

# Ground-Based Measurements of the Total Column of Freons in the Atmosphere near St. Petersburg (2009–2017)

A. V. Polyakov<sup>a,\*</sup>, Yu. M. Timofeyev<sup>a</sup>, Ya. A. Virolainen<sup>a</sup>, M. V. Makarova<sup>a</sup>,  
A. V. Poberovskii<sup>a</sup>, and H. K. Imhasin<sup>a</sup>

<sup>a</sup>*Saint Petersburg State University, St. Petersburg, 199034 Russia*

\**e-mail: a.v.polyakov@spbu.ru*

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**Abstract**—The results of the first long-term (2009–2017) ground-based spectroscopic measurements of the total content (TC) of a number of freons in Russia are presented. According to measurements in Peterhof, TCs of CFC-11 and CFC-12 decrease at a rate of ~0.6% per year and TC of HCFC-22 grows at a rate of ~2.7% per year, which is in good agreement with independent measurements. The seasonal course of freon TC in the area of St. Petersburg is registered: highs of CFC-11 and CFC-12 are observed in summer and lows are in late winter and spring. For the HCFC-22 TC, the opposite seasonal course is observed, with a maximum in winter and a minimum in summer.

**Keywords:** chlorofluorocarbons (freons), total content in the atmosphere, ground-based transmittance method

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## INTRODUCTION

Halogen-containing atmospheric gases play an important role in the destruction of the ozone layer in the Earth's atmosphere [1]. For example, the main products of photolysis of freons CFC-11 (chemical formula  $\text{CCl}_3\text{F}$ ), CFC-12 ( $\text{CCl}_2\text{F}_2$ ) and HCFC-22 ( $\text{CHF}_2\text{Cl}$ ), which are studied in this work, are chlorine atoms, which play an important role in the destruction of stratospheric ozone [2]. In addition, these gases absorb thermal infrared radiation and significantly contribute to the greenhouse effect [3]. Finally, due to the long lifetime (CFC-11 from 43 to 67 years, CFC-12 from 88 to 122 years, HCFC-22 ~ 12 years; these estimates were obtained from the results of various publications and given in [4]), these gases are good monitors in the study of transport processes in the upper troposphere and lower stratosphere [5, 6].

Measurements of the concentration of freons in the atmosphere started in the 1970s using a variety of local and remote methods (e. g., see [7]). Ground-level freon concentrations are measured at a number of stations entering the Advanced Global Atmospheric Gases Experiment (AGAGE) and NOAA's Halocarbons & other Atmospheric Trace Species Group (HATS) networks. The total content (TC) of a number of freons has been determined for a long time using the ground-based spectroscopic IR method [8–10]. The results of the first measurement of the CFC-12 TC in Russia are given in [11].

The satellite remote sensing methods have played an important role in the study of the global distribution of the content of a number of freons and in the assessment of their long-term trends [7]. Satellite observations were performed using IR Sun occultation method (ATMOS, ILAS, and ACE instruments) and a measurement of the planet's own IR radiation (instruments CIRRIS, CRISTA, CLAES, HIRDLS, and MIPAS) [12].

Ground and satellite measurements have shown that the freon content in the 1970s–1980s was constantly increasing and the surface concentrations of CFC-11 and CFC-12 in the troposphere reached their maxima in 1992 and 2003, respectively. In connection with the negative impact of these gases on the ozone layer, in September 1987 an international agreement (Montreal Protocol) was adopted that was aimed at stopping their usage and replacing them with other gases in industries and households [13]. Later, thanks to the adopted restrictions, there was a decrease in the content of freons in the atmosphere [14–16].

However, HCFC-22 continues to be widely used in developing countries and, at present, both its surface concentrations and TC exhibit an increase.

## 1. GROUND-BASED MEASUREMENTS OF SOLAR SPECTRA

Measurements of infrared spectra of solar radiation were carried out in St. Petersburg's Peterhof suburb at

**Table 1.** Parameters used in the inversion of spectral measurements and the number of degrees of freedom of the useful signal (DOFS [19])

Gas under study	CFC-11	CFC-12	HCFC-22
Spectral interval, $\text{cm}^{-1}$	830–860	922.5–923.6	828.75–829.4
Recoverable profiles	Gas under study		
Recoverable TCs of influencing gases	$\text{H}_2\text{O}$ , $\text{CO}_2$ , $\text{O}_3$ , $\text{HNO}_3$ , $\text{COCl}_2$	$\text{H}_2\text{O}$ , $\text{CO}_2$ , $\text{O}_3$	$\text{H}_2\text{O}$ , $\text{CO}_2$ , $\text{O}_3$
Spectroscopic information	PL <sup>1</sup> , HITRAN-2008, 2009		
A priori profile of studied gas	Average profile of WACCM for 1980–2020		
Calibration parameters specified in the solution of the inverse problem	Slope, curvature, parameters of sine distortion (beam) <sup>2</sup> , modulation depth, and phase shift of the interferometer (ILS parameters) <sup>2</sup>		
DOFS, average	1.3	0.9	0.8

<sup>1</sup>PL indicates pseudolines (<http://mark4sun.jpl.nasa.gov/pseudo.html>) modeling the spectroscopic parameters of absorption of CFC-11, CFC-12, and HCFC-22. <sup>2</sup>Terms used in the program SFIT4 are given in parentheses.

a station of the Network for the Detection of Atmospheric Composition Change (NDACC) St. Petersburg (29°49′15″ E, 59°52′56″ N; instrument height above sea level is 21 m). In this paper we report measurement results obtained from April 2009 to March 2017. Spectra were recorded by a Bruker 125HR instrument with a spectral resolution of  $0.005 \text{ cm}^{-1}$  [17]; interferograms were converted to spectra without apodization (boxcar). The signal-to-noise ratio, calculated for the dark noise, reached a value of 700, and typical values were 400–500. Measurements of atmospheric parameters are based on an interpretation of the IR spectra of solar radiation using international program (computer code) SFIT4 [18]. For analysis, we selected the results of a solution of the inverse problem obtained under the conditions of convergence and stability of the solution and the value of the spectral discrepancy of no more than 2%.

The information on a number of parameters for the solution of the direct and inverse problem (spectral intervals, gases that affect the transparency of the atmosphere in these intervals and are specified in the interpretation of spectra, other specified parameters, spectroscopic and a priori information, etc.) is given in Table 1. The spectral measurement intervals were chosen on the basis of two conditions: maximum absorption by the studied gases and simultaneously minimum absorption by other atmospheric gases, primarily water vapor. Therefore, we used a set of spectral intervals of work [20], since the spectral intervals of the later work [12] are optimized for the humid tropi-

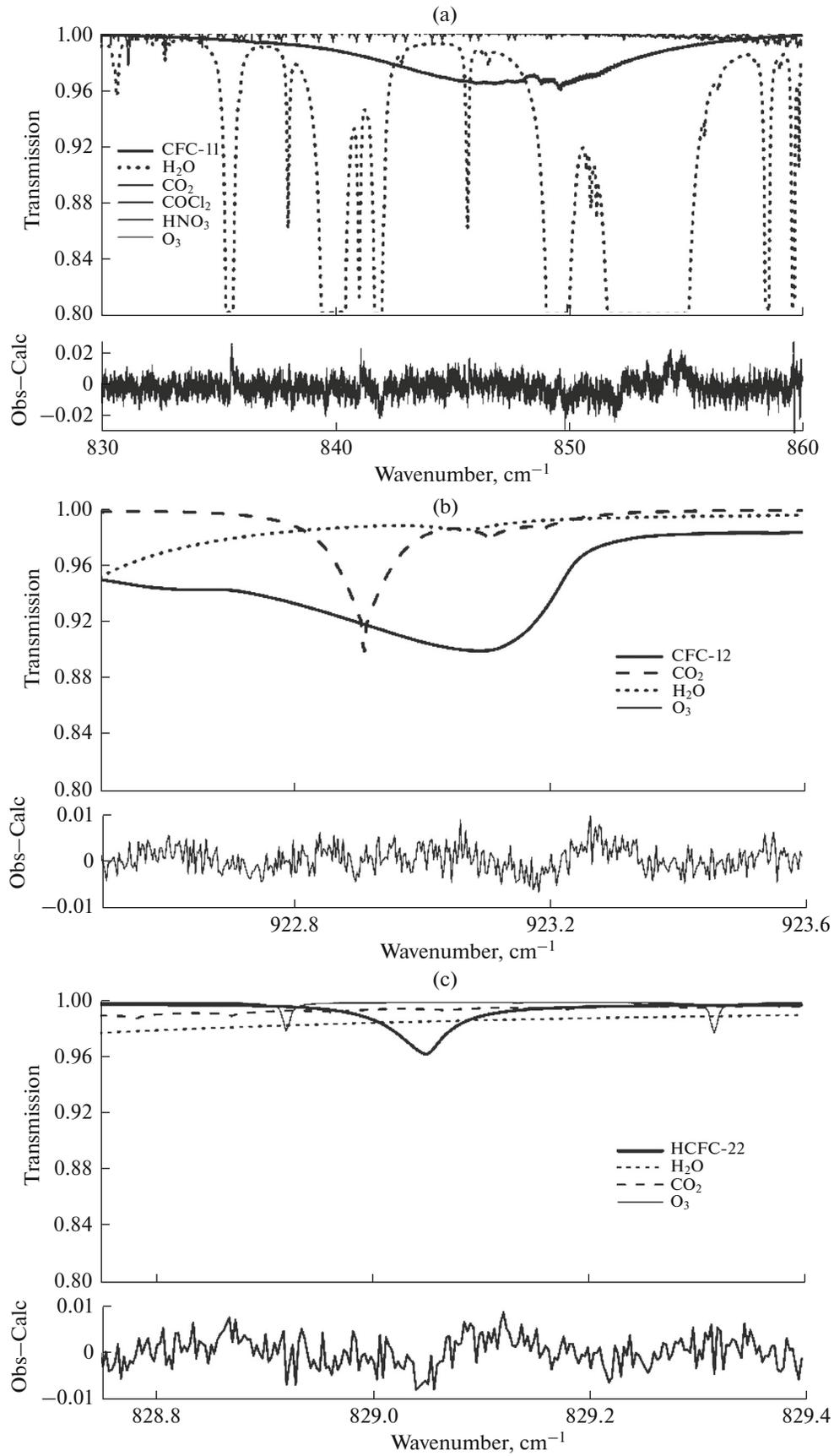
cal atmosphere and are not optimal for the station under consideration.

The values of the number of Degrees Of Freedom of the useful Signal (DOFS) [19] in the last row of Table 1 are close to 1. This indicates that the spectral measurements contain only data on the TC of the studied gases, whereas the information on their vertical profiles is practically absent.

Figure 1 shows typical examples of transmission functions (TFs) in the studied spectral intervals for conditions of Summer 2016. The difference between the measured and calculated (after solving the inverse problem) values of TF (spectral residual) is shown as well. As a rule, the residual is small in all intervals and does not exceed a fraction of a percent. As is seen from Fig. 1, all three analyzed gases absorb a small (less than 10%) fraction of solar radiation, resulting in a relatively low accuracy of the TC measurements.

The absorption of CFC-11 (Fig. 1a) does not exceed 4%. In fact, the entire spectral region used in the measurements is overlapped by a number of absorption lines of water vapor, in the centers of some of which there is complete absorption (not shown in Fig. 1a because of the scale). The absorption on the wings of these lines remains noticeable in the spectral region up to a few  $\text{cm}^{-1}$  from the centers of the lines. Obviously, this evidences the importance of accurate determination and setting the water vapor content, as well as the low information value of spectral measurements for CFC-11 and, consequently, the relatively low accuracy of determining its content in the atmo-

**Fig. 1.** Examples of TFs of atmosphere of different gases in used spectral windows: (a) CFC-11, (b) CFC-12, and (c) HCFC-22 (top of figures). The lower parts of the figures are the “experiment minus calculation” spectral residuals after solving the inverse problem. The calculations were performed for the measurement conditions in the summer of 2016. Transmittances of  $\text{O}_3$ ,  $\text{CO}_2$ ,  $\text{COCl}_2$ , and  $\text{HNO}_3$  in part (a) are shown by the same thin black line; for all these gases, transmittance is close to 1 and the corresponding curves for the most part mutually overlap.



sphere. However, due to the large width of the used spectral range of  $30\text{ cm}^{-1}$  and, consequently, the large number of used TF values ( $\sim 6000$ ) and the corresponding number of random noise suppression, the number of independently determined parameters (DOFS) is greater than for the other two gases (1.3). It should be noted that, in the course of solving the inverse problem, the water vapor TC, which is essential for evaluating the CFC-11 TC, was independently clarified from the same solar measurements, whereas the a priori profile of its concentration in the volume was preliminarily determined for each day using results of spectral measurements in specially selected spectral intervals. The results were processed by ROFFIT software [18, 21], which is used in NDACC for interpreting spectral measurements.

Figure 1b shows that for CFC-12 the maximum absorption is about 10%, with carbon dioxide absorption lines and the wing of the water absorption line being partially in the absorption region of CFC-12. As a result, DOFS is on average even slightly below 1 (0.9), which is due to the relatively narrow width of the used spectral interval ( $1.1\text{ cm}^{-1}$ ).

The absorption of HCFC-22 (Fig. 1c) reaches about 4% at maximum, with the entire absorption region being also partially overlapped by the wing of a water vapor line. A narrow spectral interval ( $0.65\text{ cm}^{-1}$ ) is used and, as a consequence, there is a relatively small number of spectral samples, which leads to the minimal value (0.8) of DOFS for this gas.

## 2. METHOD OF INTERPRETATION AND MEASUREMENT ERRORS

The measured spectra were analyzed with the help of the SFIT4 software [18], which uses the method of optimal estimation (statistical regularization) [19]. This software solves direct and inverse problems, using a variety of a priori information, and estimates the uncertainties of the solutions, which are due to numerous factors, including the characteristics of the device, the quality of a priori information (spectroscopic parameters, a priori profiles, etc.), and the measurement conditions (in total, more than 20 factors).

SFIT4 was also used to calculate all the main random and systematic errors due to the measurement noise, smoothing the profile variation in the process of solving the inverse problem, and the quality of various a priori information (such as temperature profiles, humidity and the presence of other interfering gases, intensities and half-widths of spectral lines (and pseudo-lines) and their dependences on temperature and pressure, etc.). The total errors of determining the TC of the studied gases are given in Table 2.

Random errors of determining the TC of the studied gases are 3.4–3.7% and systematic are 3.2–7.5%. The systematic and random errors are maximal for CFC-11, which is primarily caused by the large (7%)

**Table 2.** Total (systematic and random) errors (%) of determining TCs of CFC-11, CFC-12, and HCFC-22

Gas	Total error	
	systematic	random
CFC-11	7.5	3.7
CFC-12	3.2	3.5
HCFC-22	4.6	3.4

**Table 3.** Main characteristics of the results

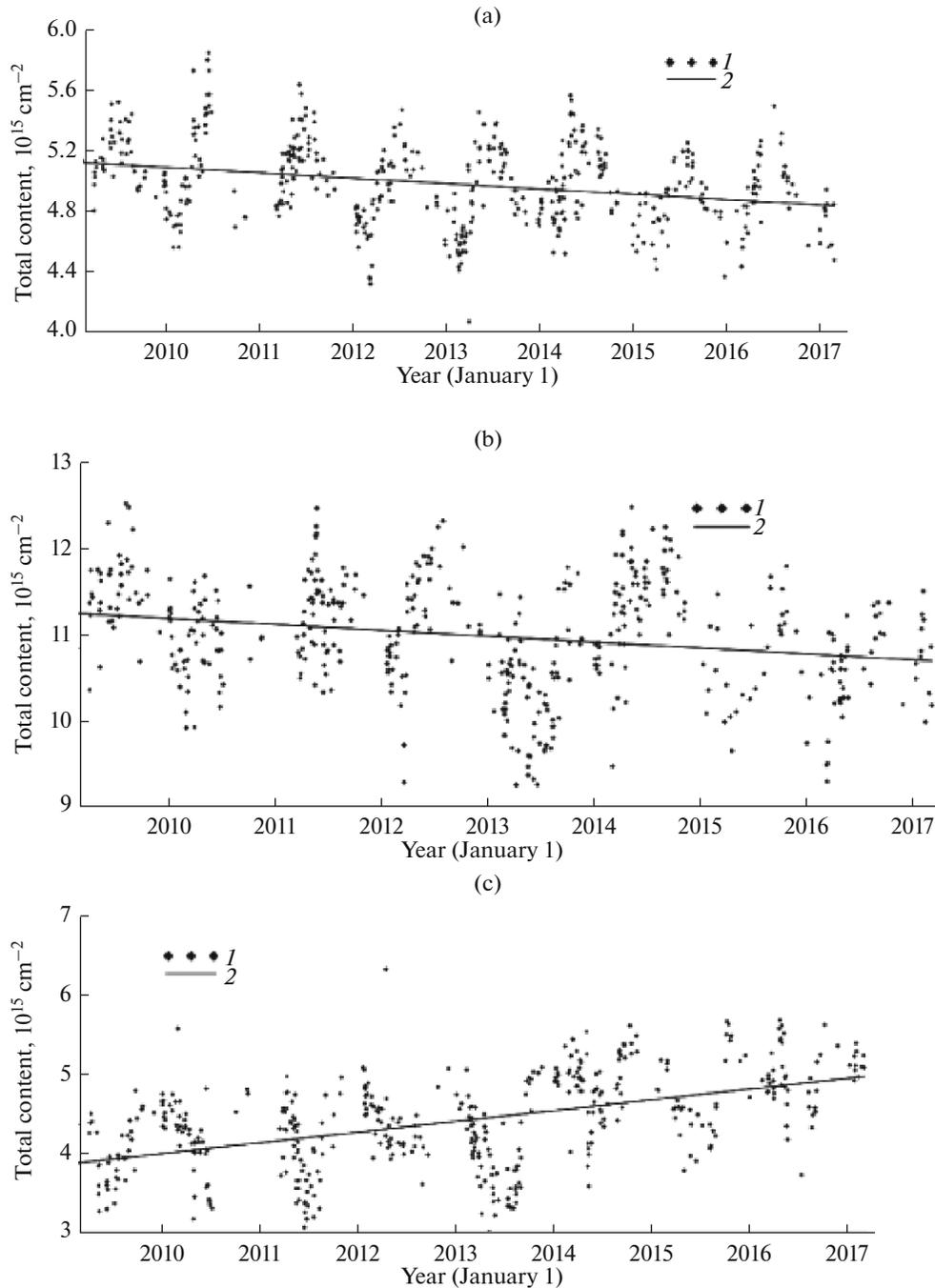
No.	Gas	CFC-11	CFC-12	HCFC-22
1	Number of measurement days	502	458	390
2	Number of single measurements	2976	2250	1631
3	RMS variability of TC during the day, %	1.4	2.2	3.9

error of the spectroscopic parameters for this gas. The errors in setting the spectroscopic parameters also dominate when determining the TCs of CFC-12 and HCFC-22. Other important factors affecting the errors of determining TC are the errors of the atmospheric temperature and the errors of fixing the height of the Sun and smoothing (especially for HCFC-22).

Comparing our errors with the errors obtained in [12], it can be noted, that for CFC-11 and CFC-12, the average errors in Peterhof are  $\sim 1.5$  times higher due to the fact that the measurements in the above work were carried out in more favorable conditions at low latitudes (station Réunion Island  $-20^\circ\text{ S}$ ) for relatively small zenith angles of the Sun.

Some characteristics of the Peterhof results are given in Table 3, including the number of days and single measurements and the mean square (MS) variability of the TC values during the day. The number of measurements is markedly different, as is the number of measurement days for each gas, which is determined by the operation of the convergence and stability criteria and the magnitude of the spectral discrepancy.

The mean square variability of TC during the day characterizes the total variability both due to random measurement errors and due to the natural variability of TCs of the studied gases. The mean square variability for CFC-11 and CFC-12 (1.4 and 2.2%, respectively) is significantly less than the previously given numerical estimates of random errors, whereas for HCFC-22 it is very close to our numerical estimates of random error components (3.7, 3.5, and 3.4%, see Table). This is likely caused by overestimating the systematic error of the used temperature profile, which we set according to the recommendations of NDACC



**Fig. 2.** Average daily TC of (a) CFC-11, (b) CFC-12, and (c) HCFC-22 measured in (1) Peterhof and (2) linear regressions for the entire measurement period.

in the range of 1–4 K, depending on the height in the atmosphere.

### 3. MEASUREMENT RESULTS

Figure 2 shows the results of measurements of freons TC (average daily values in  $\text{cm}^{-2}$ ) at Peterhof station for 2009–2017.

The following features are seen from the results in Fig. 2:

(1) CFC-11 and CFC-12 TCs are decreasing with time, whereas HCFC-22 TC is growing. As we already mentioned above, the decrease is due to the 1987 ban on these compounds in industry and in everyday life. The growth of the HCFC-22 TC is due to the continued use of this compound in developing countries.

**Table 4.** Trends of freon TCs based on the measurements in Peterhof; satellite measurements near Réunion Island [12] and according to the WMO [4]

Type of measurements/trend estimation period	Trend magnitude, % (halfwidth of interval for probability of 95%)		
	CFC-11	CFC-12	HCFC-22
Ground-based/04.2009–04.2017	−0.57 (0.14)	−0.61 (0.21)	2.73 (0.34)
ACE-FTS/2004–2010	−0.9 (0.1)	−0.4 (0.1)	3.7 (0.1)
MIPAS/2004–2011	−1.07 (0.06)	−0.48 (0.05)	4.05 (0.30)
WMO [4]	AGAGE, in situ	−0.9	2.6
	NOAA, flask & in situ	−1.0	2.5
	UCI, flask	−1.1	2.6

(2) There are noticeable temporal variations of TC of the studied gases, the amplitude of which reaches ~15% for CFC-11 and CFC-12 and ~30% for HCFC-22. The standard deviation of TC is ~5.5% for CFC-11 and CFC-12 and ~11% for HCFC-22. These variations are due to both random measurement errors and the natural variability of TC.

(3) The measurements reveal seasonal variability of TCs of the studied gases. For CFC-11 and CFC-12, the minimum values of TCs in Peterhof are observed in spring, and the maximum values are in summer. TC of HCFC-22 is maximal in winter and minimal in summer. The amplitudes of seasonal variations of freon TCs change considerably from year to year.

The results of our measurements allow us to evaluate the long-term trends for the studied freons. It should be noted that the trends for these gases have already been determined several times using the data of measurements of surface concentrations, ground spectroscopic measurements of the TC at different stations, and various satellite measurements (MIPAS, ACE, etc.). Table 4 gives the values of trends and their uncertainties according to measurements in Peterhof, as well as the trends obtained from satellite measurements using ACE-FTS and MIPAS instruments [12] and the data of the WMO report [4]. A comparison shows that, taking the differences in the analyzed time periods of measurements and different characteristics (total content, content in individual layers, and near-surface concentrations) into account, as well as spatial averaging, the data from independent measurements are in reasonable agreement.

The global trend for CFC-11 is well-matched by all independent estimates within the uncertainty range, and can be estimated as 1%. At the same time, measurements in Peterhof give a much smaller value, as can be seen from Table 4, which is likely due to the influence of the metropolis. For CFC-12, all estimates of the trend value are consistent within the given uncertainties. For HCFC-22, the estimates of the trend obtained by satellite (3.7 and 4.05%) differ from the value of ground measurements (2.5–2.73%) of

near-surface concentrations conducted in background conditions.

#### 4. ANALYSIS OF MEASUREMENTS

The time variation of freon content was studied using ground and satellite measurements. Ground measurements [8] for the Arctic regions revealed a similar seasonal dependence of the CFC-12 TC (with a maximum in summer time) and the absence of such a dependence for the CFC-11 TC. According to the measurements at the Jungfraujoch station, the seasonal variations of CFC-11, CFC-12, and HCFC-22 exhibit similar behavior with maximal TC in summer–autumn and minimal in late winter–early spring [22]. The TC data for the considered gases obtained at the NDACC Eureka station (89.05° N, 86.42° W) [23] are also close to the above dependencies. Satellite instruments (MIPAS, ACE-FTS, and HIRDLS) used the limbic geometry of measurements and therefore provided mainly information about the content of the studied gases in the stratosphere. A comparison of the climatology of the content of CFC-11 and CFC-12 obtained with the help of these instruments is reviewed [24]. According to these satellite measurements, seasonal and annual variations of CFC-11 are determined by quasi-biennial oscillations (QBOs) in the tropics and by the annual cycle at high latitudes (see, for example, [25]). The annual cycle at high latitudes is determined by the descending air flow in the winter polar vortex, which was recorded by all three satellite instruments in the Northern Hemisphere.

Paper [12] presents the results of an analysis of the seasonal course of CFC-11, CFC-12, and HCFC-22 content in the 6–30 km layer obtained by spectroscopic ground measurements at Saint Denis and Maïdo stations on Réunion Island (~20° S) in the Southern Hemisphere and measurement data obtained with the MIPAS satellite instrument. Both types of measurements reveal similar variations with maximal values in February–April (summer–early autumn in the Southern Hemisphere) and minimal values in August–October (late winter–early spring),

but the amplitude of the variations is much smaller than that of the TC variations observed at Peterhof station. The latter is explained by the fact that the 0–6 km layer is not taken into account in [12].

## CONCLUSIONS

(1) We reported the main features of ground-based spectral measurements of direct solar infrared radiation in the Peterhof district of St. Petersburg. The measurements were carried out on a high spectral resolution Fourier spectrometer and used to determine the total content of CFC-11, CFC-12, and HCFC-22.

It is shown that the absorption of solar radiation by the studied gases is insignificant and, on average, does not exceed 10%.

(2) Information on peculiarities of interpreting ground-based spectral measurements of solar infrared radiation with the help of international computer code SFIT4 is given. The total systematic and random errors of determining the TC of CFC-11, CFC-12 and HCFC-22 are in the range of 3.2–7.5% and 3.4–3.7%, respectively.

(3) The first long-term (2009–2017) regular ground-based spectroscopic measurements of TCs of a number of freons in Russia were carried out. It is shown that TCs of CFC-11 and CFC-12 decrease, whereas the TC of HCFC-22 grows. This decrease is due to the ban on CFC-11 and CFC-12 in industry and in everyday life, and the growth is due to the continued use of HCFC-22 in a number of developing countries. According to measurements at Peterhof, TCs of CFC-11 and CFC-12 decrease with a rate of ~0.6% per year, and the TC of HCFC-22 grows with a rate of ~2.7% per year, which is quite consistent with independent measurements, given the differences in methods and locations of measurements.

(4) The seasonal course of freon TCs in the St. Petersburg area was recorded. The maximum TCs of CFC-11 and CFC-12 were observed in summer and the minimal were in late winter and spring. For HCFC-22, the opposite seasonal course was observed with a maximum in winter and a minimum in summer. Features of the seasonal course require further research and modeling.

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