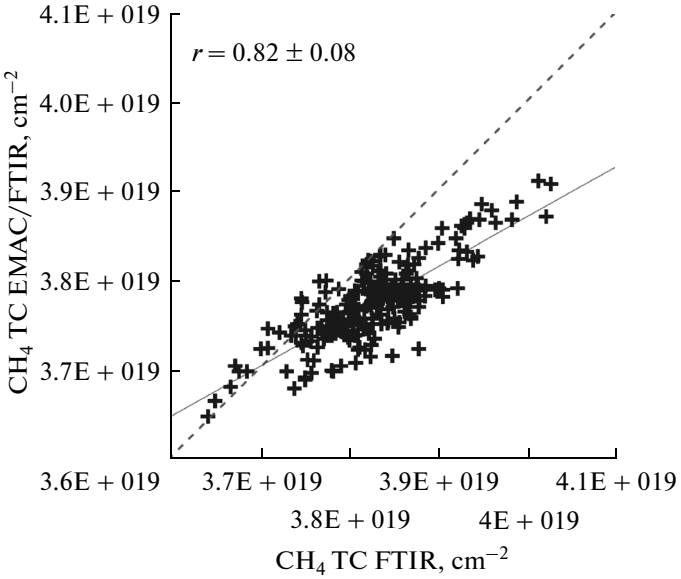

Fig. 5. Diagram of scattering for EMAC/FTIR TC and FTIR TC data.

(i) the average value of TC CH4 estimated from the sample series of EMAC/FTIR TC exceeds the "true" mean value by (0.4 ± 0.2)% (Table 1, Fig. 4). This difference is statistically significant with a probability of 95%. Since the values of μa for EMAC TC and EMAC/FTIR TC are the same, the difference in mean values is caused by a rightward shift in the entire distribution of EMAC/FTIR TC (see Fig. 3);

(ii) the mean value of MF CH4 estimated from the sample series of EMAC/FTIR MF is on average (0.2 ± 0.06)% lower than the "true" values calculated from the complete array of EMAC MF (see Table 2). The resulting difference is also statistically significant with a probability of 95%.

At latitudes where the observation station is located, these shifts are mainly caused by the relatively small number of sunny days, which are often accompanied by anticyclonic activity. The growth in atmospheric pressure leads to an increased total column of methane. Indeed, a comparison of the average levels of surface atmospheric pressure for EMAC and EMAC/FTIR showed that the pressure for the sample array is on the average of 0.6% higher than for the complete array. On the other hand, anticyclones are typically characterized by downward and horizontally divergent air flows [42]. These flows transport air with a lower concentration of methane from the upper troposphere and can serve as a cause for the negative shift for MF CH_4 obtained by us.

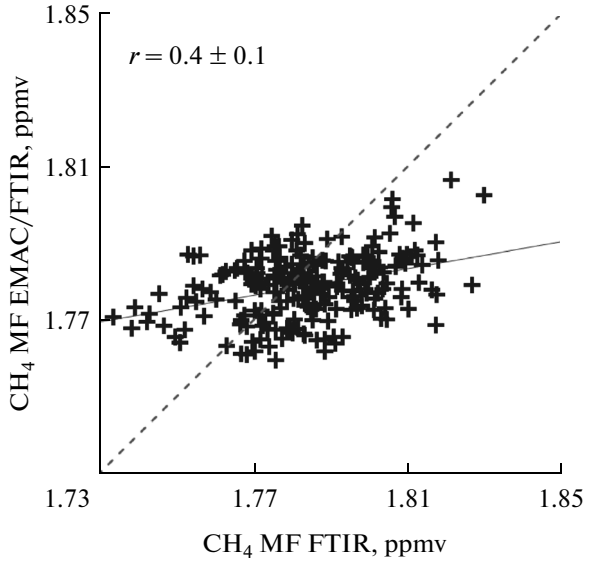

Fig. 6. Diagram of scattering for EMAC/FTIR MF and FTIR MF data.

Thus, a comparison of average levels of methane TC and MF obtained from FTIR measurements with, for example, climatological or model data for northwestern Russia requires that possible systematic shifts should be taken into account.

5. CONCLUSIONS

A combined analysis of experimental data and simulation results on atmospheric methane at Petergof station showed that the following:

(1) The data of FTIR measurements of the total column (TC) of methane are systematically higher than the simulation results (the difference for 2009–2012 is 1.3%).

The variations in experimental data on TC are higher than in model results: the σ values are 1.6% and 1.2%, respectively.

(2) The values of column-averaged mole fraction (MF) of methane obtained from observations are 0.3% higher than for the EMAC model. The reduction of the systematic difference between measured and simulated data for MF by 1% in comparison with TC is caused by the fact that the model data on surface pressure are lower than measurement data by 1% on the average.

The variations in experimental and model values of MF CH_4 are 1% and 0.5%, respectively, which is ~0.6-0.7% less than for TC.

(3) The correlation coefficients calculated for experimental and simulated data of TC and MF for 2009–2012 are statistically significant with a probability of 95%: 0.82 ± 0.08 and 0.4 ± 0.1, respectively. This means that

(i) the EMAC model reproduces the meteorological impact on observed changes in TC CH_4 in the atmosphere well;

(ii) the mechanisms of MF CH_4 formation (for example, the sources and sinks of methane) are currently known with insufficient accuracy.

(4) Estimates for the relative contribution of pressure variations to the total variance of experimental (FTIR TC) and model (EMAC/FTIR TC) series differ in two times: 36% and 74%, respectively.

(5) Based on an analysis of EMAC model data for 2009–2012, we found that

(i) the average value of the EMAC/FTIR TC series (days of FTIR measurements) is 0.4% higher than the "true" average value calculated for the complete model array of EMAC TC;

(ii) the average value of the EMAC/FTIR MF series is 0.2% lower than the "true" average value calculated for the complete array of EMAC MF.

This is mainly caused by the relatively small number of sunny days (measurement days) accompanied by anticyclonic meteorological conditions.

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