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Variations in the total column abundances of atmospheric carbon monoxide and methane in the polar regions

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Abstract: Atmospheric carbon monoxide and methane were studied spectroscopically in the Arctic and Antarctic. Seasonal variations of CO are evident in both polar regions, absolute values of abundance being three times larger in the Arctic than in the Antarctic. Increasing concentration trends were confirmed for both gases: 0.8% per year for Antarctic CO, 0.5% per year for Antarctic CH_4 and 3.1% per year for Arctic CH_4.

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Introduction

Carbon monoxide and methane are permanent components of the terrestrial atmosphere and their study is very important. Atmospheric cycles of these gases are interconnected; oxidation of tropospheric methane is one of the important sources of CO (Crutzen, 1983). The main sink of both gases is oxidation by tropospheric hydroxyl. Sources of these gases are diverse, with a considerable human-induced contribution which has disturbed the natural budget. The concentrations of CO and CH_4 demonstrate a tendency for temporal increase at the rate of 1 to 2 % per year (Dianov-Klokov & Yurganov 1989, Khalil & Rasmussen 1990). According to recent estimates, the mean global methane concentration has more than doubled relative to the natural level (Stauffer *et al.* 1985), and the Northern Hemispheric CO concentration has probably tripled (Dianov-Klokov & Yurganov 1989).

The turnover time of carbon monoxide in the atmosphere is an order of magnitude lower than that of methane, and therefore CO has a remarkable seasonal course and is unevenly distributed between the hemispheres (Dianov-Klokov & Yurganov 1989, Dianov-Klokov *et al.* 1989). An averaged picture of a seasonal-latitudinal global CO distribution is displayed in Fig.1 (Dianov-Klokov & Yurganov 1981). The features of the methane global distribution are similar to that of carbon monoxide but the variations are much less and do not extend beyond a few percent (a difference between the hemispheres, according to Steele *et al* (1987), no greater than 6%.

The study of the abundances of both gases is important for three reasons. Firstly, the above-mentioned increases of both gases are probably caused by human activities. Secondly, methane contributes significantly to the absorption of IR radiation generated by the Earth's surface and consequently

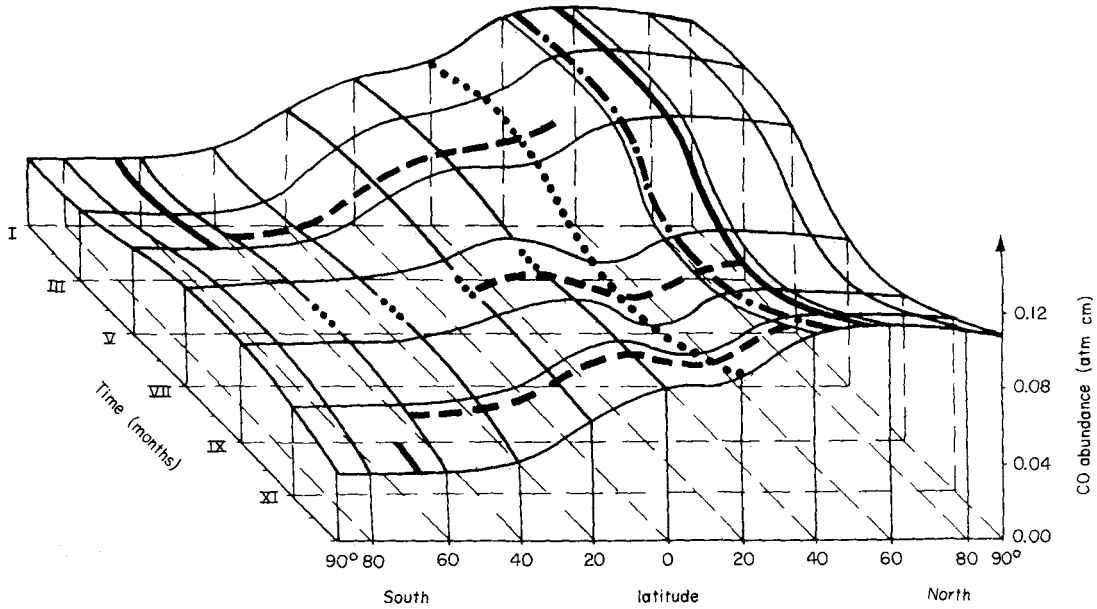

Fig. 1. Model for the global latitude-time distribution of the total content of CO given by Dianov-Klokov & Yurganov (1981). The thick curves are smoothed experimental data for 1970–1978

to the atmospheric "greenhouse effect". Thirdly, their increase diminishes the concentration of hydroxyl which is the main destroyer of many chlorofluorocarbons, hydrocarbons, organics etc.

In this paper the results of spectroscopic measurements of total column abundances of carbon monoxide and methane in the Arctic and Antarctic atmospheres are reported. These results extend the polar data base published earlier by Voskresensky *et al.* (1989) and Dianov-Klokov & Yurganov (1989). Some peculiarities of atmospheric budgets of these gases in polar regions are discussed.

Methods

Spectra of the IR solar radiation (about 3.3 μm for CH_4 and 4.6 μm for CO) absorbed by the total atmospheric layer were measured using grating spectrometers with a spectral resolution 0.2 to 0.3 cm^{-1} designed by the U.S.S.R. Institute of Atmospheric Physics. Total column gas abundance was determined by the area inside the spectral line contour. The measurements are possible at solar zenith angles lower than 75°. The unit of measurement is atm.cm, i.e. the depth of the gas layer reduced to normal conditions. 1 atm.cm = 2.69 x 10^{19} molec cm^{-2}. The measurement techniques are described in more detail by Dianov-Klokov *et al.*(1989).

It should be noted that the spectroscopic methods average gas concentration over the whole tropospheric thickness. Thus gas fluctuations, both natural and artificial, are smoothed out, in contrast to *in situ* methods (e.g. gas chromatography). But on the other hand the measurement precision diminishes due to unavailable information about atmospheric parameters through a long, inhomogeneous atmospheric path. In particular, an important disadvantage of this technique is the dependence of a measured gas abundance on the shape of the gas vertical distribution. Measurement error for the case of an unknown CO gas profile was studied by Dianov-Klokov *et al.* (1989) with the aid of a computer model. It has been noted that spectra are not sensitive to variations of stratospheric concentrations of the gas; after the recalculation of the total abundance U into the mean concentration of the gas R (using the relation R=U/H, where H=7.9 km), the latter value does not differ significantly from the mean tropospheric mixing ratio (maximum possible error for all likely shapes of vertical CO distributions is ± 4%).The CH_4 vertical distribution is more even than that of CO and such a problem does not arise. The total uncertainty of a single measurement for both gases is estimated by Dianov-Klokov *et al.*(1989) as ± 8 to 10%.

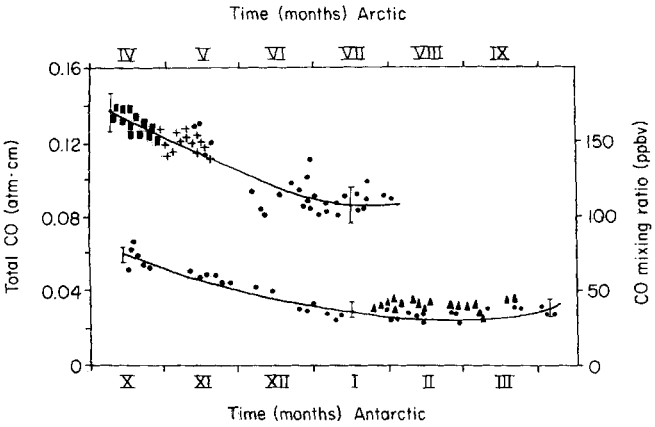

Fig. 2. Seasonal variations of the total CO abundance in Arctic (upper curve and upper axis) and in Antarctic (lower curve and lower axis). The right scale (here and in Fig. 3) is the tropospheric mean on CO mixing ratio. Vertical segments are daily means for Zhokhov Island (1982) (Gabriyelyan *et al.* 1983), crosses are for Severnaya Zemlya (1983), and dots are for Wrangel Island (1987). For the Mirny (Antarctic) data, results are indicated by dots (1987) and triangles (1989)

Results

Measurements of CO and CH_4 total abundances were carried out in the Soviet Arctic (Severnaya Zemlya Archipelago, Novosibirskiye Island, Wrangel Island) and in Antarctic (Molodezhnaya and Mirny Stations). Weather and solar height conditions limited the period of measurements to spring and summer. During one month we had usually 10–15 days of measurements and for one day 10–20 spectra were measured as a rule. A full list of data is given in the appendix. Standard deviations of daily means from monthly means did not exceed the experimental uncertainty, i.e. ±8%.

Some daily means of CO abundance are shown in Fig. 2. Curves are the result of smoothing all the measured values (listed in the Appendix). Absolute values of carbon monoxide content in Antarctica were about three times lower than in the Arctic, but seasonal variations in both regions had similar shapes: diminishing during spring and a minimum in late summer. It should be noted that for comparability the smoothed curves in Fig. 2 were shifted one relative to another by six months.

The apparent similarity of CO seasonal variations suggests a common cause; obviously it is the variations of the main CO destroyer hydroxyl. During a winter an accumulation of carbon monoxide associated with the action of all its sources, including man-made ones, takes place in both hemispheres. During a spring increasing solar radiation generates more hydroxyl thus destroying CO. The relatively short CO life-time (a few months) makes seasonal changes possible.

Total column contents of methane are displayed in Fig. 3. One can see that methane abundances do not show any seasonal variations larger than precision of our technique in either hemisphere. It is interesting to note that the reaction with hydroxyl is the main sink for methane as well. The lack of seasonal variations larger than 8% is a consequence of the longer life-time.

The length of the experimental data record (1978–1989 for the Antarctic and 1982–1988 for the Arctic) permits us to evaluate the degree of inter-annual variability of these gases

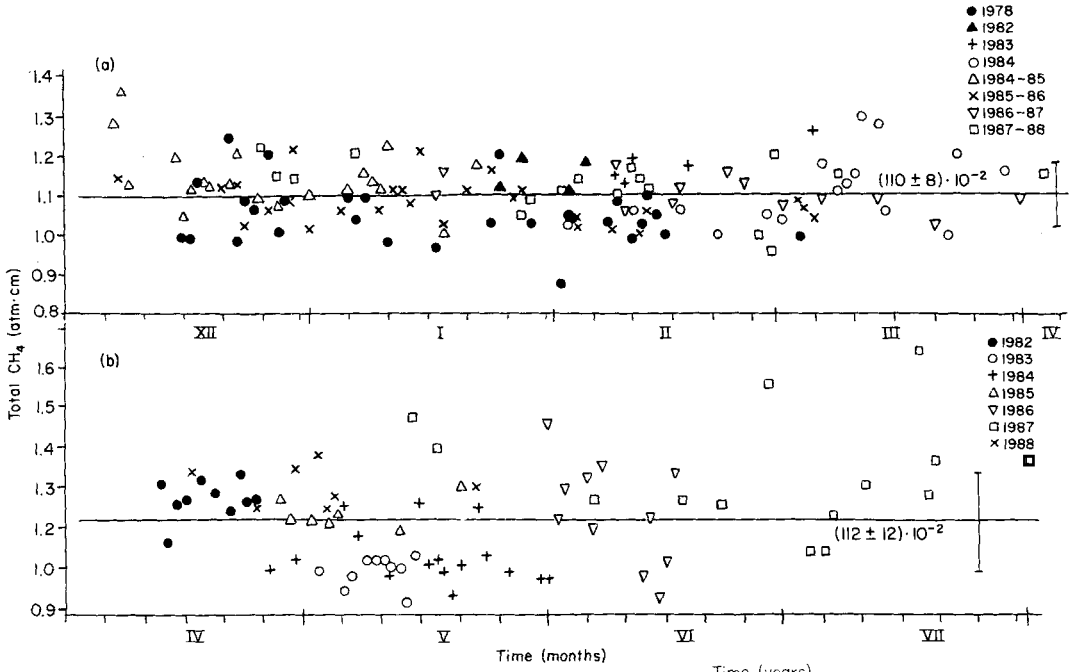

Fig. 3. Daily means of column CH_4.
a. Antarctic.
b. Arctic

and possible trends in the polar regions. Unfortunately, the Arctic measurements were carried out in different months in different years and hence strong CO seasonal variations prevented trend analyses. To exclude seasonal variations from the Antarctic measurements only January–February results were used; the variations are at a minimum during these months (see Fig. 2). Values of CO and CH_4 abundances averaged over all the results of the given year (CH_4), or over January–February values (CO) are shown in Fig. 4.

The slopes of regression lines, recalculated into mean tropospheric mixing ratio areas follows:

(a) for Antarctic CO, 0.3±0.1 ppbv. yr^{-1} (excluding (scattered data for 1982 and 1989),
(b) for Antarctic CH_4, 6.7±1.2 ppbv. yr^{-1} and
(c) for Arctic CH_4, 43±13 ppbv. yr^{-1}.

In all cases above, root-mean-square uncertainties are given and the trend is significant at the 95% probability level. The trend of CO values in May in the Arctic is positive but statistically insignificant. The values of trend in percent per year can be seen in Fig. 4.

Inter annual fluctuations of CO abundances seem to be real. For example, Fraser *et al.* (1986) observed reduced values of the CO mixing ratio in the summer of 1982 at the station Mawson, located 600 km from Mirny (cf. Fig.4). The trends reported here are in accordance with previously published earlier globally averaged estimates: CH_4(16.6±0.4) ppbv. yr^{-1} (Khalil & Rasmussen 1990), CO (0.8±0.3) ppbv. yr^{-1} (Khalil & Rasmussen 1988). Khalil & Rasmussen (1988) obtained statistically insignificant CO trends for Tasmania (0.2±0.4)ppbv. yr^{-1} and for Alaska (0.5±0.5)ppbv. yr^{-1}.

The spectroscopic measurements of these gases in both polar regions complement measurements made by *in situ* methods (Fraser *et al.* 1986, Khalil & Rasmussen 1990). The northern and the southern polar regions have obvious similarities: a long polar night, frequent temperature

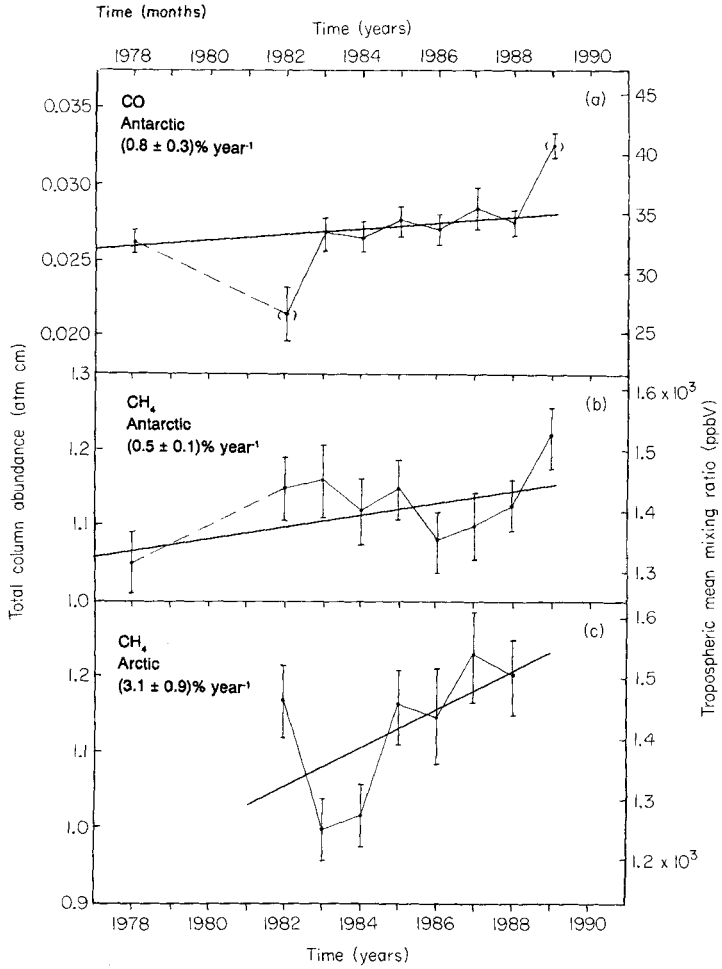

Fig. 4. Values of CO and CH_4. **a.** Total abundance of CO for Antarctic stations averaged over January–February. **b.** Total abundance of CH_4 for Antarctic stations averaged over whole measurement periods given in Appendix. **c.** As **b.** for Arctic stations. In all cases, confidence limits correspond to 90% probability.

inversions, an absence of local pollutions and so on. A significant difference between them is the different impact of Man's industrial activity on the chemical composition of the atmosphere on a chemispheric scale. For carbon monoxide this difference was possibly reflected in different percentage increases in the two hemispheres: 1.5–2% yr^{-1} for mid latitudes of Northern Hemisphere (Dianov-Klokov & Yurganov 1989) and less than 1% yr^{-1} in Antarctic. A difference between the estimated Arctic and Antarctic methane trends is unlikely in view of its long life-time, which is sufficiently longer than the time of interhemispheric mixing.

Conclusions

Results of simultaneous spectroscopic measurements of total column CO and CH_4 in the Arctic and Antarctic reveal both seasonal, inter annual and long-term variations of these gases. Increasing trends for both carbon monoxide and methane are observed in both polar regions, at least qualitatively. The results are consistent with previously published *in situ* tropospheric data. The variations (especially long-term ones) have very important climatological and photo chemical consequences.

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Appendix 1. Monthly measurements of carbon monoxide abundance from the Arctic and Antarctic for the periods covering 1977–1978 and 1982–1990

Table I. Molodezhnaya (68°S, 49°E) 1977/78. Carbon monoxide abundance measurements in atm.cm x 10 000. (Left column = sampling date)

Dec. 77		Jan. 78		Feb.78		Mar. 78		Apr.78	
15	380	05	280	01	210	14	250	06	432
16	356	06	300	02	250	03	249		
17	346	07	280	07	245	20	428		
21	324	10	293	08	243	21	404		
22	361	16	250	09	267	06	432		
23	302	23	242	10	304				
24	299	24	275	11	247				
26	292	26	277	12	259				
27	286	28	234	13	286				
28	283			14	250				
30	268								
31	313								
Mean	318		270		256		360		432
sd	(34)		(22)		(24)		(79)		()

Table II a. Mirny station (67°S, 93°E) 1982–1983. Carbon monoxide abundance measurements in atm.cm x 10 000. (Left column = sampling date)

Jan. 82		Feb. 82		Feb. 83		Mar. 83	
20	222	02	203	08	245	03	278
22	198	04	260	09	266	04	300
24	197			10	246	09	283
27	222			11	262	10	266
				12	302	12	308
				14	282	13	306
				15	265	17	325
				17	254	22	319
				28	282		
Mean	210		232		267		298
sd	(12)		(28)		(18)		(18)

Table II b. Mirny station (67°S, 93°E) 1984–1985. Carbon monoxide abundance measurements in atm.cm x 10 000. (Left column = sampling date)

Feb. 84		Mar. 84		Dec. 84		Jan. 85		Dec. 85	
01	245	06	287	06	355	05	279	20	323
02	269	08	303	07	399	06	296	21	296
10	261	09	312	08	367	07	283	26	335
16	274	10	266	10	324	08	298	28	307
21	267	11	286	14	333	09	269	29	308
27	277	13	285	15	317	10	273	31	304
		14	278	16	300	12	269		
		22	253	17	255	14	264		
		23	267	18	280	17	273		
		29	306	21	298	21	261		
				22	287				
				24	298				
				27	263				
				31	265				
Mean	265		284		310		276		317
sd	(10)		(18)		(41)		(12)		(17)

Table II c. Mirny station (67°S, 93°E) 1986. Carbon monoxide abundance measurements in atm.cm x 10 000. (Left column = sampling date)

Jan.		Feb.		Mar	
04	272	02	271	03	284
09	310	03	286	04	303
10	304	11	271	15	308
11	255	12	274		
12	259				
13	257				
14	277				
17	266				
20	248				
23	262				
26	272				
27	256				
Mean	270		275		298
sd	(18)		(10)		(26)

Table II d. Mirny station (67°S, 93°E) 1987. Carbon monoxide abundance measurements in atm.cm x 10 000. (Left column = sampling date)

Jan		Feb		Mar		Apr		Oct		Nov		Dec	
12	335	04	267	04	278	02	290	16	503	10	502	08	407
13	285	05	260	06	276	03	326	17	627	15	480	15	404
14	308	07	279	09	268	05	334	18	627	16	485	25	322
16	311	08	265	13	284			19	592	21	483	27	325
17	288	12	293	19	310			21	563	23	455	29	335
23	279	15	303	20	303			28	533	27	430		
25	254	16	270	28	312								
30	258	17	252	31	318								
		22	300										
		24	291										
		28	274										
Mean	290		278		294		317		581		474		359
sd	(10)		(16)		(18)		(19)		(57)		(21)		(38)

Table II e. Mirny station (67°S, 93°E) 1988. Carbon monoxide abundance measurements in atm.cm x 10 000. (Left column = sampling date)

Jan		Feb		Mar		Apr 1	
06	284	01	259	08	296	01	302
08	260	03	263	24	301	02	279
09	291	08	278	25	314	03	289
27	281	10	275				
		11	285				
		12	269				
		26	295				
		27	287				
		28	267				
Mean	279		275		304		290
sd	(12)		(11)		(8)		(9)

Table II f. Mirny station (67°S, 93°E) 1989-90. Carbon monoxide abundance measurements in atm.cm x 10 000. (Left column = sampling date)

Jan 89		Feb 89		Mar 89		Feb 90		Mar 90	
27	313	01	313	01	306	25	298	05	286
28	325	02	333	02	306	26	295	13	295
29	333	10	333	05	295	28	282	15	298
31	345	11	332	06	257			20	303
		13	314	17	355			22	304
		14	326	18	359			24	320
		25	324					27	325
		27	307						
Mean	329		322		313		292		304
sd	(13)		(10)		(38)		(8)		(14)

Table III. Zhokhov Island (76°N, 153°E) and Severnaya, Zemlya (79°N, 95°E). Carbon monoxide abundance measurements in atm.cm x 10 000. (Left column = sampling date).

Zhokhov Is.		Severn.Zemlya			
Apr 82		Apr 83		May 83	
09	1340	30	1270	01	1190
10	1380			02	1140
11	1380			03	1160
12	1390			05	1260
13	1320			06	1190
14	1300			07	1270
15	1270			08	1190
17	1350			09	1200
19	1240			10	1240
21	1300			11	1040
22	1240			12	1040
23	1250			13	1200
24	1220			14	1140
				17	1110
Mean	1306	12	70		1176
sd	(57)		()		(71)

Table IV a. Wrangel Island (71°N, 178°W) 1984–85. Carbon monoxide abundance measurements in atm.cm x 100(Left column = sampling date).

Apr 84		May 84		Apr 85		May 85	
26	1100	02	1220	27	1150	01	1140
28	1240	05	1260	28	1080	03	1160
29	1260	07	1030			04	1100
		10	1100			12	900
		11	1170			15	1020
		15	1070			20	960
		16	1130				
		17	1120				
		18	1130				
		19	1060				
		20	1090				
		22	1150				
		23	980				
		26	1070				
		30	1080				
		31	1100				
Mean	1200		1110		1115		1046
sd	(71)		(67)		(35)		(79)

Table IV b. Wrangel Island (71°N, 178°W) 1987–88. Carbon monoxide abundance measurements in atm.cm x 100. (Left column = sampling date).

May 87		Jun 87		July 87		Apr 88		May 88	
13	1290	06	930	03	820	12	1790	02	1390
14	1300	09	830	05	820	16	1320	03	1350
18	1130	10	810	06	870	27	1360	04	1300
19	1200	17	960	09	900	29	1340	10	1310
		22	990	10	800			21	1040
		23	920	17	920			22	1090
		24	860	18	840				
		25	880	19	850				
		26	850	22	890				
		27	1010	23	980				
		28	1100	30	910				
		30	910	31	900				
Mean	1230		921		875		1452		1247
	(70)		(80)		(49)		(195)		(132)

Appendix 2. Monthly measurements of methane abundance from the Arctic and Antarctic for the periods covering 1977–1978 and 1982–1990

Table V. Molodezhnaya (68°S, 49°E) 1977–78. Methane abundance measurements in atm.cm x 100. (Left column = sampling date)

Dec 77		Jan 78		Feb 78		Mar 78	
15	99	05	109	01	87	03	100
16	98	06	104	02	104		
17	113	07	109	07	103		
21	124	10	98	08	109		
22	98	16	96	10	99		
23	108	23	103	11	113		
24	106	24	120	12	110		
26	120	28	103	13	105		
27	100			14	100		
28	108						
Mean	107		105		103		100
sd	(9)		(7)		(7)		( )

Table VI a. Mirny station (67°S, 93°E) 1982–83. Methane abundance measurements in atm.cm x 100. (Left column = sampling date)

Jan 82		Feb 82		Feb 83		Mar 83	
24	112	02	111	08	115	05	126
27	119	04	118	09	113		
				10	119		
				17	117		
Mean	115		114		116		126
sd	(3)		(3)		(2)		( )

Table VI b. Mirny station (67°S, 93°E) 1984–85. Methane abundance measurements in atm.cm x 100. (Left column = sampling date)

Feb 84		Mar 84		Dec 84		Jan 85		Dec 85	
02	103	06	118	06	127	05	111	07	114
10	106	08	111	07	135	07	115	20	111
16	106	09	112	08	112	08	113	21	112
21	100	10	115	14	118	09	111	23	102
27	105	11	130	15	104	10	122	26	106
29	104	13	128	16	111	17	110	28	108
		14	106	17	113	21	117	29	121
		22	100	18	112			31	101
		23	120	21	112				
		29	116	22	120				
Mean	104		116		115		114		109
sd	(2)		(9)		(8)		(4)		(6)

Table VI c. Mirny station (67°S, 93°E) 1986-8. Methane abundance measurements in atm.cm x 100. (Left column = sampling date)

Jan 86		Feb 86		Mar 86	
04	106	02	104	04	107
09	106	03	102	05	105
10	110	07	101	13	108
11	111	11	100		
12	111	12	106		
13	108				
14	121				
17	102				
20	101				
23	116				
26	109				
27	111				
Mean	109		103		107
sd	(6)		(2)		(1)

Table VI d. Mir ny station (67°S, 93°E) 1987. Methane abundance measurements in atm.cm x 100. (Left column = sampling date)

Jan		Feb		Mar		Dec	
16	110	05	108	01	107	25	122
17	116	08	117	06	109	27	114
		09	106	13	109	29	114
		15	108	20	102		
		16	112	31	109		
		22	116				
		24	113				
Mean	113		111		107		117
sd	(3)		(4)		(3)		(4)

Table VI e. Mirny station (67°S, 93°E) 1988. Methane abundance measurements in atm.cm x 100. (Left column = sampling date)

Jan		Feb		Mar		Apr	
06	120	01	111	08	115	03	116
08	133	03	114				
27	105	08	110				
28	109	10	117				
		11	114				
		12	112				
		26	100				
		27	96				
		28	120				
Mean	117		110		115		116
sd	(11)		(7)		( )		( )

Table VI f. Mirny station (67°S, 93°E) 1989–90. Methane abundance measurements in atm.cm x 100. (Left column = sampling date)

Jan 89		Feb 89		Mar 89		Feb 90		Mar 90	
31	128	01	125	01	126	23	125	15	116
		02	126	02	115	25	124	20	114
		03	115	05	130	26	118	22	116
		10	108	18	123				
		11	105						
		13	124						
		14	118						
		20	122						
		25	134						
Mean	128		121		123		122		115
sd	( )		(9)		(6)		(4)		(1)

Table VII. Zhokhov Island (76°N, 153°E) and Severnaya, Zemlya (79°N, 95°E) 1982–83. Methane abundance measurements in atm.cm x 100. (Left column = sampling date).

Zhokhov Isl. Apr 82		Severnaya Zemyla May 83	
12	121	02	99
13	107	05	94
14	116	06	98
15	117	08	102
17	122	09	102
19	119	10	102
21	115	11	100
22	124	12	100
23	117	13	91
24	117	14	103
Mean	118		99
sd	(4)		(4)

Table VII a. Wrangel Island (71°N, 178°W) 1984–86. Methane abundance measurements in atm.cm x 100. (Left column = sampling date).

Apr 84		May 84		Apr 85		May 85		Jun 86	
26	100	05	115	27	117	01	112	01	112
29	102	07	108	28	112	03	111	02	120
		11	98			04	113	05	123
		15	116			12	109	06	110
		10	101			20	120	07	126
		17	102			31	136	12	98
		18	99					13	113
		19	93					14	92
		20	101					15	102
		22	115					16	124
		23	103						
		26	99						
		30	97						
		31	97						
Mean	101		103		114		117		112
sd	(1)		(7)		(2)		(9)		(12)

Table VII b. Wrangel Island (71°N, 178°W) 1987–88. Methane abundance measurements in atm.cm x 100. (Left column = sampling date).

May 87		Jun 87		Jul 87		Apr 88		May 88	
14	138	06	117	03	104	16	124	02	128
18	130	17	117	05	104	24	116	03	114
		22	116	06	113	29	125	04	118
		28	146	10	121			22	120
				17	155				
				18	118				
				19	127				
				31	127				
Mean	134		124		121		122		120
sd	(4)		(12)		(15)		(4)		(5)