

Climate gas monitoring at the Zeppelin station
Annual report 2001

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**Climate gas monitoring at
the Zeppelin station**

**Report
856/02**

Annual report 2001



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Statlig program for forurensningsovervåking

Preface

The air monitoring station on the Zeppelin mountain, close to Ny Ålesund at Svalbard is situated in an undisturbed arctic environment, away from major pollution sources. This unique location together with the infrastructure of the scientific research community at Ny Ålesund, makes it a well suited platform for monitoring the global changes of climate gases.

The Norwegian Institute for Air Research (NILU) has a long history of research in Ny Ålesund. Since 1988, most of these activities have been connected to the research station at the Zeppelin Mountain. Monitoring of climate related factors have been one of NILU's main fields in this work.

In 1999 the Norwegian Pollution Control Authority (SFT) and NILU signed a contract commissioning NILU to run a programme for monitoring of climate gases at the station. The funding from SFT enabled NILU to broadly extend the measurement programme and associated activities, making the Zeppelin station a major contributor of data on a global as well as a regional scale.

The measurement programme includes a range of Chlorofluorocarbons (CFC), Hydrofluorocarbons (HFC), Hydrochlorofluorocarbons (HCFC), Halons as well as other halogenated organic gases, Sulphurhexafluoride (SF₆), Methane (CH₄) and Carbon monoxide (CO). The amount of particles in the air is measured by the use of an aethalometer.

The station is also basis for measurements of Carbon dioxide (CO₂) and particles performed by the Department of Meteorology at Stockholm University. These activities are funded by the Swedish Environmental Protection Agency.

Data from the monitoring activities will be processed and used as input data in the work on international agreements like the Kyoto-agreement and the Montreal-protocol.

This report summarises the activities and results of the climate monitoring programme during year 2001.

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Summary

This is an annual report describing the activities in the project *Climate gas monitoring at the Zeppelin station*, year 2001.

A brief description is given on radiative forcing caused by climate gases and aerosols. The report describes the Zeppelin monitoring station and presents some of the activities at the station, as well as current status for instruments and measurement methods used for climate gas monitoring. Results from the measurements are presented as monthly averages and plotted as daily averages.

A wide range of anthropogenic as well as natural forcing mechanisms may lead to climate change. At present the known anthropogenic forcing mechanisms include well mixed greenhouse gases (carbon dioxide, nitrous oxide, methane, SF₆ and halogenated hydrocarbons including CFC's, HFC's, HCFC's, halones and perfluorocarbons), ozone, aerosols (direct and indirect effects), water vapour and land surface albedo.

In 1999 the Norwegian Pollution Control Authority (SFT) and NILU signed a contract commissioning NILU to run a programme for monitoring of climate gases at the Zeppelin station. The funding from SFT enabled NILU to broadly extend the climate gas measurement programme and associated activities, making the Zeppelin station a major contributor of data on a global as well as a regional scale. Equipment for measurements of halogenated climate gases (CFC's, HFC's, HCFC's, halones, perfluorocarbons and a range of other halogenated hydrocarbons) was installed in the new monitoring station building year 2000 and the extended measurement programme started in the autumn the same year. The measurement programme is continuously extended, with sulphur hexafluoride (SF₆) as one of the new compounds included in 2001.

Monitoring of methane was started at the Zeppelin station by NILU in 1997. The establishment of the monitoring programme in 1999 enabled NILU to strengthen this activity by increased maintenance and required upgrades of old equipment. This has enhanced the data quality and completeness of the data series.

The measurement programme was further extended in 2001 by installation of instrumentation for monitoring carbon monoxide.

Particles have mainly been monitored by NILU as black carbon for shorter periods during measurement campaigns. NILU will extend the monitoring of particles by implementation of new instruments and extend the cooperation with other institutions performing measurements in the same field. Close contact is established with the Department of Meteorology at Stockholm University which has been monitoring carbon dioxide and particles at the Zeppelin station since 1988, funded by the Swedish Environmental Protection Agency.

The measurement programme at the Zeppelin station covers all major climate gases except nitrous oxide, one of the strongest contributors to radiative forcing.

Measurements of climate gases at the Zeppelin station can be used together with data from other remote stations for monitoring of global changes as well as for regional emissions and

tracing of emission sources. Results from the climate gas monitoring will be used for assessment of compliance with the Montreal and Kyoto protocols.

Measurements so far confirms the Zeppelin stations status as a global background station for climate gas monitoring. As the data series are expanded over time, they will make a good basis for investigations of global levels and trends.

The high frequency of data sampling makes a good basis for the investigation of polluted air transport episodes. Combined with meteorological data and measurements from other European measurement stations, this can be used for the investigation of regional emission inventories.

Table 1: Monthly mean concentration levels of climate gases at the Zeppelin station year 2001. All concentrations in pptv, except for methane (ppb) and carbon dioxide (ppm). Monthly averages for halogenated compounds are not calculated for the month of May due to instrumental problems and lack of data. Annual mean for methane is not calculated due to lack of data during the summer period

Compound	Formula	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Year
Methane	CH ₄	1840	1829	1838	1830	1821				1816	1820	1837	1829	
Carbon dioxide*	CO ₂	375	375	375	375	376	372	367	361	362	367	373	374	371
Chlorofluorocarbons														
CFC-11	CFCl ₃	268	267	264	263		263	262	261	261	262	263	261	263
CFC-12	CF ₂ Cl ₂	545	551	553	552		552	553	549	547	548	542	554	550
CFC-113	CF ₃ Cl	79	79	78	78		78	78	78	81	82	82	82	79
CFC-114	CF ₂ CICF ₂ C 	17.4	17.5	17.5	17.5		17.5	17.4	17.3	17.9	17.6	17.5	17.4	17.5
CFC-115	CF ₃ CF ₂ Cl	8.3	8.3	8.2	8.2		8.4	8.3	8.2	8.3	8.3	8.3	8.3	8.3
Hydrofluorocarbons														
HFC-125	CHF ₂ CF ₃	1.66	1.65	1.62	1.66		1.79	1.89	2.19	2.58	2.78	2.77	3.02	2.15
HFC-134a	CF ₃ CH ₂ F	20.2	21.1	21.4	21.7		20.3	20.6	21.1	22.2	22.7	23.2	23.7	21.6
HFC-152a	CH ₃ CHF ₂	3.4	3.4	3.6	3.6		2.7	2.6	2.8	3.3	3.5	3.7	4.2	3.3
Hydrochlorofluorocarbons														
HCFC-22	CHF ₂ Cl	156	157	158	158		159	160	161	162	163	165	165	161
HCFC-123	CHCl ₂ CF ₃	1.45	1.42	1.43	1.49		1.67	1.72	1.44	1.22	1.16	1.19	1.25	1.40
HCFC-124	CHFCICF ₃	1.46	1.49	1.49	1.51		1.55	1.53	1.50	1.53	1.56	1.56	1.61	1.53
HCFC-141b	CH ₃ CFCl ₂	16.2	16.3	16.2	16.2		16.8	16.9	17.0	16.9	17.6	17.8	17.6	16.9
HCFC-142b	CH ₃ CF ₂ Cl	14.7	14.8	14.7	14.7		14.8	14.9	14.9	15.0	15.1	15.1	15.5	14.9
Halons														
H-1301	CF ₃ Br	2.91	2.94	3.01	3.08		3.00	2.99	2.95	3.06	3.11	3.11	3.24	3.04
H-1211	CF ₂ ClBr	4.37	4.39	4.38	4.40		4.44	4.41	4.42	4.41	4.44	4.46	4.47	4.42
Halogenated compounds														
Methylchloride	CH ₃ Cl	517	538	548	552		497	480	455	458	474	474	534	502
Methylbromide	CH ₃ Br	9.6	9.7	10.4	10.8		9.9	9.4	9.6	8.9	9.3	9.2	9.4	9.7
Methyliodide	CH ₃ I						3.2	1.7	0.8	0.7	0.5	0.4	0.2	1.0
Methylenedichloride	CH ₂ Cl ₂	35.4	36.1	35.7	35.8		29.7	27.1	24.1	24.2	25.5	27.8	30.1	30.1
Chloroform	CHCl ₃	11.1	10.8	10.3	10.1		10.0	9.7	10.2	11.7	11.7	12.3	11.6	10.9
Methylchloroform	CH ₃ CCl ₃										36	35	33	35
Carbontetrachloride	CCl ₄	89	91	91	93		100	99	99	95	96	96	94	95
Trichloroethylene	CHCl ₃	2.05	2.09	0.73	0.36		0.19	0.22	0.22	0.58	0.52	0.36	0.87	0.75
Perchloroethylene	C ₂ Cl ₄						5.4	4.2	3.4	3.5	3.4	3.5	3.4	3.8
Sulphurhexafluoride	SF ₆	4.85	4.89	4.90	4.91		4.88	4.94	4.99	5.00	5.00	5.03	5.06	4.95

*Measurements of CO₂ performed by MISU.

1. Climate gases and aerosols

1.1 Radiative forcing

Changes in climate is caused by internal variability within the climate system and external factors, natural and anthropogenic. Their effect can be described through the effect on radiative forcing caused by each factor. Increasing concentrations of climate gases tends to increase radiative forcing, hence contributing to a warmer global surface, while some types of aerosols have the opposite effect. Natural factors such as changes in solar output or explosive volcanic activities will also influence on radiative forcing. Changes in radiative forcing, relative to preindustrial time, is indicated in Figure 1.

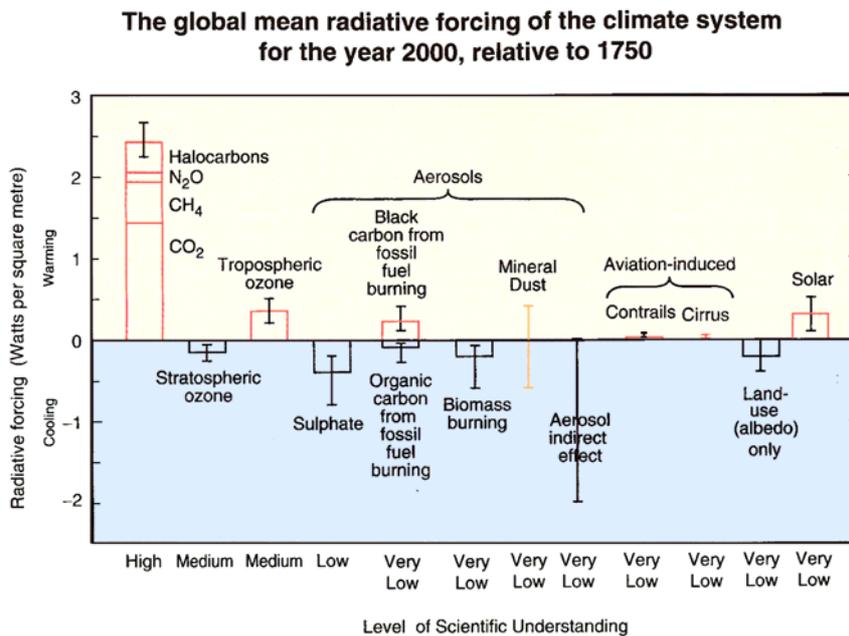


Figure 1: Known factors and their influence on radiative forcing relative to preindustrial time. The vertical lines indicates the uncertainties for each factor. (Source: IPCC.)

1.2 Natural greenhouse gases

Gases that absorb infrared radiation contribute to warming of the Earth's surface and these are called greenhouse gases. Some of these gases have large natural sources, like carbon dioxide (CO₂), methane (CH₄) and nitrous oxide (N₂O). They have sustained a stable atmospheric abundance for the centuries prior to the industrial revolution. Emissions due to human activities have caused large increases in their concentration levels over the last century (figure 2), adding to radiative forcing.

The atmospheric concentration of CO₂ has increased by 30% since 1750. The rate of increase has been about 1.5 ppm (0.4%) per year over the last two decades. About three quarters of the anthropogenic emissions to the atmosphere is due to fossil fuel burning, the rest is mainly due to land-use change, especially deforestation.

The atmospheric concentration of CH₄ has increased by 1060 ppb (150%) since 1750 and continues to increase. More than half of the current emissions are anthropogenic; use of fossil

fuel, cattle, rice plants and landfills. Carbon monoxide (CO) emissions have been identified as a cause of increasing CH₄ concentration. This is caused by CO reacting with reactive OH, thus preventing OH from reacting with CH₄, a primary loss reaction for methane (ref. Daniel, Solomon).

The atmospheric concentration of N₂O has increased by 45 ppb (17%) since 1750 and continues to increase. About a third of the emissions are anthropogenic; agriculture, cattle feed lots and chemical industry.

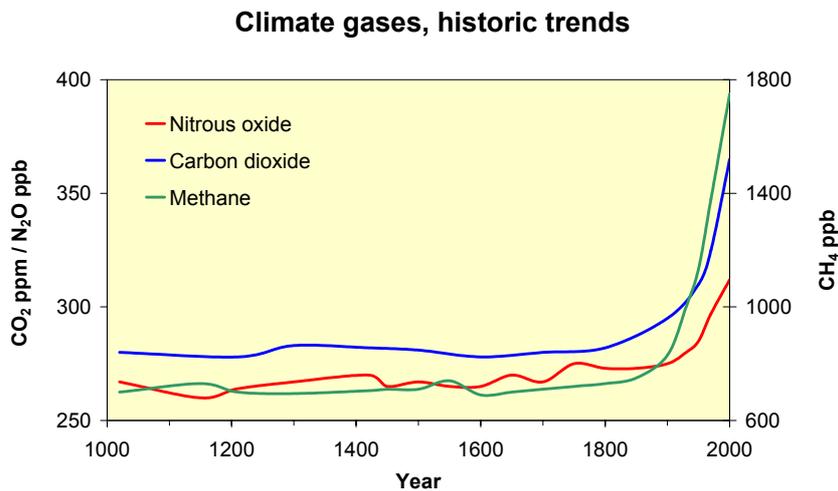


Figure 2: Changes in concentration levels over time for some natural climate gases.

Ozone (O₃) is a reactive gas with relatively large variation in concentration levels. The amount of tropospheric O₃ have increased by 35% since 1750, mainly due to anthropogenic emissions of O₃-forming gases like volatile organic compounds (VOCs), carbon monoxide (CO) and nitrogen oxides. O₃ forcing varies considerably by region and responds more quickly to changes in emissions than more longlived greenhouse gases.

Water vapour in the lower stratosphere is a effective greenhouse gas. The amount of water vapour is temperature dependent, increasing with higher temperatures. Another source of H₂O is the oxidation of CH₄ and possibly future direct injection of H₂O from high-flying aircrafts.

1.3 Synthetic greenhouse gases

Another class of gases are the man made greenhouse gases, such as CFCs, HCFCs, HFCs, PFCs, SF₆ and halons. These gases did not exist in the atmosphere before the 20th century. Although these gases have much lower concentration levels than the natural gases mentioned above, they are strong infrared absorbers, many of them with extremely long atmospheric lifetimes resulting in high global warming potentials (table 2). Some of these gases that are ozone depleting, are under the regulations of the Montreal protocol. Concentrations of these gases are increasing more slowly than before 1995, some of them are decreasing. Their substitutes however, mainly HFCs, and other synthetic greenhouse gases are currently increasing.

Table 2: Global warming potential for some selected greenhouse gases (Source IPCC).

Compound	Formula	Abundance* ppt	Trend ppt/year*	Annual emission	Lifetime, years	GWP ¹ 100-year
Non-halogenated compounds						
Carbon dioxide***	CO ₂	367	-1.5	~8 PgC		1
Methane**	CH ₄	1745	7	600 Tg	8.4/12	23
Nitrous oxide**	N ₂ O	315	0.8	16.4 TgN	120/114	296
Carbon monoxide**	CO	80	6	~2800 Tg	0.08-0.25	
Chlorofluorocarbons						
CFC-11	CFCl ₃	268	-1.4		45	4 600
CFC-12	CF ₂ Cl ₂	533	4.4		100	10 600
CFC-13	CF ₃ Cl	4	0.1		640	14 000
CFC-113	CF ₃ Cl	84	0.0		85	6 000
CFC-114	CF ₂ ClCF ₂ Cl	15	<0.5		300	9 800
CFC-115	CF ₃ CF ₂ Cl	7	0.4		1 700	7 200
Hydrofluorocarbons						
HFC-23	CHF ₃	14	0.55	~7 Gg	260	12 000
HFC-125	CHF ₂ CF ₃				29	3 400
HFC-134a	CF ₃ CH ₂ F	7.5	2.0	~25 Gg	13.8	1 300
HFC-143a	CH ₃ CF ₃				52	4 300
HFC-152a	CH ₃ CHF ₂	0.5	0.1	~4 Gg	1.4	120
Hydrochlorofluorocarbons						
HCFC-22	CHF ₂ Cl	132	5		11.9	1 700
HCFC-123	CHCl ₂ CF ₃				1.4	120
HCFC-124	CHFClCF ₃				6.1	620
HCFC-141b	CH ₃ CFCl ₂	10	2		9.3	700
HCFC-142b	CH ₃ CF ₂ Cl	11	1		19	2 400
Halons						
H-1301	CF ₃ Br	2.5	0.1		65	6 900
H-1211	CF ₂ ClBr	3.8	0.2		11	1 300
H-2402	CF ₂ BrCF ₂ Br	0.45	~0		<20	
Other halogenated compounds						
Methylchloride	CH ₃ Cl				1.3	16
Methylbromide	CH ₃ Br				0.7	5
Methyliodide	CH ₃ I					
Methylenedichloride	CH ₂ Cl ₂				0.46	10
Methylenedibromide	CH ₂ Br ₂				0.41	1
Chloroform	CHCl ₃				0.51	30
Bromoform	CHBr ₃					
Methylchloroform	CH ₃ CCl ₃	69	-14		4.8	140
Carbontetrachloride	CCl ₄	102	-1.0		35	1 800
Trichloroethylene	CHCl ₃					
Perchloroethylene	C ₂ Cl ₄					
1,2-Dichloroethane	CH ₂ ClCH ₂ Cl					
Perfluoromethane	CF ₄	80	1.0	~15 Gg	>50 000	5 700
Perfluoroethane	C ₂ F ₆	3.0	0.08	~2 Gg	10 000	11 900
Sulphurhexafluoride	SF ₆	4.2	0.24	~6 Gg	3 200	22 200

*Global mixing ratios 1998 (ppt = parts per trillion). **Concentration levels and trends in ppb and ppb/year (ppb = parts per billion).

***Concentration levels and trends in ppm and ppm/year (ppm = parts per million). ¹Global warming potential relative to CO₂.

1.4 Aerosols

Major sources of anthropogenic aerosols are fossil fuel and biomass burning. Aerosols like sulphate, biomass burning aerosols and fossil fuel organic carbon produce negative radiative forcing, while fossil fuel black carbon has a positive radiative effect. Aerosols vary considerably by region and respond quickly to changes in emissions.

Natural aerosols like sea salt, dust and sulphate and carbon aerosols from natural emissions, are expected to increase as a result of climate change. In addition to their direct radiative forcing, aerosols have an indirect radiative forcing through their effect on cloud formation.

2. The Zeppelin station

2.1 Description of the station

The monitoring station is located on the Zeppelin Mountain, close to Ny Ålesund at Svalbard. At 79° north the station is placed in an undisturbed arctic environment, away from major pollution sources. Situated 474 meters asl above the inversion layer, there is minimal influence from local pollution sources in the nearby small community of Ny Ålesund.



Figure 3: The monitoring station is located at the Zeppelin mountain.

The Zeppelin station is owned and maintained by the Norwegian Polar Institute. NILU is responsible for the scientific activities at the station. The station was built in 1989-1990. After 10 years of use, the old building was no longer sufficient for operation of advanced equipment and the increasing amount of activities. The old building was removed to give place to a new modern station that was opened in May 2000. The new monitoring station was realised by funds from the Norwegian Ministry of Environment and the Wallenberg Institution via MISU (Meteorological Institution Stockholm University).

The station building was constructed using selected materials to minimise contamination and influence on any ongoing measurements. All indoor air is ventilated away down from the mountain. The building contains several separate laboratories, some for permanent use by NILU and MISU, others intended for short term use like measurement campaigns and visiting scientists. A permanent data communication line permits on-line contact with the station for data reading and instrument control.

The unique location of the station makes it an ideal platform for the monitoring of global atmospheric change. The station is part of the EU-financed Large Scale Facility for Arctic Environmental Research, resulting in a number of visiting scientists and research groups.

The measurement activities at the Zeppelin station contributes to a number of global, regional and national monitoring networks:

- SOGE (System for Observation of halogenated Greenhouse gases in Europe)
- EMEP (European Monitoring and Evaluation Programme under "UN Economic Commission for Europe")
- Network for detection of stratospheric change (NDSC under UNEP and WMO)

- Global Atmospheric Watch (GAW under WMO)
- Arctic Monitoring and Assessment Programme (AMAP)

2.2 Activities at the station

2.2.1 NILU activities

The main goals of NILU's research activities at the Zeppelin station are:

- Studies of climate related matters and stratospheric ozone
- Exploration of atmospheric long range transport of pollutants
- Characterization of the arctic atmosphere and studies of atmospheric processes and changes

NILU performs measurements of halogenated climate gases as well as methane and carbon monoxide using automated gas chromatographs with high sampling frequencies. A mass spectrometric detector is used to determine more than 30 halogenated compounds, automatically sampled 6 times per day. Methane and CO are sampled 3 times per hour. This high sampling frequency gives valuable data for the examination of episodes caused by long range transport of pollutants as well as a good basis for the study of trends and global atmospheric change. Tight cooperation with SOGE-partners on the halocarbon instrument and audits on the methane and CO-instruments (performed by EMPA on the behalf of GAW/WMO) shows that the instruments deliver data of high quality.

The amount of particles in the air is monitored by a continuous aethalometer.

The station at Zeppelin is also used for a long range of measurements not directly related to climate gas monitoring, including daily measurements of sulphur and nitrogen compounds (SO_2 , SO_4^{2-} , $(\text{NO}_3^- + \text{HNO}_3)$ and $(\text{NH}_4^+ + \text{NH}_3)$), main compounds in precipitation, mercury, persistent organic pollutants (HCB, HCH, PCB, DDT, PAH etc.), as well as tropospheric and stratospheric ozone.

2.2.2 MISU

MISU (Department of Meteorology at Stockholm University) measure carbon dioxide (CO_2) and atmospheric particles on Zeppelin Mountain.

MISU maintains a continuous infrared CO_2 instrument on Zeppelin Mountain. The continuous data are enhanced by weekly flask sampling programmes in co-operation with other institutions. The flask data give CO_2 , ^{13}C , ^{18}O , CH_4 and CO data.

The CO_2 monitoring project at Zeppelin Station has three goals:

- provide a baseline measurement of European Arctic CO_2 concentrations
- allow detailed analysis of the processes behind CO_2 variations in the Arctic on time-scales from minutes to decades
- understand how human activities and climate change perturb the global carbon cycle and thus give variations of atmospheric CO_2 and CH_4 .

MISU has several instruments at Zeppelin Station which measure particles in the atmosphere. Aerosol particles tend to reflect light and can therefore alter Earth's radiation balance. The optical particle counter (OPC) operated by MISU gives the concentration of aerosol particles and, combined with data from the nephelometer, clues to the particles' age and origin. Size distribution is acquired from a differential mobility analyser (DMA).

Understanding atmospheric chemical processes requires more than just CO₂ and aerosols and scattering data. A total filter allows MISU to create a bi-daily record of the chemical composition of aerosol particles.

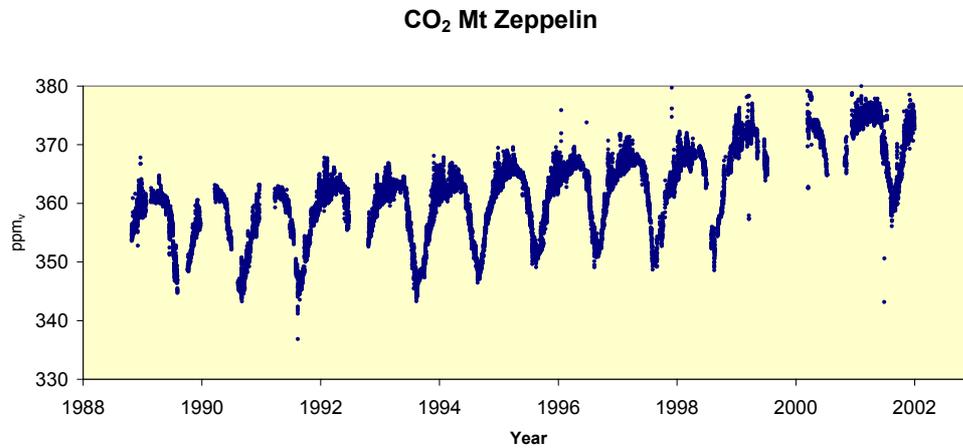


Figure 4: MISU have been measuring CO₂ at Mt Zeppelin since 1988.

2.2.3 NOAA

NOAA CMDL (The Climate Monitoring and Diagnostics Laboratory at The National Oceanic and Atmospheric Administration in USA) operates a global air sampling network. The Zeppelin station is included in this network (Figure 5).

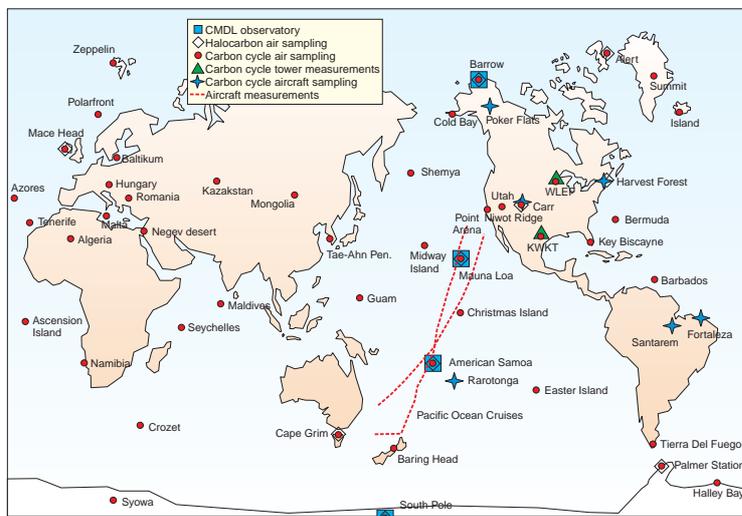


Figure 5: NOAA's global air sampling network.

Air is sampled on a weekly basis on steel canisters and sent to be analysed in the laboratories at Boulder, Colorado in USA. The measurement programme includes CH₄, CO, H₂, N₂O and SF₆. Results from the analysis are used in studies of trends, seasonal variations and global distribution of climate gases.

2.3 SOGE

SOGE is an integrated system for observation of halogenated greenhouse gases in Europe. SOGE builds on a combination of observations and modelling. High resolution in situ observation at four background stations form the backbone of SOGE. A network is being developed between the four stations. This includes full inter-calibration and common quality control which is adopted from the global monitoring network of Advanced Global Atmospheric Gases Experiment (AGAGE).

The in situ measurements will be combined with vertical column measurements, which have been made at two of the network sites for up to about 15 years, as a part of Network for Detection of Stratospheric Change (NDSC). One purpose of this combination is determination of trends in the concentrations of the gases under consideration. Integration of the observations with a variety of model tools will allow extensive and original exploitation of the data. The integrated system will be used to verify emissions of the measured substances in Europe down to a regional scale. This will be obtained by the use of a model labelling air-parcels with their location and time of origin, so it is possible to identify the various sources that contribute to the concentrations measured at the network sites. The results can be used to assess compliance with the Kyoto and Montreal protocols, and they will be utilised also to define criteria for future monitoring of halocarbons in Europe.

Global models will be used to estimate impacts of the observed compounds on climate change and the ozone layer. The impacts will be evaluated in terms of radiative forcing and Global Warming Potential (GWP), and ozone destruction and Ozone Depletion Potential (ODP), respectively.

SOGE is funded by European Commission Directorate General Research 5th Framework Programme Energy, Environment and Sustainable Development.

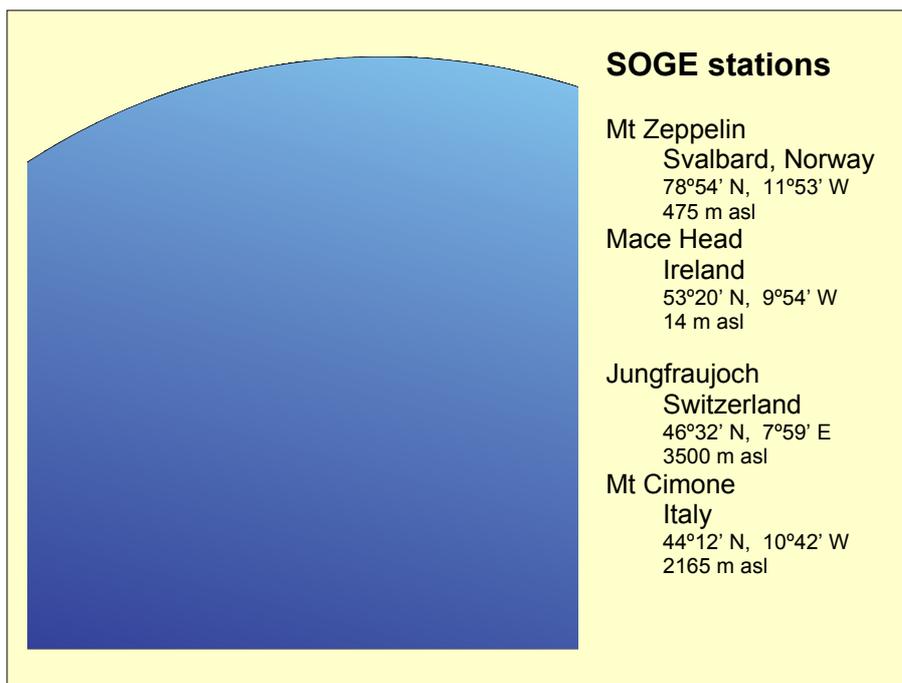


Figure 6: The SOGE climate gas monitoring stations.

3. Measurements

3.1 Instruments and methods

3.1.1 Halocarbons

Measurements of volatile halocarbons started in late spring 2000, when a specially designed instrument was installed. The instrument currently monitors more than 30 compounds, including CFCs, HFCs, HCFCs, halons and a range of other halogenated species. The gases monitored by the instrument are listed together with CH₄, CO and CO₂ in table 3.

A fully automated adsorption/desorption device is used for pre concentration of 2000 ml air over a time period of 40 minutes. The compounds are then separated and detected by the use of an automatic gas chromatograph with a mass spectrometric detector (GC-MS). The total analysis time is 2 hours. Each air sample run is followed by analysis of air from a pressurized cylinder, which is used as working standard. The four sites within the SOGE project are using working standards, which are pressurized simultaneously at Mace Head and then calibrated to AGAGE (Advanced Global Atmospheric Gases Experiment) before shipment to the individual sites. After about four months of use they are shipped back to the laboratory in Bristol for end calibration. This final calibration of the working standard is used together with the initial concentration to define the concentrations of the air samples analysed within this time period.

The instrument is remote controlled from NILU, but there is a daily inspection at the site from personnel from the Norwegian Polar Institute. There are about 4 to 6 visits from NILU each year for major maintenance work. All data are transferred to NILU on a daily basis. All data are processed by software, which is common for all AGAGE or SOGE stations. In the data network from all stations, data are accessible for each participant one day after the measurements are performed.

Instrumental problems in May/June 2001 resulted in a period of data loss. There have been short interruptions in the measurements due to periodical maintenance, but the overall data coverage is considered very good for the year 2001. Some new halogenated compounds have been included in the measurement programme this year, methyl iodide (CH₃I), methylchloroform (CH₃CCl₃) and perchloroethylene (C₂Cl₄).

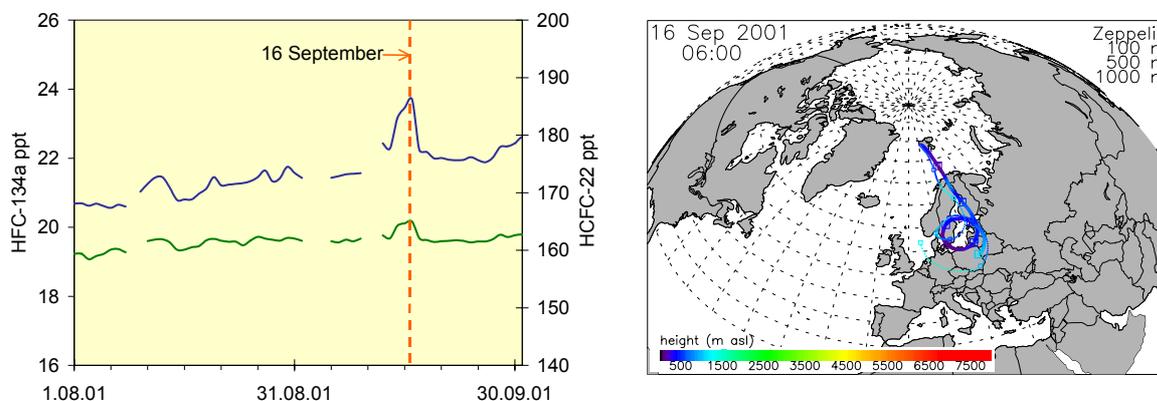


Figure 7: Measurements of HFC-134a (blue curve) and HFC-22 (green curve) showing a significant increase in concentration levels 16 September 2001. The trajectory plot indicates polluted air arriving that day is coming from the continent.

3.1.2 Methane

Methane is monitored by the use of an automatic gas chromatograph with a flame ionisation detector (GC/FID). Air is sampled three times an hour and calibrated against an air standard once an hour.

Measurements were interrupted for a three months period in the summer due to needs for maintenance and repair and late delivery of new standards. New equipment for data collection and instrument control was installed in August 2001, permitting the instrument to be controlled on-line from NILU. Data have been of some reduced quality in periods due to reduced chromatographic separation. This problem was addressed through the installation of a new chromatographic column in September 2001.

The instrument produces a large amount of data requiring a specially made system for the extensive data handling. The installation of new data collection equipment was the first step to enable the methane data being processed by the same system as the halocarbon data. This data system is specially made at the Scripps Institution of Oceanography in California, but needs an upgrade before it can include the methane measurements. The upgrade was expected to take place in September 2001, but has been delayed from the supplier. Therefore all methane data must be considered as preliminary until the new system can be taken into use.

The instrument has been running for a while against uncalibrated standards. Old standards have now been calibrated and the instrument runs with new traceable standards with references to standards used under the AGAGE programme. In September 2001 the instrument was controlled by personnel from the Swiss Federal Laboratories for Materials Testing and Research (EMPA) which is assigned by the World Meteorological Organization's (WMO) to operate the Global Atmospheric Watch (GAW) World Calibration Center for Surface Ozone, Carbon Monoxide and Methane. The results are published in EMPA-WCC report 01/3, concluding that methane measurements at Zeppelin can be considered to be traceable to the GAW reference standard. The conclusions are as follows:

“All measurements of the audited parameters (O_3 , CO , CH_4) at Ny Ålesund were performed at a high level of accuracy. The whole system from the air inlet to the instrumentation, including maintenance and data handling, is operated with great care. The staff involved in measurements and data evaluation is highly motivated and experienced. The station offers an excellent infrastructure for atmospheric research and measurement campaigns.”

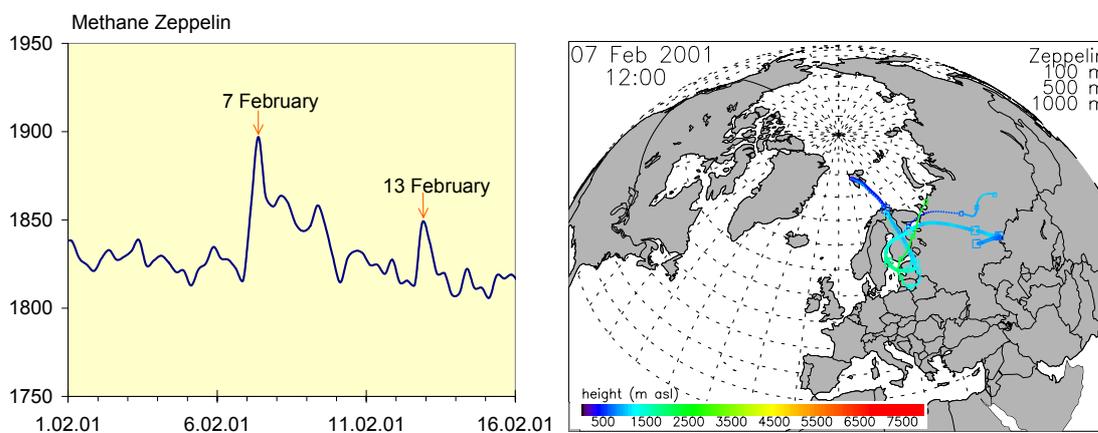


Figure 8: Trajectory plots for February 2001 indicates transport of air masses from the continent 7 February as well as 13 February. This is well reflected in the measurement data for the same period.

3.1.3 Carbon Monoxide

NILU's CO instrument was reinstalled at Zeppelin in September 2001. An international calibration during an audit from Swiss Federal Laboratories for Material Testing and Research (EMPA) was performed the same month to assess the quality of the measurements. EMPA represented the Global Atmosphere Watch (GAW) programme to include the measurements on the Zeppelin Mountain in the GAW programme. The conclusions referred to for the methane instrument (above) is also applied for the CO-instrument.

The instrument is an automatic gas chromatograph with mercury oxide reduction followed by UV detection. It has been running without interruptions since installation. The instrument is connected to the same system for data collection as the methane instrument and will use the new upgraded system from Scripps for data handling. Until the new system is upgraded, all measurements have to be considered as preliminary.

The instrument is performing analysis of 5 air samples and one standard within a time period of 2 hours. The standard is calibrated directly to the Mace Head standards, which are part of AGAGE.

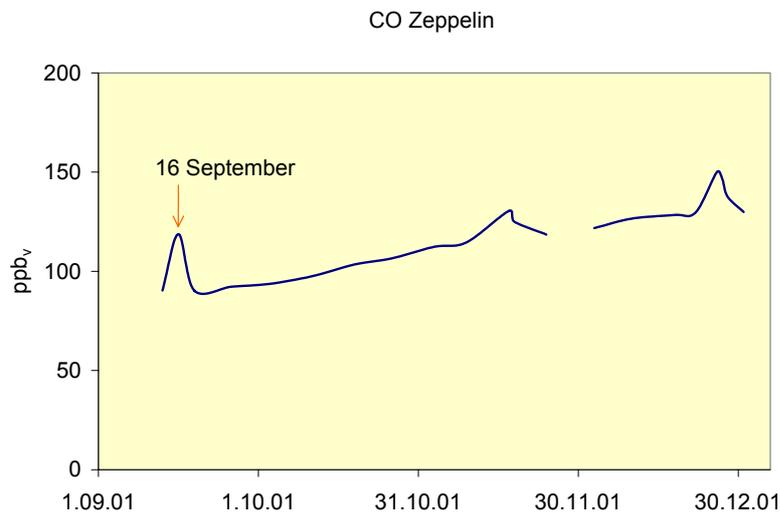


Figure 9: Plot of preliminary CO-measurement data at the Zeppelin-station. The plot shows an expected seasonal increase in concentration level. A sharp rise on 16 September fits well with the episode presented in Figure 7.

3.2 Measurements

Concentration levels for each compound monitored are plotted in Appendix 1. Monthly and annual averages are shown in table 3.

Table 3: Monthly mean concentration levels of climate gases at the Zeppelin station year 2001. All concentrations in pptv, except for methane (ppb) and carbon dioxide (ppm). Monthly averages for halogenated compounds are not calculated for the month of May due to instrumental problems and lack of data. Annual mean for methane is not calculated due to lack of data during the summer period

Compound	Formula	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Year
Methane	CH ₄	1840	1829	1838	1830	1821				1816	1820	1837	1829	
Carbon dioxide*	CO ₂	375	375	375	375	376	372	367	361	362	367	373	374	371
Chlorofluorocarbons														
CFC-11	CFCl ₃	268	267	264	263		263	262	261	261	262	263	261	263
CFC-12	CF ₂ Cl ₂	545	551	553	552		552	553	549	547	548	542	554	550
CFC-113	CF ₃ Cl	79	79	78	78		78	78	78	81	82	82	82	79
CFC-114	CF ₂ CICF ₂ C 	17.4	17.5	17.5	17.5		17.5	17.4	17.3	17.9	17.6	17.5	17.4	17.5
CFC-115	CF ₃ CF ₂ Cl	8.3	8.3	8.2	8.2		8.4	8.3	8.2	8.3	8.3	8.3	8.3	8.3
Hydrofluorocarbons														
HFC-125	CHF ₂ CF ₃	1.66	1.65	1.62	1.66		1.79	1.89	2.19	2.58	2.78	2.77	3.02	2.15
HFC-134a	CF ₃ CH ₂ F	20.2	21.1	21.4	21.7		20.3	20.6	21.1	22.2	22.7	23.2	23.7	21.6
HFC-152a	CH ₃ CHF ₂	3.4	3.4	3.6	3.6		2.7	2.6	2.8	3.3	3.5	3.7	4.2	3.3
Hydrochlorofluorocarbons														
HCFC-22	CHF ₂ Cl	156	157	158	158		159	160	161	162	163	165	165	161
HCFC-123	CHCl ₂ CF ₃	1.45	1.42	1.43	1.49		1.67	1.72	1.44	1.22	1.16	1.19	1.25	1.40
HCFC-124	CHFCICF ₃	1.46	1.49	1.49	1.51		1.55	1.53	1.50	1.53	1.56	1.56	1.61	1.53
HCFC-141b	CH ₃ CFCl ₂	16.2	16.3	16.2	16.2		16.8	16.9	17.0	16.9	17.6	17.8	17.6	16.9
HCFC-142b	CH ₃ CF ₂ Cl	14.7	14.8	14.7	14.7		14.8	14.9	14.9	15.0	15.1	15.1	15.5	14.9
Halons														
H-1301	CF ₃ Br	2.91	2.94	3.01	3.08		3.00	2.99	2.95	3.06	3.11	3.11	3.24	3.04
H-1211	CF ₂ ClBr	4.37	4.39	4.38	4.40		4.44	4.41	4.42	4.41	4.44	4.46	4.47	4.42
Halogenated compounds														
Methylchloride	CH ₃ Cl	517	538	548	552		497	480	455	458	474	474	534	502
Methylbromide	CH ₃ Br	9.6	9.7	10.4	10.8		9.9	9.4	9.6	8.9	9.3	9.2	9.4	9.7
Methyliodide	CH ₃ I						3.2	1.7	0.8	0.7	0.5	0.4	0.2	1.0
Methylenedichloride	CH ₂ Cl ₂	35.4	36.1	35.7	35.8		29.7	27.1	24.1	24.2	25.5	27.8	30.1	30.1
Chloroform	CHCl ₃	11.1	10.8	10.3	10.1		10.0	9.7	10.2	11.7	11.7	12.3	11.6	10.9
Methylchloroform	CH ₃ CCl ₃										36	35	33	35
Carbontetrachloride	CCl ₄	89	91	91	93		100	99	99	95	96	96	94	95
Trichloroethylene	CHCl ₃	2.05	2.09	0.73	0.36		0.19	0.22	0.22	0.58	0.52	0.36	0.87	0.75
Perchloroethylene	C ₂ Cl ₄						5.4	4.2	3.4	3.5	3.4	3.5	3.4	3.8
Sulphurhexafluoride	SF ₆	4.85	4.89	4.90	4.91		4.88	4.94	4.99	5.00	5.00	5.03	5.06	4.95

*Measurements of CO₂ performed by MISU.

4. Method for assessment of compliance with the Montreal and Kyoto protocols

One objective of the climate gas monitoring is to model the transport of halocarbons from areas of emissions to the observational site in Ny-Ålesund and to contribute to validated emission inventories for the halocarbons measured in the program. These results can be used by regulatory bodies to evaluate emission estimates of the respective substances.

As part of the EU funded project SOGE (System for observation of halogenated greenhouse gases in Europe) best estimates for emissions of several target species will be derived from databases maintained by industry, regulatory bodies and publications. Ideally, the high temporal resolution *in situ* data from the Zeppelin station will be used in combination with a dispersion model to trace the origin of the observed compounds. This will allow an evaluation of the derived emissions and comparison with emission inventories provided by European countries.

Such an integrated observation/modelling system has already proven reliable and useful for a few selected compounds measured at Mace Head in Ireland, and offers a unique opportunity to monitor the compliance with the protocols. However, the observations made at the Zeppelin so far reveal that the episodes of elevated halocarbon concentrations are fewer than at Mace Head, and also the peaks are weaker, indicating a larger degree of dispersion from the source regions to the station. The data from the Zeppelin station can be regarded as supplementary to data from other stations closer to the emissions. The connection between the Zeppelin station and the other stations in the SOGE network is thus crucial for the purpose of emission verification.

Previous modelling studies with the model that will be used in the monitoring program have focussed on the long range transport to Mace Head, Ireland and the model has provided a detailed analysis of long-range transport from North America to Europe. Furthermore, the dispersion model has quantified the European emission source strengths of approaching 20 individual halocarbons and greenhouse gases required to support the observations made at Mace Head, Ireland.

The model concentrations will be compared with the observations taken at 4-hour intervals at the Zeppelin station providing continuous *in situ* data. For statistical comparison, model values will be linearly interpolated in time to correspond with the observations. The comparisons will be made with the short-term fluctuations observed above background levels (see Figure 10), which represent years of emissions since industrial manufacture began. This procedure has proven to yield good agreement between observations and model results. A high correlation has been found between observed and model traces, e.g. for CFC-11 as shown in Figure 10.

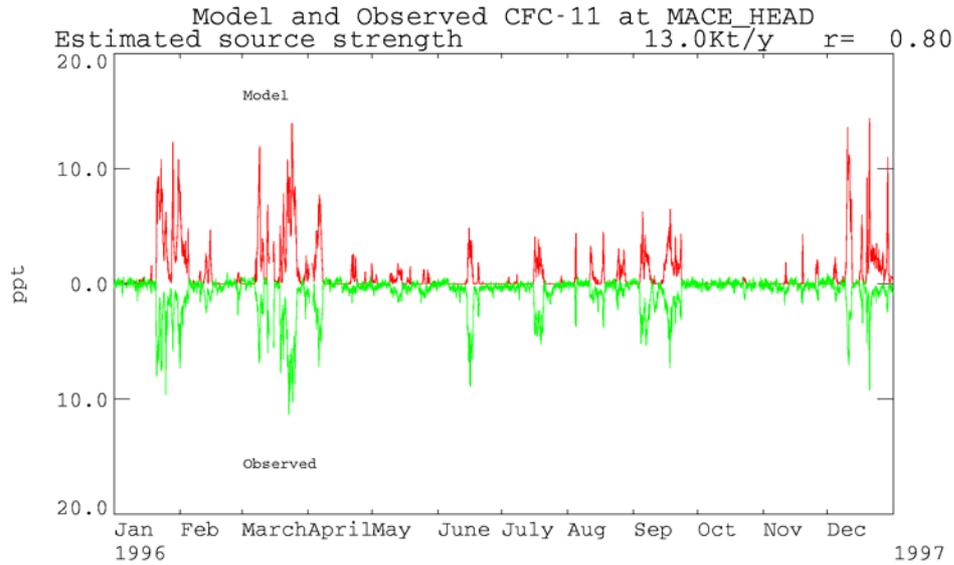


Figure 10: Observations of CFC-11 at Mace Head and estimates by a Lagrangian dispersion model that will be used in this project.

The dispersion model is of a Lagrangian type, in which emissions are modelled by releasing large number of ‘particles’ into the ‘model atmosphere’. The particles are carried along passively by the ambient three-dimensional wind, with turbulent diffusion simulated by random walk or Wiener techniques. Each particle represents a mass of released pollutant. All meteorological data, such as wind and temperature fields, are taken from a numerical weather prediction model, at six-, three- and one-hour intervals, with horizontal resolution of about 90, 50 and 16 km, respectively. A nested structure is used. The model has successfully been used to simulate dispersion of a wide range of trace gases.

The main aim of the model studies with the dispersion model is to establish the particular sources that contribute to the concentrations at given receptors. In the model particles are labelled with their location and time of origin, so it is possible to identify the various sources that contribute to a defined receptor and quantify their relative contributions.

5. Radiative forcing in the high northern latitude atmosphere

A wide range of anthropogenic as well as natural forcing mechanisms may lead to climate change. As a starting point for estimation of radiative forcing in the climate gas monitoring programme we have performed calculations of the radiative forcing due to anthropogenic emissions leading to changes in the concentrations of several trace gases and aerosols. Increase in all major well-mixed greenhouse gases are included as well as changes in tropospheric and stratospheric ozone. The radiative forcing due to certain aerosols is also estimated. The calculations have been performed for the Northern Hemisphere, and the main focus in this report is on high latitudes.

Several climate forcing mechanisms may lead to climate change. At present the known anthropogenic forcing mechanisms include well-mixed greenhouse gases, ozone, aerosols (direct and indirect effects), and land surface albedo. We include a short description of the radiative transfer models, which have been used. Usually the radiative forcing is seen in a global context. However, in this report we discuss matters of particular interest for high latitudes in the Northern Hemisphere, to relate them to the measurements of climate gases in Ny Ålesund. We present results for each of the individual contributions to the radiative forcing.

The radiative forcing is calculated using radiative transfer models. We use an emissivity/absorptivity broadband model for thermal infrared radiation. The model includes about 50 absorption bands including all the major greenhouse gases. Clouds are included in the model. For solar radiation a multi-stream model using the discrete ordinate method is used. Absorption by O₂, O₃, CO₂, and H₂O, Rayleigh scattering, aerosols, and clouds are taken into account. The spectral resolution varies from 1 nm to dividing the spectral interval into 70 regions.

Table 4 shows estimates of all radiative forcings since industrialization (from 1750 to 1995). The numbers are global and seasonal averages. The global distribution of the radiative forcing due to all the anthropogenic contributions discussed in the following is shown in Figure 11 for seasonally averaged conditions.

We have previously established formulas for global radiative forcing as a function of atmospheric concentrations for the well-mixed greenhouse gases based on calculations with the models described above. We have applied these functions along with changes in the global concentrations of well-mixed greenhouse gases to establish the radiative forcing in 1995. CO₂, CH₄, N₂O and a group of halocarbons yield a forcing of 2.30 Wm⁻², out of which CO₂ contributes with 1.38 Wm⁻². There is a marked latitudinal gradient in the forcing, with largest values in the tropics, due to the large temperature between the warm surface and the cold tropopause region. This pattern can clearly be seen in Figure 11. The forcing at high latitudes in the Northern Hemisphere is about 70% of the global average.

In a previous study we have investigated the evolution of tropospheric ozone and its radiative forcing from 1850 to 1995, due to emissions of ozone precursors (NO_x, CO, CH₄ and VOC). Regional differences in the radiative forcing due to tropospheric ozone are marked, with maxima at longitudes near the regions of maximum emissions of the ozone precursors. The maxima in radiative forcing, which are displaced towards lower latitudes, e.g. due to the larger difference in the vertical temperature gradient, as for the well-mixed greenhouse gases,

can also be seen in Figure 11. Consequently, the radiative forcing is weak at high northern latitudes. The global radiative forcing is 0.38 Wm^{-2} .

In our calculation of the radiative forcing due to change in stratospheric ozone, we have adopted zonal mean stratospheric ozone changes based on satellite observations and ozone sonde measurements in the Polar Regions. Long wave and short wave calculations are performed with the models described above. Strong radiative forcing is found at northern high latitudes, with values between -0.3 Wm^{-2} and -0.4 Wm^{-2} . In the tropical region the radiative forcing is close to zero. The global and annual mean radiative forcing due to change in stratospheric ozone is calculated to -0.15 Wm^{-2} . The forcing due to changes in stratospheric ozone is uncertain, as the vertical distribution of the ozone loss in the stratosphere is uncertain, especially in the lower stratosphere where it has a strong impact on the forcing. Our result is more negative than the bulk of other calculations, most of which give a slightly positive forcing as a global average.

Several different aerosol components are of climatic importance, and we have included the direct effect of two types of aerosols in this report, namely sulphate and soot. These are important components of the Arctic ambient particulate matter, e.g. in Arctic haze. We have previously calculated the value -0.36 Wm^{-2} for the 1995 forcing due to sulphate. Several recent estimates of the forcing due to sulphate are performed, some in line with this estimate and some with stronger radiative forcing. Uncertainties in representation of hygroscopic effects of the sulphate aerosols and forcing in cloudy areas are major reasons for the large differences found. Likewise, we have estimated a soot forcing of 0.14 Wm^{-2} . This estimate is slightly lower than other estimates. Large uncertainties for this aerosol component also exist, mostly related to the mass and the distribution of the soot, optical properties, and mixing with other types of aerosols.

Thus, large uncertainties also exist for the forcing at high northern latitudes. In general, the radiative forcing due to the largely scattering sulphate aerosol is reduced at very high latitudes due to the high surface albedo in the areas covered with ice and snow. Conversely, the radiative forcing due to the absorbing soot aerosols is enhanced in the same region.

There are several features which make radiative forcing values at high northern latitudes deviate from the global averages. The well-mixed greenhouse gases are of somewhat less importance due to the lower vertical gradient between the surface and the tropopause. This is the case also for tropospheric ozone, in this case also because of the distance to the regions of emissions of ozone precursors.

Stratospheric ozone changes, on the other hand, maximize at high latitudes and in particular in the Arctic, so the radiative forcing due to depletion of the ozone layer is largest there. Whereas the well-mixed greenhouse gases have increased in atmospheric burden since the time of industrialization, the most significant ozone depletion has taken place over only about two decades. Consequently, at high northern latitudes, the heating due to well-mixed greenhouse gases has been nearly balanced by the cooling due to reductions in stratospheric ozone over the last two decades.

Aerosols also play a role in the radiative balance at high northern latitudes, in the so-called Arctic haze events. The uncertainties in the radiative forcing estimates are large, but it is clear that the effect of scattering sulphate aerosols is reduced and the effect of the absorbing soot

aerosol is enhanced relative to the globally averaged conditions due to the high local surface albedo.

In the Arctic there is a large seasonal variation in the radiative forcing. This is most noticeable for the forcing due to Arctic haze and changes in stratospheric ozone. The Arctic haze maximizes during the winter and spring. The radiative effect is largest in the visible wavelengths. Consequently it peaks during spring after the winter time polar night. Likewise, the radiative effects of changes in stratospheric ozone maximize during spring, which is the time of the strongest ozone depletion, which also depends on solar radiation.

Table 4: Global-mean radiative forcing in 1995 since industrialization.

Species	Radiative forcing (Wm^{-2})
CO ₂	+1.38
CH ₄	+0.47
N ₂ O	+0.12
CFCs	+0.33
<i>Total for well mixed greenhouse gases</i>	<i>+2.30</i>
Stratospheric ozone	-0.15
Tropospheric ozone	+0.38
<i>Total for ozone</i>	<i>+0.23</i>
Sulphate	-0.36
Soot	+0.14
<i>Total for sulphate and soot aerosols (external mixing)</i>	<i>-0.22</i>

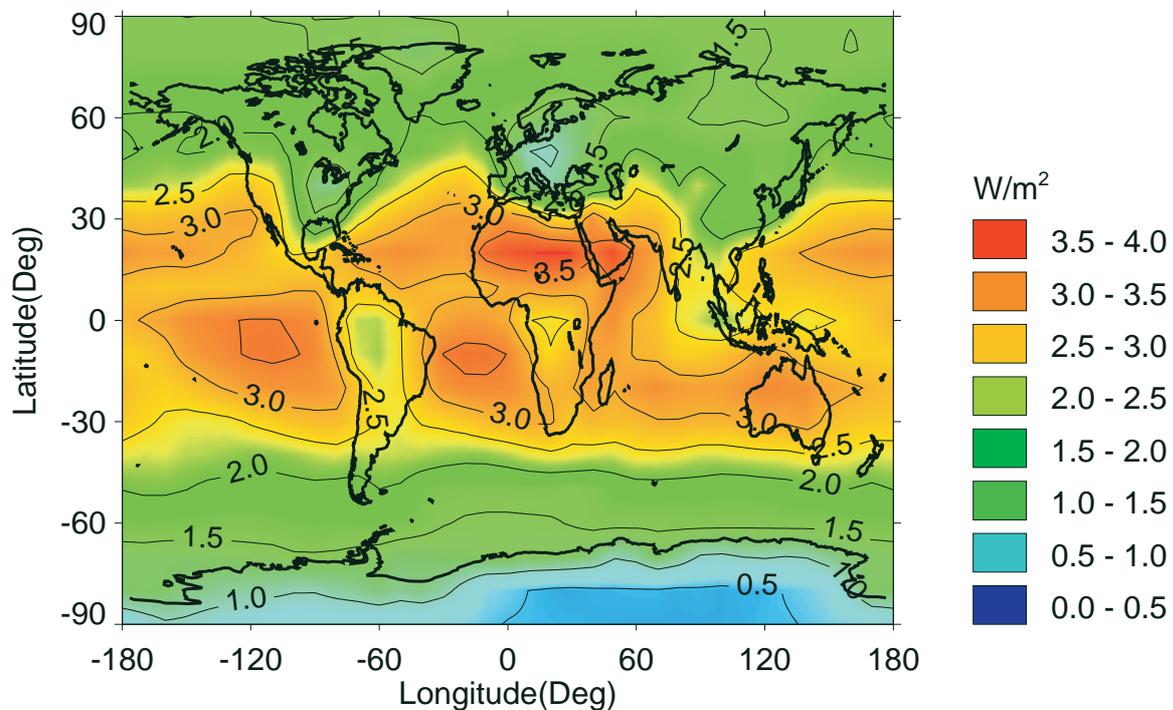


Figure 11: Radiative forcing due to anthropogenic activity (in Wm^{-2}). The results are for changes in well mixed greenhouse gases, tropospheric and stratospheric ozone, sulphate and soot aerosols from pre-industrial to 1995 conditions.

6. References

Houghton, J.T., Ding, Y., Griggs, D.J., Noguer, M., van der Linden, P.J., Dai, X., Maskell, K. and Johnson, C.A. (eds.) (2001) *Climate Change 2001: The Scientific basis*. Cambridge, Cambridge University Press.

Daniel, J.S. and Solomon, S. (1998) On the climate forcing of carbon monoxide. *J. Geophys. Res.*, *103*, 13 249-13 260.

Zellweger, C., Buchmann, B., Klausen, J. and Hofer, P. (2001) System and performance audit for surface ozone, carbon monoxide and methane, global GAW station Zeppelin Mountain Ny-Ålesund, Svalbard, September 2001 (EMPA-WCC report 01/3).

Appendix A

Measurement results

Comments to the measurement results

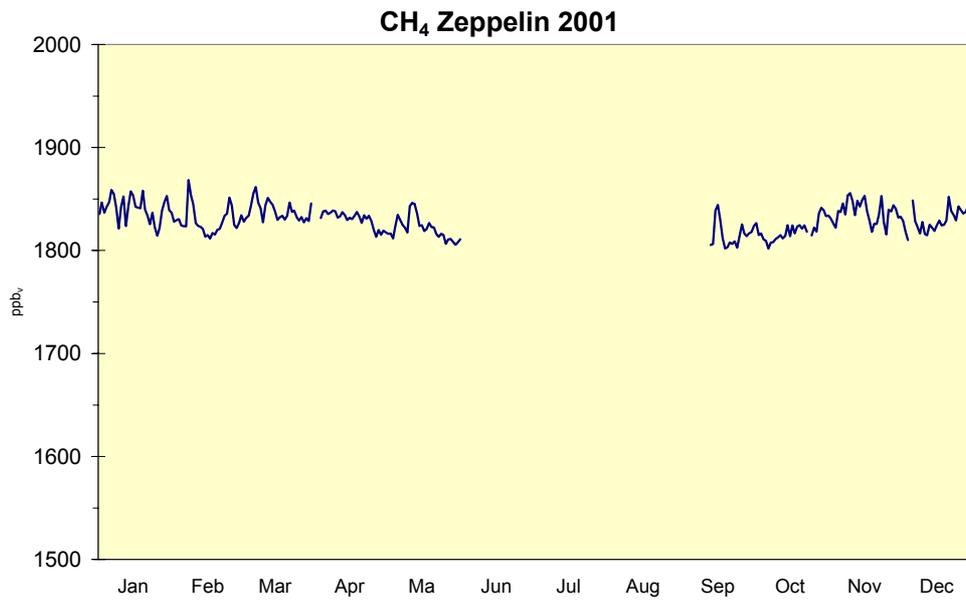
The appendix presents plots of daily averages for methane (CH₄), carbon dioxide (CO₂), chlorofluorocarbons (CFC), hydrofluorocarbons (HFC), hydrochlorofluorocarbons (HCFC), halons, other halogenated hydrocarbons and SF₆.

Trends of each compound will be further investigated as the time series are expanded. Some annual variations due to seasonal changes in photochemical activities and emissions can be seen for the more reactive compounds like CH₃Cl and CO₂.

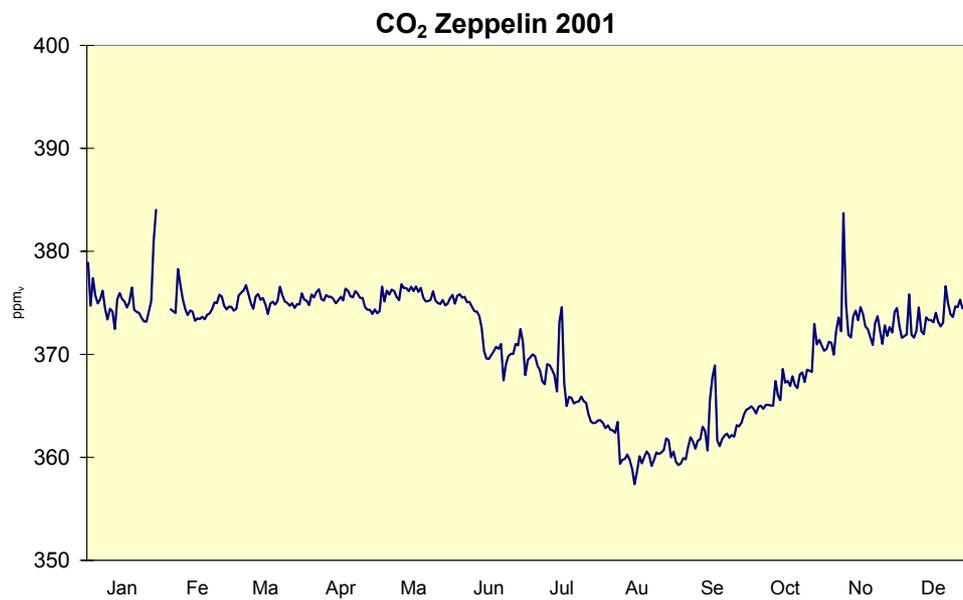
There are some wholes in the data series due to instrument stops for maintenance work. Methane has a big gap during the summer months when the instrument was running without calibration standard. All halogenated compounds have a gap from the middle of April to the beginning of June due to instrumental problems.

Some sudden jumps in concentration levels when substituting calibration standards indicates a calibration problem for some compounds (i.e. CFC-12 and CFC-113). Routines for calibration are still under development.

Methane



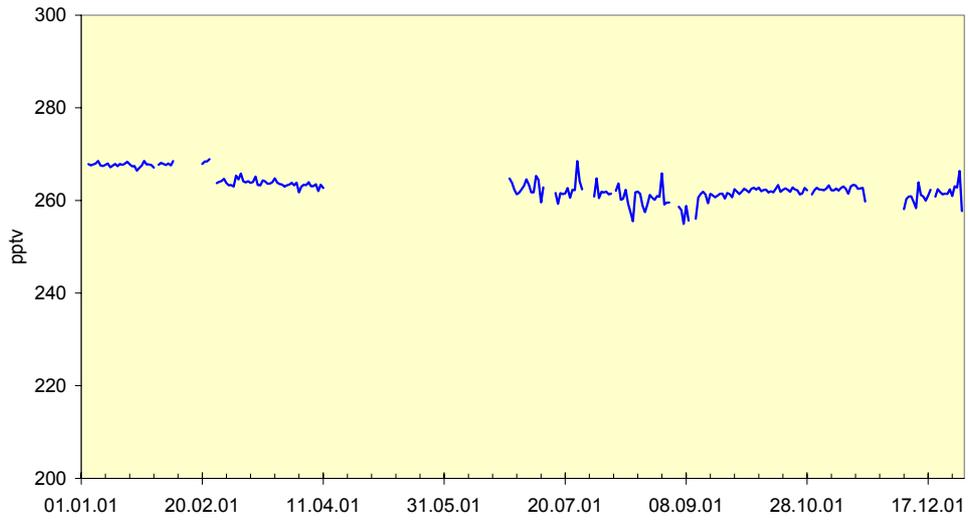
Carbon dioxide



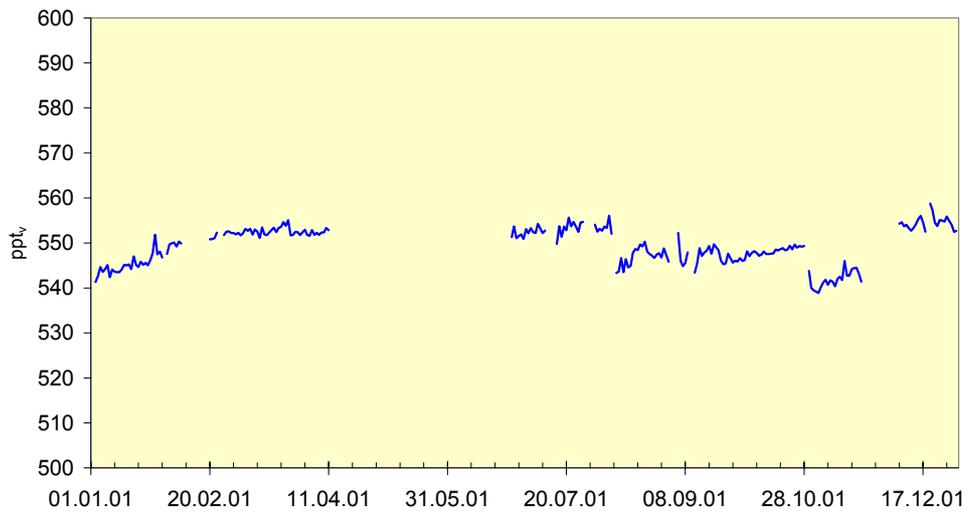
Measurements of CO₂ are performed by the Department of Meteorology, Stockholm University (MISU).

Chlorofluorocarbons (CFC)

CFC-11



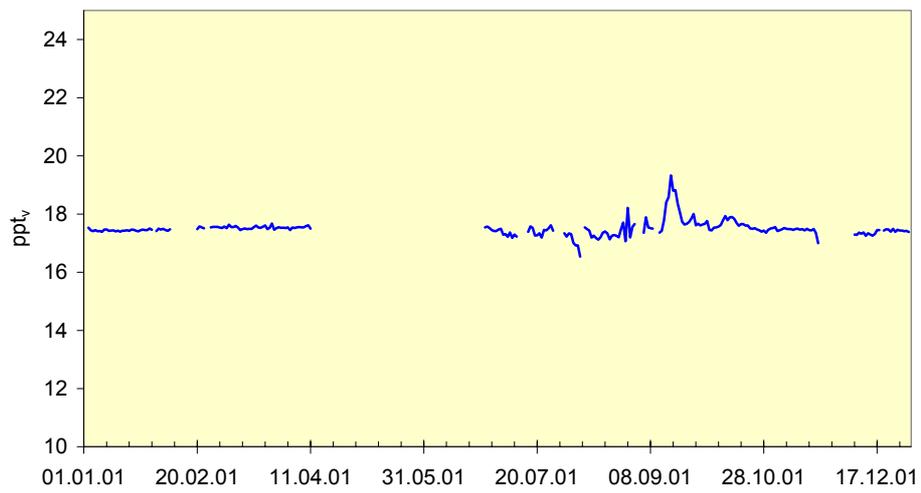
CFC-12



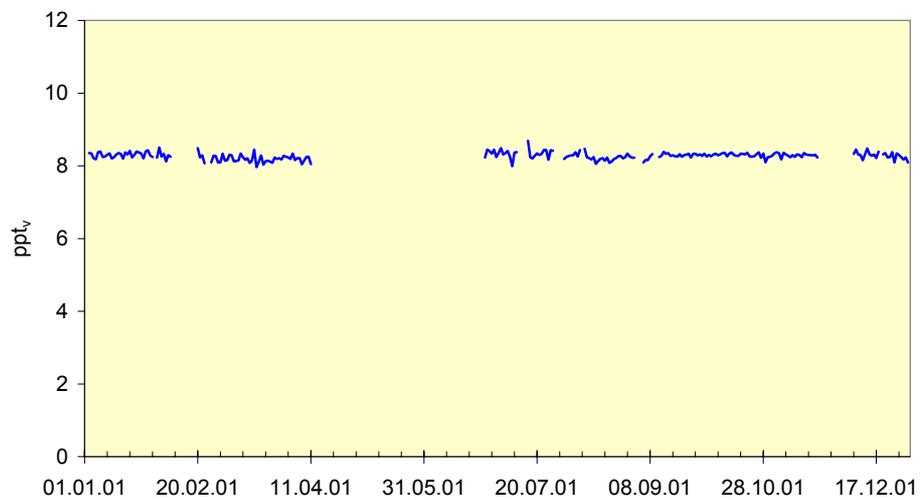
CFC-113



CFC-114

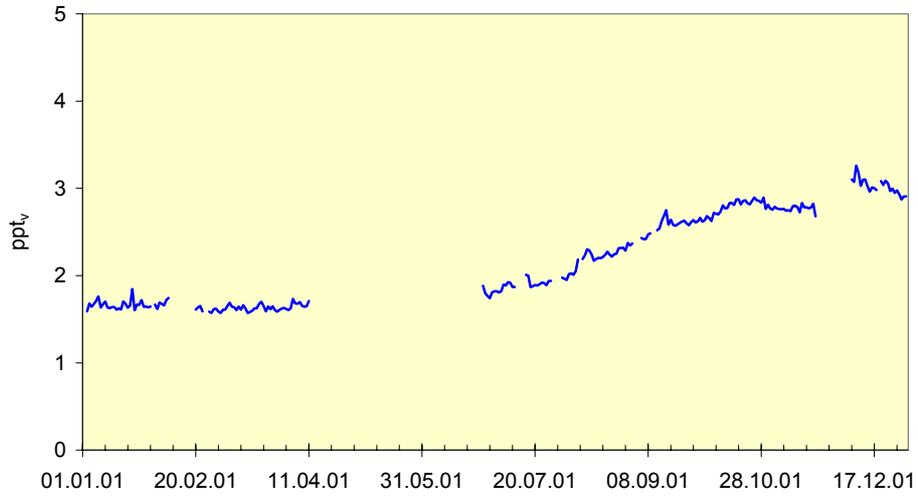


CFC-115

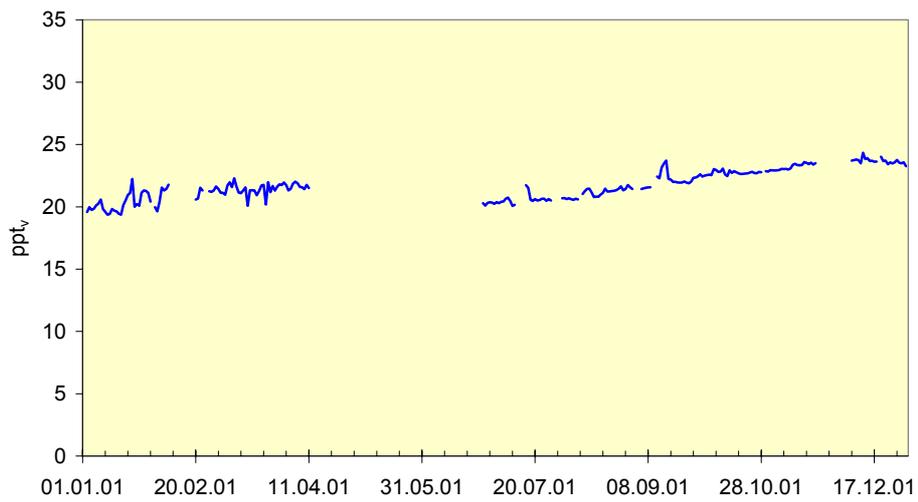


Hydrofluorocarbons (HFC)

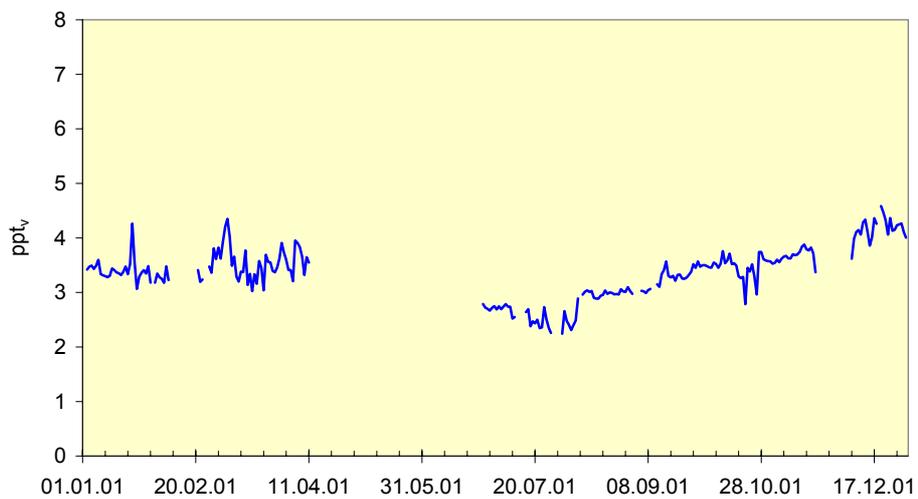
HFC-125



HFC-134a

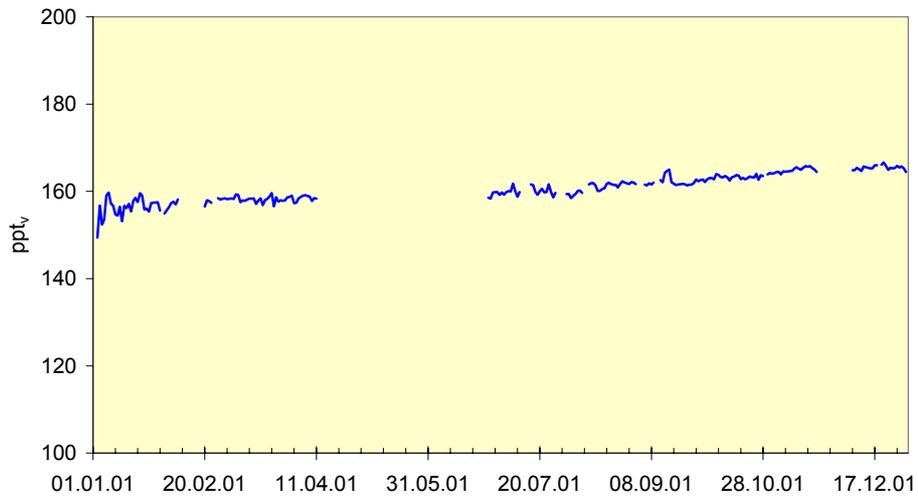


HFC-152a

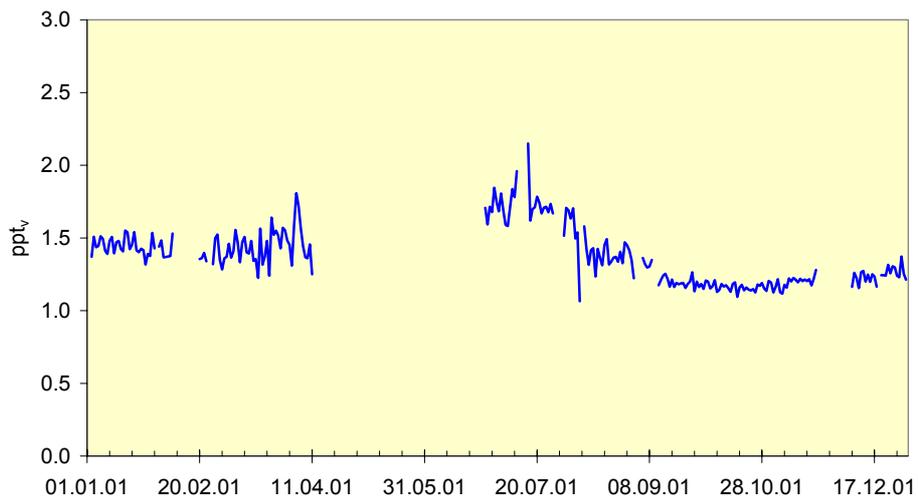


Hydrochlorofluorocarbons

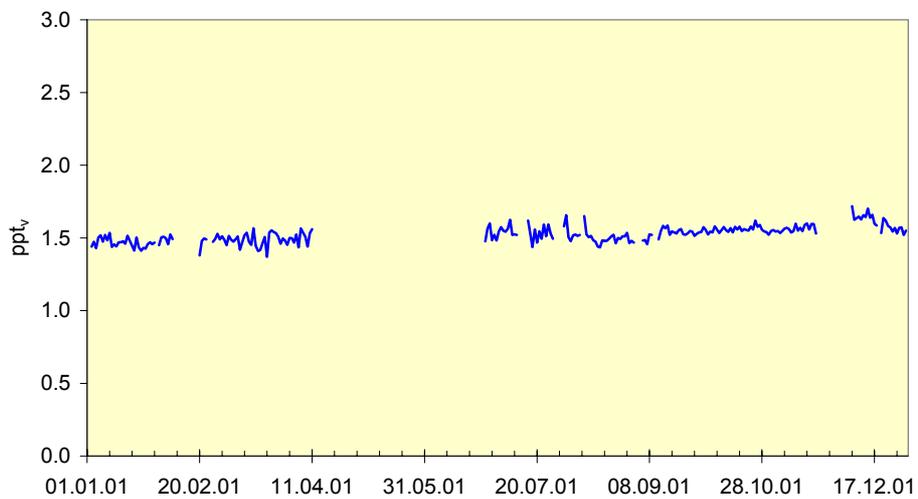
HCFC-22



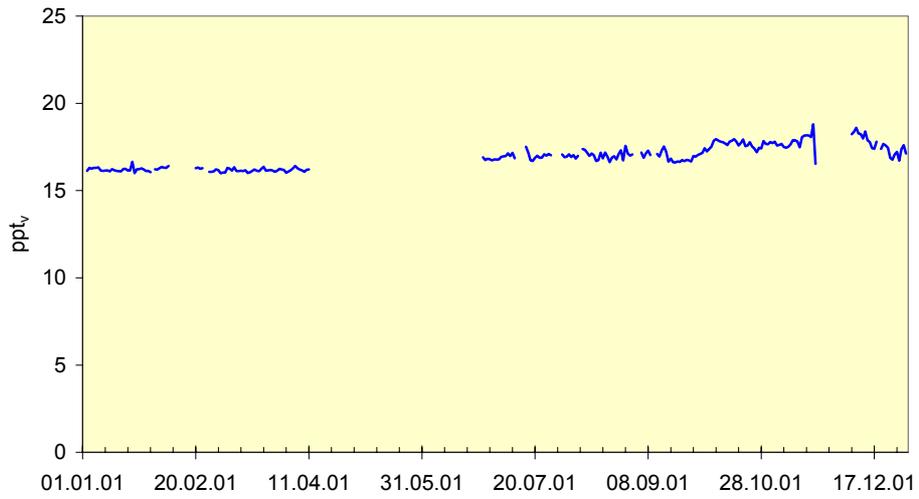
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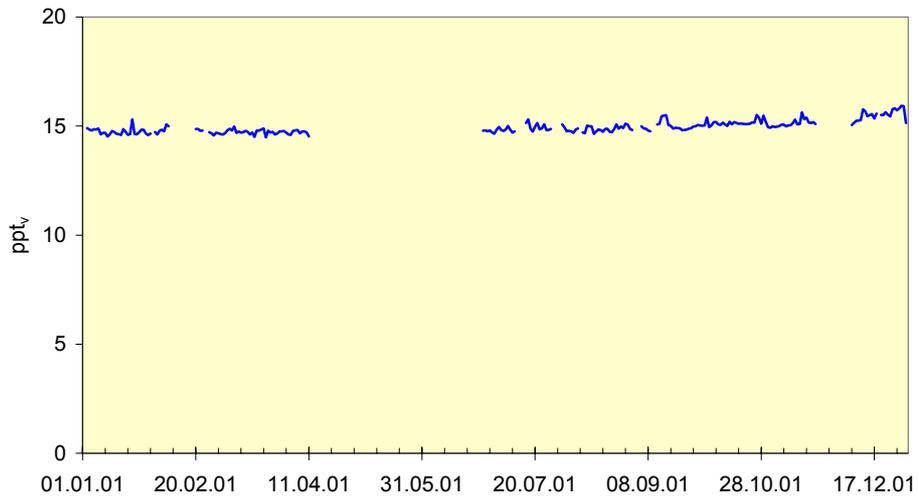
HCFC-124



HCFC-141b

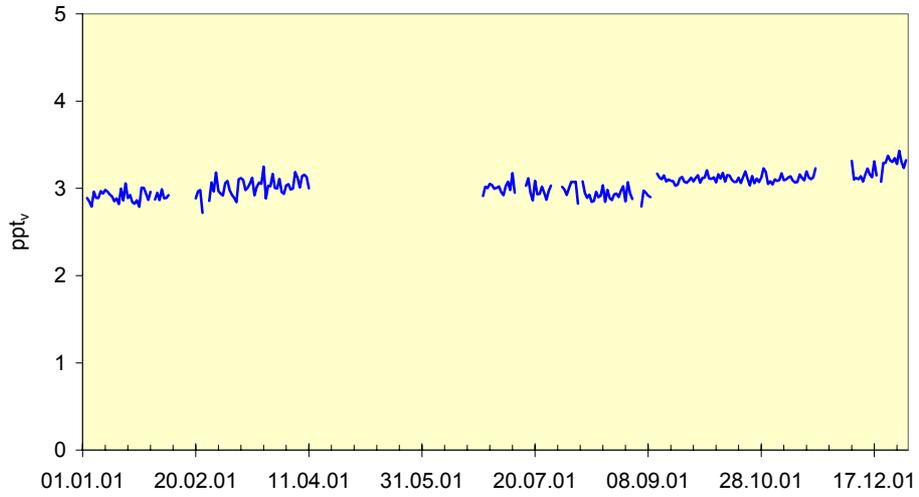


HCFC-142b

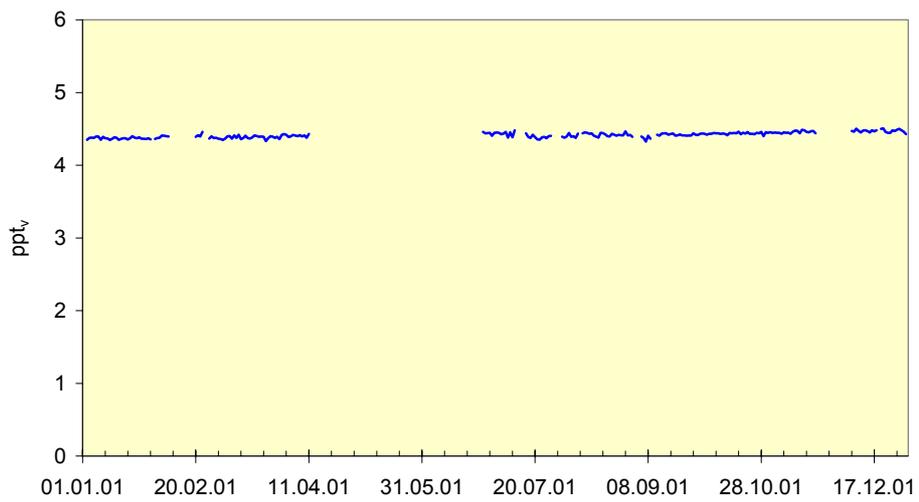


Halons

H-1301

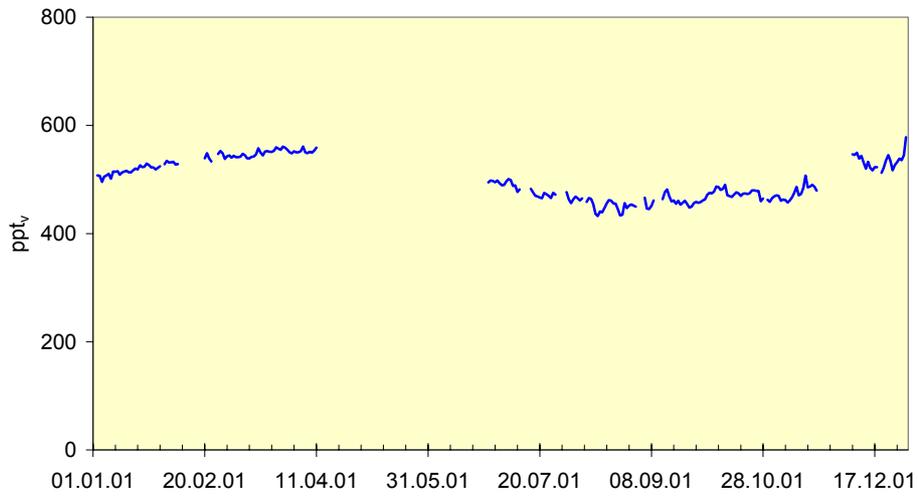


H-1211

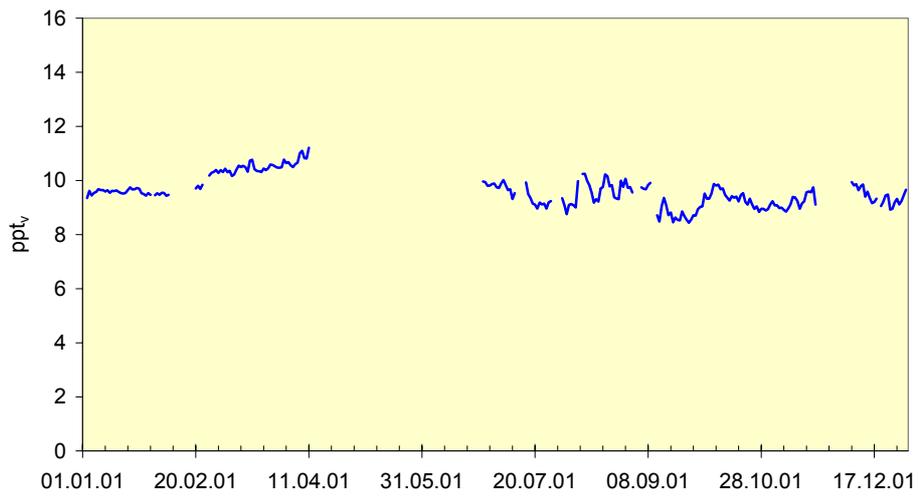


Halogenated hydrocarbons

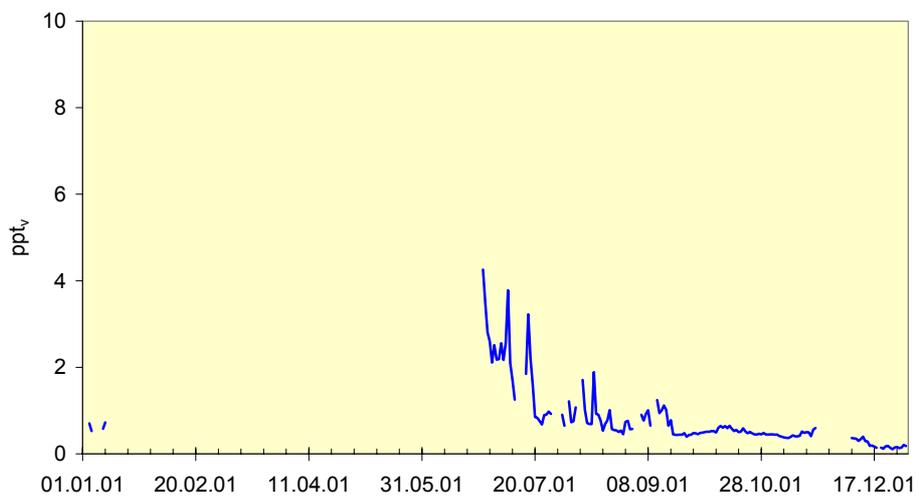
CH₃Cl



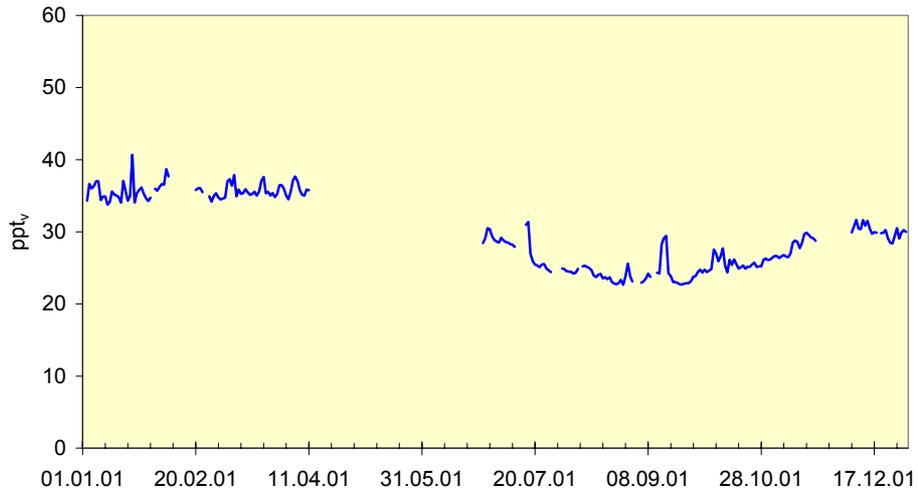
CH₃Br



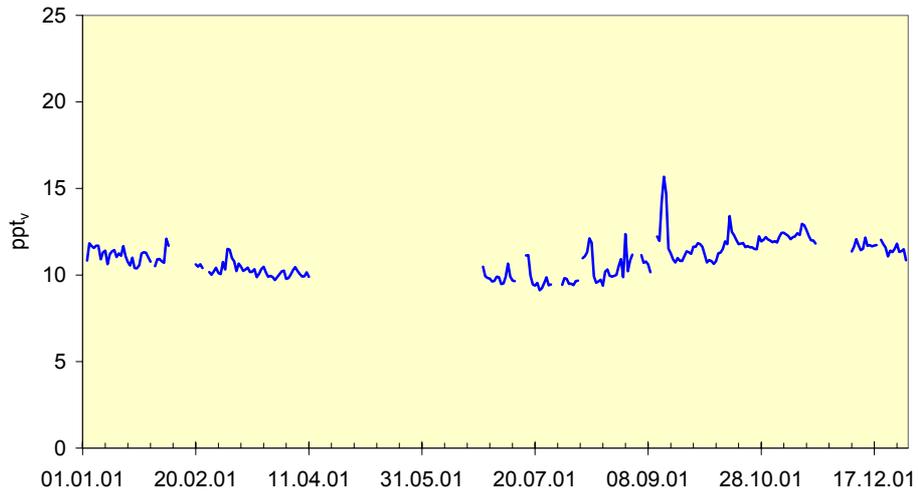
CH₃I



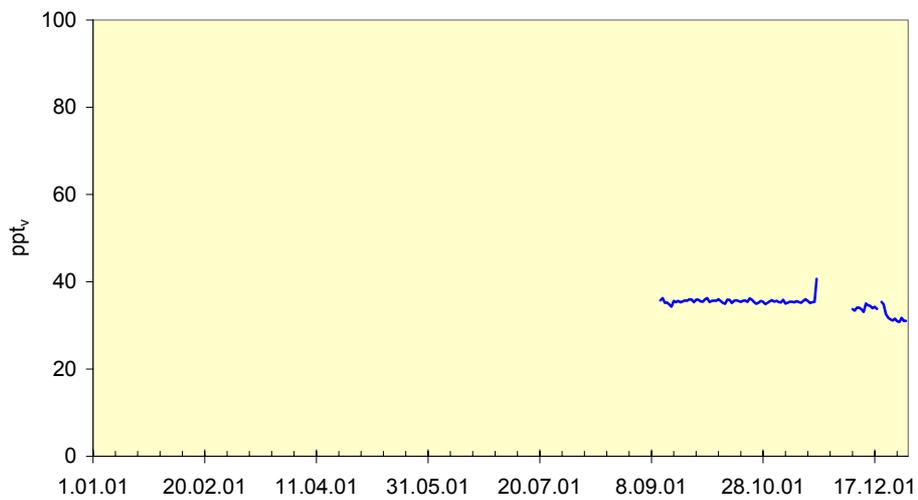
CH₂Cl₂



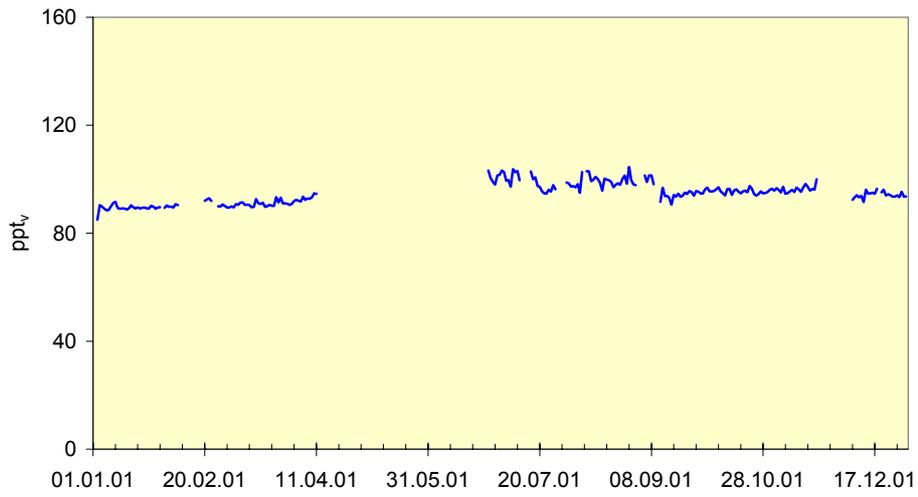
CHCl₃



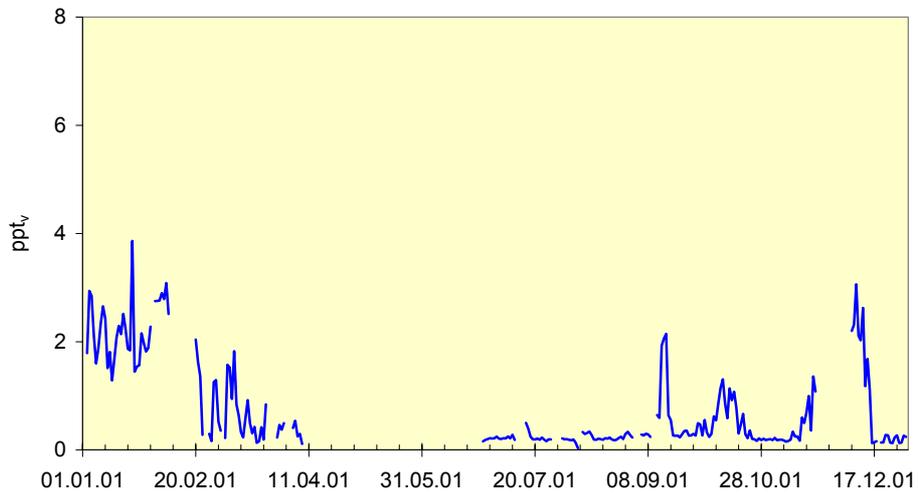
Methylchloroform



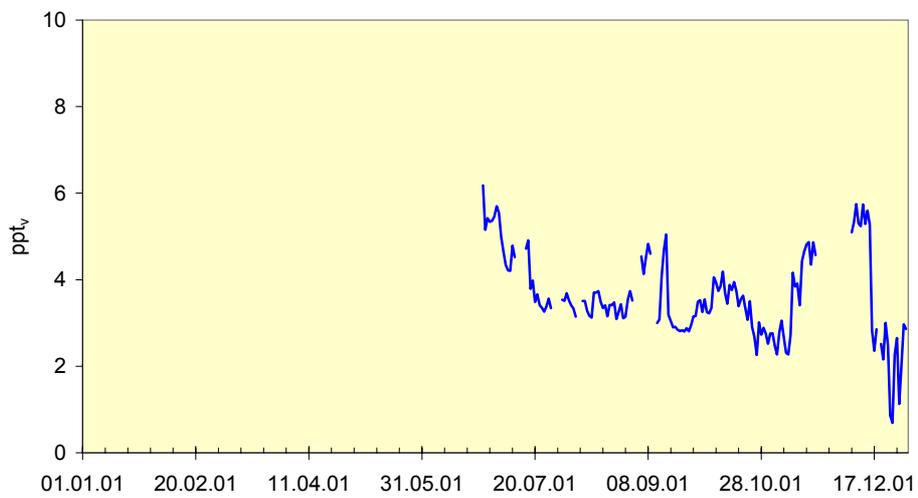
CCl₄



CHCl₃

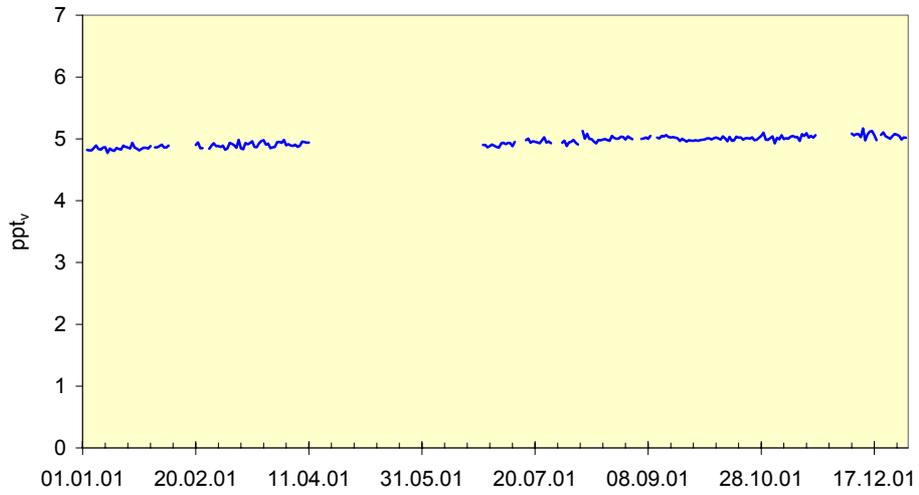


C₂Cl₄



Sulphurhexafluoride

SF₆





Statens forurensningstilsyn
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