The Air Chemistry Observatory at Neumayer Station (Antartica)

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Abstract: The Air Chemistry Observatory at the German Antarctic station Neumayer (formerly Georg von Neumayer) is in continuous operation since nearly 25 years. It thus provided unique records of atmospheric trace constituents of climatic relevance. The focus of the established observational programme is on characterizing the physical properties and chemical composition of the aerosol, as well as on monitoring the changing trace gas composition of the background atmosphere, especially concerning greenhouse gases. Here a brief outline of the technical concept of the observatory and an excerpt of the most relevant results are presented.

Zusammenfassung: Das Spurenstoff Observatorium an der deutschen Antarktis Station Neumayer (vormals Georg von Neumayer) ist mittlerweile seit fast 25 Jahren kontinuierlich in Betrieb und liefert einzigartige Messreihen klimarelevanter atmosphärischer Spurenstoffe. Der Schwerpunkt des Messprogramms liegt in der Charakterisierung der physikalischen Eigenschaften und chemischen Zusammensetzung des Aerosols sowie in der Dokumentation der langfristigen Änderung der Spurengas-Zusammensetzung in der Hintergrund-Atmosphäre, speziell hinsichtlich der wichtigsten Treibhausgase. In diesem Beitrag werden eine kurze Beschreibung der technischen Auslegung des Observatoriums sowie einige der wichtigsten Ergebnisse präsentiert.

1. INTRODUCTION

The atmosphere above Antarctica constitutes the cleanest part of the Earth's troposphere which allows here to study the composition and temporal change of the background atmosphere without any direct impact of civilization. Furthermore, with the exception of very few rocky terrains, the Antarctic continent is largely free of aerosol and trace gas sources, so that the main part of atmospheric trace compounds must be advected by long range transport to Antarctica or has its source region in the surrounding Southern Ocean. Due to this unique position, Antarctica is an outstanding place to document long-term changes of the composition of our atmosphere in the so called “Anthropocene”, the era that started with the industrial revolution about 200 years ago. The Neumayer Air Chemistry observatory is one of the few clean air laboratories operated in Antarctica with an extensive scientific program, in parts established already in 1982. Since 1997 the air chemistry observatory is part of the GAW (***G***lobal ***A***tmosphere ***W***atch; http://www.empa.ch/gaw/gawsis) global station network. Another important aspect of studying tropospheric trace constituents in Antarctica is the need to interpret records of particulate or reactive trace compounds observed in firn and ice cores. Addressing the coupling between climate and biogeochemical cycles, polar ice cores provide a unique archive of climate proxies from which information about past changes of temperature and atmospheric aerosol load can be derived even in

sub-annual resolution, provided that the controlling mechanisms of the air to firn transfer of relevant species are thoroughly investigated in the present atmosphere. In this context, the realization of deep drilling activities on the central Antarctic ice sheet (EPICA-project; http://www.awi-bremerhaven.de/GPH/EPICA/index.html), the need for recent atmospheric observations to interpret ice core records is strongly emphasized.

The purpose of this contribution is to give a brief overview on the technical layout of the observatory and highlight some outstanding scientific results. Concerning the latter, we will focus on conclusions drawn from the established long-term observational program. For a more comprehensive view, especially concerning particular case studies which we will not address here, see original publications listed below and in http://www.awi-bremerhaven.de/GPH/AirChemistryObservatory/SPUSO.html.

2. TECHNICAL ASPECTS OF THE AIR CHEMISTRY OBSERVATORY

The first Air Chemistry Observatory at Neumayer was initiated and constructed by the Institut für Umweltphysik, University of Heidelberg (UHEI-IUP) in 1982. Following almost 13 years of operation, the technical equipment and the data acquisition facilities had to be renewed. The present observatory was designed in collaboration between AWI and UHEI-IUP as a container building placed on a platform some metres above the snow surface (Figs. 1 & 2). All experiments installed in the Air Chemistry Observatory are under daily control and daily performance protocols are available. A ventilated stainless steel inlet stack (total height about 8 m above the snow surface) supplies the experiments with ambient air.

Fig. 1: *The present Air Chemistry Observatory (on the left) together with the first observatory, which was installed in 1983 and finally dismantled in January 1995.*

Concerning anthropogenic pollutants, the Antarctic atmosphere is "ultra clean". Local pollution by vehicles and the base itself is, thus, a potential problem for many measurements aimed at representing the background status of the Antarctic troposphere. Consequently, a central aspect of the technical concept concerns the ability of contamination-free sampling of aerosols and trace gases. This is realized by several means: (i) The Air

Fig. 2: *A view inside the Air Chemistry Observatory: Aerosol sampling on filters (right hand), integrating nephelometer and condensation particle counters (left hand).*

Chemistry Observatory is situated in a clean air area about 1500 m south of Neumayer. Due to the fact that northern wind directions are very rare, contamination from the base can be excluded for most of the time. (ii) The power supply (20 kW) is provided by a cable from the main station, thus no fuel-driven generator is operated in the very vicinity. (iii) Contamination-free sampling is controlled by the permanently recorded wind velocity, wind direction and by condensation particle concentration. Contamination is indicated if one of the following criteria is given: Wind direction within a 330°-30° sector, wind velocity <2.2 m s^{-1} or >17.5 m s^{-1}, or condensation particle concentrations >2500 cm^{-3} during summer, >800 cm^{-3} during spring/autumn and >400 cm^{-3} during winter. If one or a definable combination of these criteria are given, high and low volume aerosol and part of the trace gas sampling are interrupted. Interestingly, most of the data loss is provoked by blizzards and drifting snow (wind velocity >17.5 m s^{-1}). During such harsh weather conditions aerosol sampling has to be switched off entailing a data loss of roughly 10% per year. Note, that only about 2% of data loss is actually caused by potential contamination!

3. SELECTED RESULTS

3.1. Overview of the scientific program

Tables 1 and 2 present a summary of the current monitoring and sampling programs established at the Neumayer Air Chemistry Observatory. Generally, the observatory provides a platform for both, long-term observations and particular studies performed during one or two over-wintering periods or during summer seasons only. The majority of sample analyses have to be conducted in the home laboratories. The long-term observational program can be divided into three categories. (i) High volume aerosol sampling on precleaned Whatman 541

Table 1: *Summary of the actual sampling program. Abbreviations and symbols: asterisks (*): measured only during special campaigns; AWI, Alfred Wegener Institut für Polar- und Meeresforschung, Bremerhaven; GSF, Forschungszentrum für Umwelt und Gesundheit GmbH, Institut für Hydrologie, München-Neuherberg; IAR, Institut für Atmosphärische Radioaktivität, Freiburg; UHEI-IUP, Institut für Umweltphysik der Universität Heidelberg.*

category	sampling method	analysed compounds	institute
long lived trace gases	compressed air (200 bar)	SF_6, CH_4, $^{13}CH_4$, $CH_3{}^2H$, $^{14}CH_4$	UHEI-IUP
		^{85}Kr, N_2O, $\delta^{15}N$-N_2O*, $\delta^{18}O$-N_2O*	UHEI-IUP/IAR
	compressed air (2 bar) (flask sampling)	CO_2, ^{13}C-CO_2, ^{18}O-CO_2 N_2O, CH_4, SF_6, CO, H_2	UHEI-IUP
	absorption (in NaOH)	$^{14}CO_2$	UHEI-IUP
water vapour			
	cryogenic sampling	H_2O, δ^2H- H_2O, $\delta^{18}O$- H_2O, ^3H- H_2O	UHEI-IUP
reactive trace gases	low volume sampling (teflon/nylon filter combination)	SO_4^{2-}, NO_3^-, Cl^-, MSA, Na^+, NH_4^+, HNO_3, SO_2	AWI
aerosol	high volume sampling (Whatman 541 filter)	SO_4^{2-}, NO_3^-, Cl^-, MSA, Na^+, NH_4^+, ...	UHEI-IUP/AWI
		trace elements*	AWI
	high volume sampling	^{210}Pb, ^7Be, ^{10}Be*	UHEI-IUP
fresh snow		SO_4^{2-}, NO_3^-, Cl^-, MSA, Na^+, NH_4^+, ^{210}Pb, ^{10}Be, ...	AWI/UHEI-IUP

Table 2*. Summary of the actual in situ measuring program. Abbreviations and symbols see Table 1; DLR, Deutsches Zentrum für Luft- und Raumfahrt, Institut für Physik der Atmosphäre, Oberpfaffenhofen.*

category	property	method	institute
aerosols physical properties	particles (>10 nm)	condensation particle counter (CPC)	AWI
	ultra fine particles (>3 nm)	CPC	AWI
	non volatile particles	thermo-denuder / CPC	DLR/AWI
	aerosol size distribution 0.5-20 μm	optical particle counter	AWI
	black carbon, aerosol absorption	Aethalometer and Multi Angle Absorption Photometer (MAAP)	DLR/AWI
	aerosol scattering	integrating nephelometer	AWI
trace gases	^{222}Rn	α-spectroscopy of ^{214}Po	UHEI-IUP
	surface O_3	UV-absorption	AWI
trace gases	column density of O_3, NO_2, OClO, ...	UV spectroscopy (DOAS)	IUPH

cellulose filters (diameter 240 mm) at a flow rate of around 120 m^3/h. Typically about 20 000 m^3 and 40 000 m^3 of ambient air are sucked through filter pairs for analyses of ionic compounds and radio isotopes, respectively.

The main objective of these investigations is to determine the chemical composition of the aerosol, its seasonality and source apportionment. (ii) In situ measurements of reactive trace gases like ozone and aerosol physical properties (number concentration, light scattering, size distribution) for a comprehensive characterization of the aerosol and to assess the photooxidation potential of the Antarctic troposphere. (iii)

Whole air samples to determine long-term concentration trends of greenhouse gases (CO_2, CH_4, N_2O) and their isotopic compositions and anthropogenic tracers (SF_6 and ^{85}Kr) in the background atmosphere. Selected results from each category will be present below. Note that throughout this article atmospheric mixing ratios of trace gases are given in ppm, ppb, and ppt, i.e. molar parts per 10^6, 10^9, and 10^{12}, respectively.

3.2. Biogenic sulfur and particle number concentrations

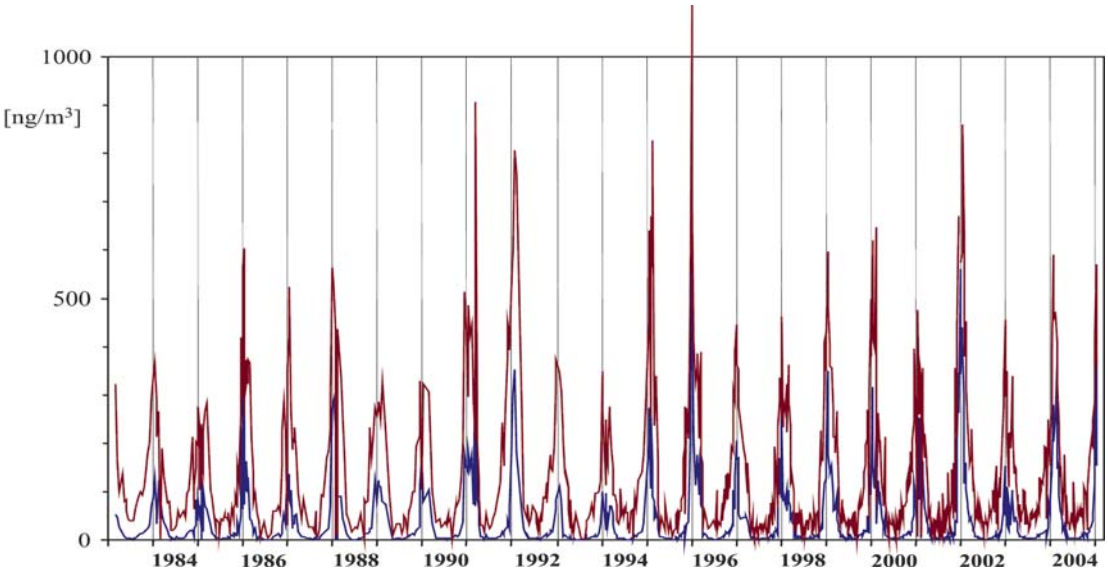

Fig. 3: *Atmospheric methane sulfonic acid (MSA, blue line) and non-seasalt sulfate (nss-sulfate, red line) concentrations sampled with the high volume sampling system (sampling period: one week). Note the distinct seasonality with maximum concentrations in January for both compounds.*

Aerosol formation from biogenic precursor gases such as algae-derived dimethyl sulfide (DMS, CH_3SCH_3) plays an important and crucial role in determining the Earths albedo by direct and indirect effects. This is especially true for the Southern Hemisphere, where the emission of man-made aerosol is still much less dominant than in the Northern Hemisphere. Considering the naturally derived aerosol load of the Southern Hemisphere, the atmospheric photooxidation of DMS leading to the final reaction products methane sulfonic acid (MSA) and sulfuric acid is believed to be the most important process [YIN et al., 1990]. In contrast to sulfate, which comprises a composite signal of marine biogenic, sea salt, terrestrial, and volcanic sources, MSA is known to be virtually exclusively formed by photooxidation of DMS [YIN et al., 1990]. As for the marine biogenic tracer MSA, long-term aerosol measurements at Neumayer revealed that the main source region is the South Atlantic primarily south of 50°S with increasing contribution from more local sources south of 60°S [MINIKIN et al., 1998]. The non-sea salt sulfate (nss-sulfate) and MSA records from Neumayer (Fig. 3) covering now more than 20 years of quasi-continuous observations reveal the strong seasonality of the signal with maximum concentrations in January. In addition the close correlation of nss-sulfate and MSA indicate that most of the sulfate amount in Neumayer aerosol is of marine biogenic origin and thus DMS dominates the overall sulfur budget at this site. The extraordinarily pronounced seasonality of atmospheric MSA and nss-sulfate concentrations is characteristic for coastal Antarctica [MINIKIN et al., 1998] and is linked with the seasonality of the sea ice coverage and insolation. Sea ice retreat provokes distinct phytoplankton blooms [GÜNTHER & DIECKMANN, 1999] associated with DMS emissions into the atmospheric boundary layer. The impact of volcanic

eruptions (El Chichon 1983, Cerro Hudson and Pinatubo 1991) are identifiable in the nss-sulfate signal but secondary compared to the biogenic sulfur source strength [LEGRAND & WAGENBACH, 1999]. This is in contrast to continental Antarctica, where the imprints of prominent volcanic events clearly exceed the biogenic sulfate signal for 1-2 years after the eruption date [TRAUFETTER et al., 2004].

3.3. Condensation particle concentrations

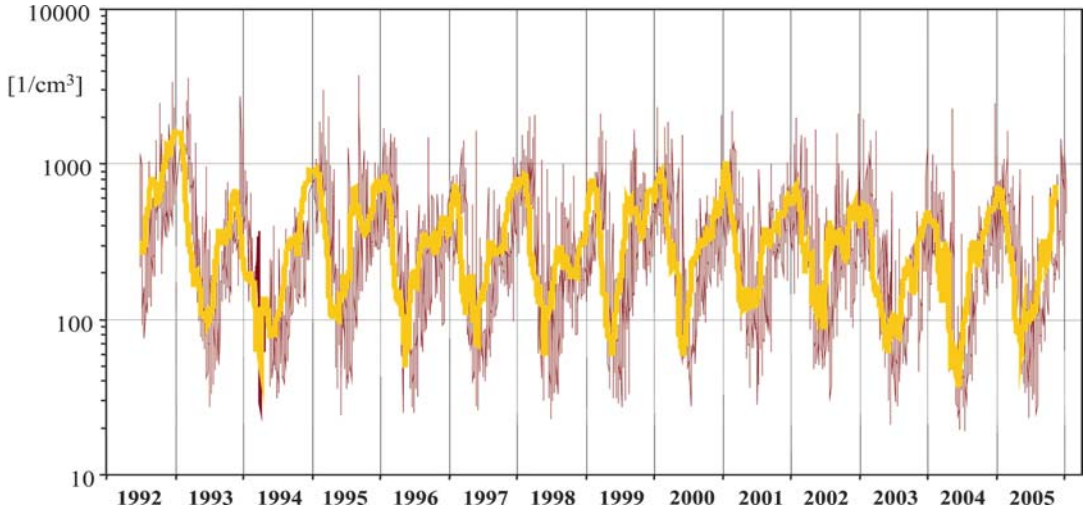

Fig. 4: *Daily means of condensation particle (larger than 10 nm) concentrations at Neumayer. The bold yellow line is a low-pass smoothed representation of the data by a 30 points Gaussian filter.*

Condensation particle (CP) comprise all atmospheric particles which can act as nuclei for condensation of low volatile gaseous compounds like organic species (biogenic or from any combustion process), organic and mineral acids which are both reaction products of atmospheric photooxidation processes and most notably water vapour. Generally particles >3 nm in diameter can act as condensation nuclei. Especially particles within the accumulation mode (particle diameter 0.1 – 2.5 µm) can act as efficient cloud condensation nuclei and play an important role in cloud formation. The latter fact emphasizes the crucial role of aerosols in determining atmospheric radiation transfer. CP concentrations at Neumayer exhibit a stepwise increase from polar winter (below 100 particles/cm^3) to a maximum in late austral summer of around 1000 particles/cm^3 (Fig. 4; data can be retrieved from http://www.awi-bremerhaven.de/GPH/AirChemistryObservatory/SPUSO.html). During summer the chemical composition of these particles is mainly MSA and nss-sulfate, thus CN are formed in the marine troposphere by photooxidation of DMS emitted by the phytoplankton. During winter and stormy weather conditions, however, sea salt dominates the aerosol mass. Interestingly, the maximum of condensation particle concentration typically appears in late February to early March, i.e. shifted by around 4-6 weeks compared to the MSA and nss-sulfate maxima. Our measurements suggest that during late summer the concentration of very small particles between 3 and 5 nm diameter (nucleation mode) is significantly enhanced, indicating new particle formation. DMS could act via its photooxidation product sulphuric acid as gaseous precursor for nucleation mode particles, a process known as gas to particle conversion. Due to the relatively short atmospheric lifetime (a few hours) of nucleation mode particles, regional sources should dominate the measured signal. We believe that following the retreat of sea ice in the nearby Atka bay during late February, considerable amounts of DMS are released by the now emerging phytoplankton bloom in this area. Note that nucleation mode particles do not

contribute much to the total aerosol mass due to their small size, therefore nss-sulfate and MSA concentration maxima do not necessarily coincide with the particle number concentration maximum!

3.4. Tropospheric ozone

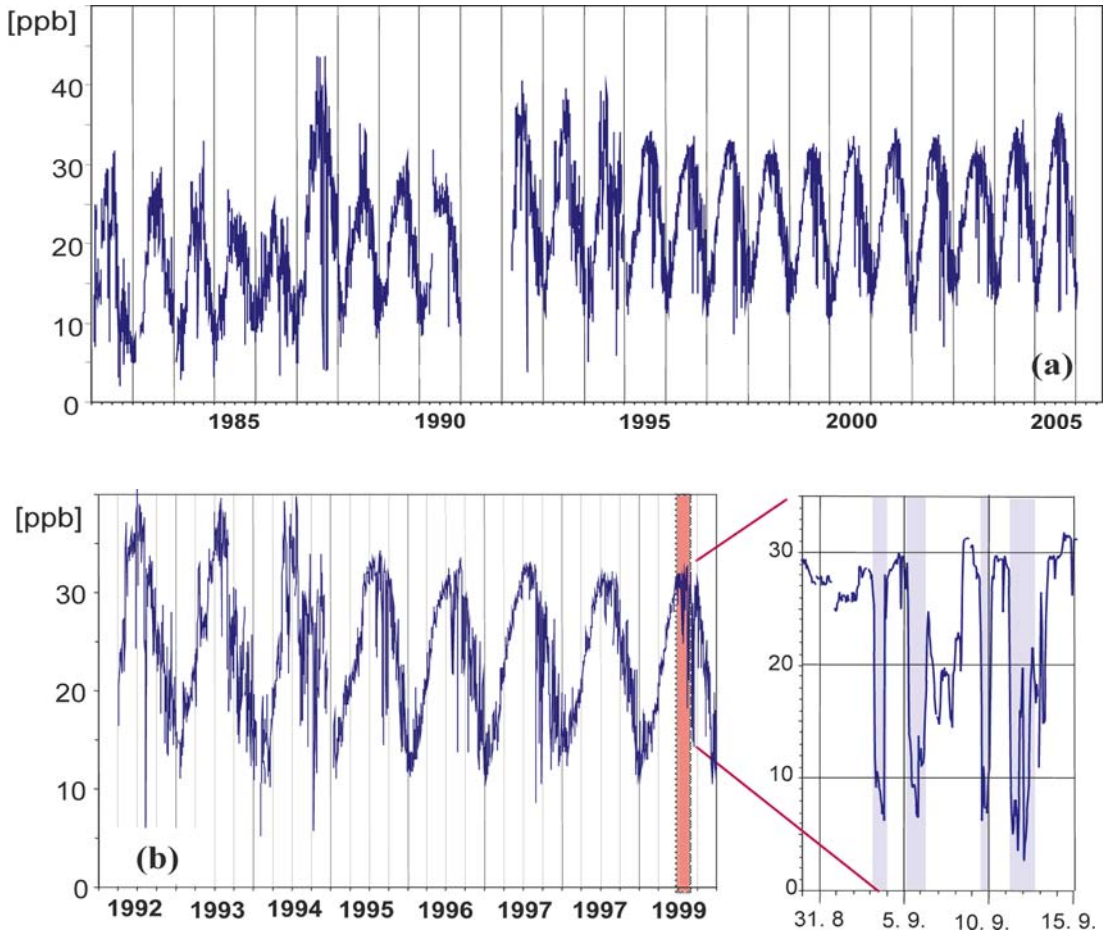

Figure 5: *Surface ozone mixing ratios (daily means) record at Neumayer from 1982 to 2005 (a). The enlarged section on the right hand side of (b) shows typical tropospheric ozone depletion events (shaded areas) in more detail (hourly averages).*

It is generally accepted that the photooxidation of trace gases to water soluble compounds, followed by rainout, is the major cleaning procedure of tropospheric air. Photooxidation in the troposphere consists of typical radical chain reactions which need to be initiated, mainly by OH radicals. OH in turn is primarily generated via ozone photolysis and subsequent reaction of the so produced O^1D (i.e. electronically excited oxygen) atoms with water vapour. Thus tropospheric ozone is certainly a key trace gas in controlling the chemical composition of the troposphere. Surface ozone is continuously measured since 1982 at Neumayer Station by electro-chemical concentration cells (ECC, until 1994) and uv-absorption from 1994 ongoing (Fig. 5a). O_3 mixing ratios measured at the former Georg von Neumayer Station by ECC seem to be significantly lower before 1987, a probably artificial peculiarity which is not yet clarified. Nevertheless from this record, covering now 24 years of observation, no significant trend can be deduced. A more detailed section of this times series is depicted in Fig. 5b. Maximum ozone values of about 32 ppbv are generally observed in August while during polar summer (December-January) a distinct minimum of around 13 ppbv is typical. In strong contrast to urban areas where

nitrogen oxides (NO_x) levels are about three orders of magnitude higher, photochemical ozone destruction and not formation occurs in summer leading to surface ozone minima in pristine regions like Antarctica. In addition, from August to September extraordinary tropospheric ozone depletion events can frequently be detected (Fig. 5b). Neumayer Station was the first Antarctic site where this peculiarity was described [WESSEL et al., 1998]. Comparable to stratospheric ozone depletion, reactive halogen compounds, here especially BrO, are responsible for this anomaly [FRIEß et al., 2004]. However, in contrast to the chemical processes occurring in the stratosphere, tropospheric ozone depletion in polar regions is a natural phenomenon most probably caused by release of reactive bromine compounds eventually derived from sea-salt over sea-ice. There is some recent evidence that frost flowers, which frequently grow on newly-formed sea ice, play an important, if not crucial role as a source for reactive tropospheric halogen compounds [KALESCHKE et al., 2004].

3.5. Greenhouse gases

Neumayer is the only station in Antarctica where the main greenhouse gases carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O) as well as their isotopic compositions $\delta^{13}C\text{-}CO_2$, $\delta^{18}O\text{-}CO_2$, $^{14}C\text{-}CO_2$, $\delta^{13}C\text{-}CH_4$, $\delta D\text{-}CH_4$, $\delta^{18}O\text{-}N_2O$, and $\delta^{15}N\text{-}N_2O$ are measured simultaneously. In particular from the isotopic signature information about the corresponding greenhouse gas sources and sinks can be deduced [for details see LEVIN et al., 1987; HESSHAIMER et al., 1994; LEVIN & HESSHAIMER, 2000; POß, 2003; RÖCKMANN & LEVIN, 2005]. The mixing ratio and isotopic analyses are made on whole air samples collected in high pressure cylinders as well as in 1-litre glass flasks.

Carbon dioxide: The long-term increase of CO_2, caused by an ongoing input of CO_2 from fossil fuel burning and land-use change into the atmosphere, was almost linear over the last decade (growth rate 1.8 ppm yr^{-1}, Fig. 6), however, large inter-annual variations are obvious. For example in 1997/98 during a strong El Niño period a positive CO_2 anomaly was observed which is accompanied by a significant decrease of $\delta^{13}C\text{-}CO_2$ (not shown), indicating unusually large continental biogenic CO_2 emissions during this period.

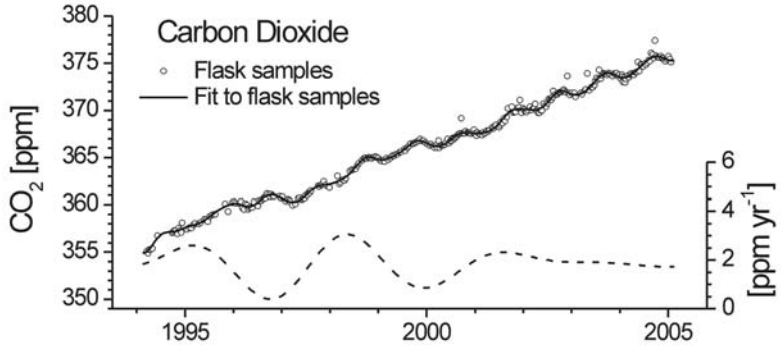

Fig. 6: *Atmospheric carbon dioxide mixing ratios at Neumayer, measured on flask samples by gas chromatography. The solid lines are smoothed curve fits calculated with a fitting procedure described by NAKAZAWA et al. [1997], the dashed line gives the annual growth rate in ppm/year.*

Methane: At Neumayer, methane mixing ratio has been measured on high-volume air samples since 1986 and on flask samples since 1994. Methane increased (globally) by approximately 75 ppb over the last 15 years. Interestingly, the growth rate decreased continuously from about 10-15 ppb yr^{-1} in the 1980s to about 5 ppb yr^{-1} in the 1990s and finally fluctuates around zero since about the year 2000 (Fig. 7). This trend in the CH_4 growth rate indicates that, in the last years, the sources of this greenhouse gas approach an equilibrium with its sinks (photochemical oxidation). However, with increasing emissions in the future associated with growing population and energy demand, atmospheric CH_4 mixing ratios will possibly increase again in the future. The distinct sinusoid like seasonal cycle of methane in the Southern Hemisphere with minimum mixing ratios during austral summer is caused by the seasonality of the photochemical CH_4 oxidation.

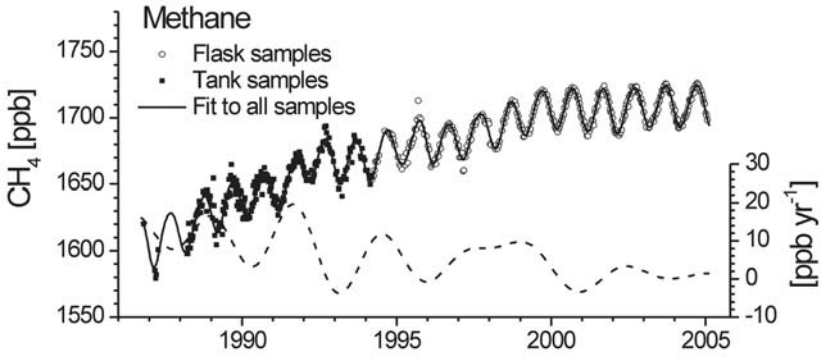

Fig. 7: *Atmospheric methane mixing ratios at Neumayer, measured on high-volume air samples by gas chromatography. The solid lines are smoothed curve fits calculated with a fitting procedure described by NAKAZAWA et al. [1997], the dashed line gives the annual growth rate in ppb/year.*

Nitrous oxide: During the last decade N_2O mixing ratio at Neumayer increased by 0.83 ppb yr^{-1} (Fig. 8). Our isotope observations, the first long-term measurements of this species in the atmosphere, show corresponding trends of $\delta^{15}N\text{-}N_2O$ of -0.040‰ yr^{-1} and $\delta^{18}O\text{-}N_2O$ of -0.021‰ yr^{-1} providing observational evidence that mainly isotopically depleted N_2O from soils must be responsible for a large fraction of the observed N_2O increase in the global atmosphere [RÖCKMANN & LEVIN, 2005]. The seasonal variation of the N_2O mixing ratio with a mean peak-to-peak amplitude of 0.83 ppb is mainly caused by seasonally varying oceanic N_2O emissions as well as by stratosphere-troposphere exchange processes [LEVIN et al., 2002; NEVISON et al., 2005].

Sulfur hexafluoride (SF_6). Sulfur hexafluoride is almost exclusively emitted into the atmosphere by man and mainly used in electric switch gear and for degassing molten reactive metals. Due to its extremely long atmospheric life time of more than 1000 years, almost all SF_6 remains in the atmosphere. Consequently, starting around 1970, mixing ratios persistently increased by several percent per year during the last three decades (Fig. 9). The linear trend observed over the last decade indicates a virtually constant emission rate since 1995. Sulfur hexafluoride is also an excellent tracer to validate atmospheric transport models as the distribution of its sources is well known and mainly restricted to the Northern Hemisphere [LEVIN & HESSHAIMER, 1996]. From the well

established north-south difference of SF_6 mixing ratios, a mean inter-hemispheric exchange time of 1 to 1.5 years can be deduced [MAISS & LEVIN, 1994].

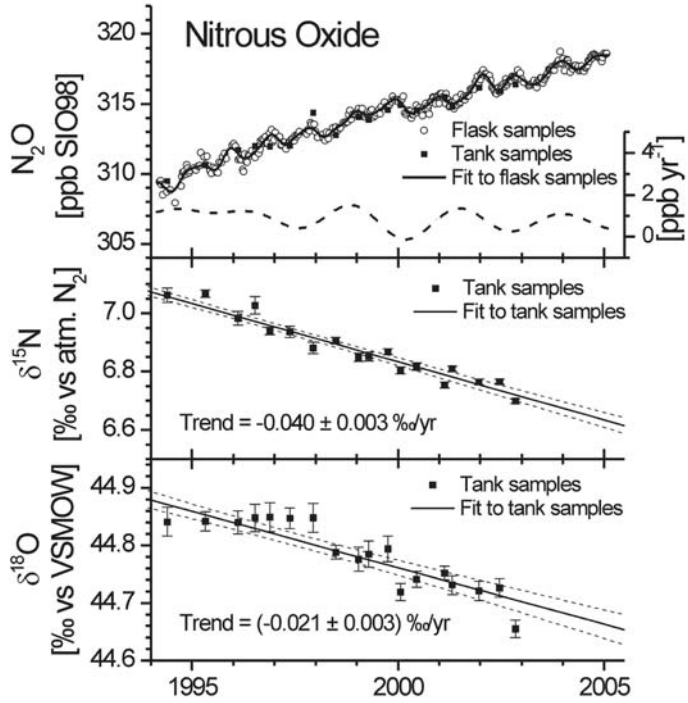

Fig. 8: *Atmospheric nitrous oxide mixing ratios and isotopic composition at Neumayer, measured on air samples by gas chromatography and isotope ratio mass spectroscopy (IR-MS). The solid lines are smoothed curve fits calculated with a fitting procedure described by NAKAZAWA et al. [1997], the dashed line gives the annual growth rate in ppb/year.*

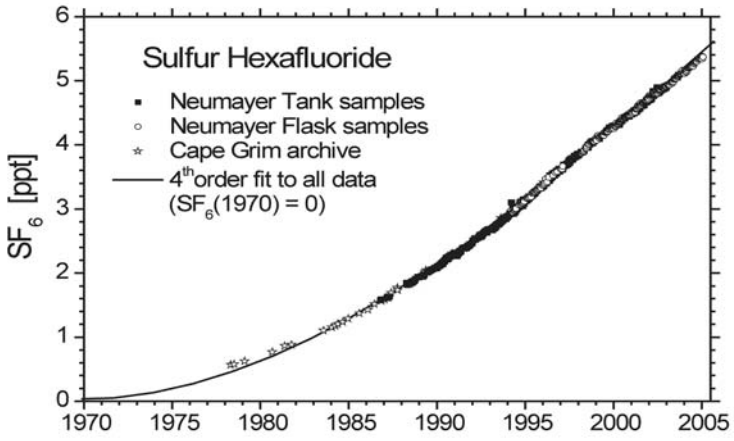

Fig. 9: *Atmospheric sulfur hexafluoride mixing ratios at Neumayer, measured on different air samples (high volume (tank), flasks and from archived tank samples) by gas chromatography.*

4. CONCLUSIONS, PERSPECTIVES, AND OUTLOOK

Contamination free air sampling as realized in the Air Chemistry Observatory at Neumayer has proved one's worth and is partly copied in the meanwhile at other stations, e.g. CASlab (Clean Air Sector Laboratory) at Halley. After 13 years of permanent operation, the present Air Chemistry Observatory (or "Spuso" as affectionately called by the over-winterers) is expected to be replaced in austral summer 2008 concurrently to the planned assembly of Neumayer III, the new German Antarctic Station. Both containers of the present observatory are now seriously stained (see Figure to the right) while part of the equipment is near failure preventing prolonged operation.

Concerning new scientific projects, we intend to install a specially designed Lidar system allowing remote monitoring of tropospheric aerosols. Until now, only very limited information on the vertical aerosol stratification above Antarctica are available. This new experiment will provide us year-round data on vertical aerosol distribution, similar to the set-up already installed at Koldewey Station, Spitzbergen (Arctic). Furthermore, especially regarding ice core projects like EPICA, there is a need to expand our year-round aerosol sampling program to the Antarctic Plateau. Only very few year-round measurements of atmospheric constituents are available from continental Antarctica and thus e.g. the seasonality of most ionic aerosol compounds is so far unknown for continental Antarctica. Within the EPICA project, an automated aerosol sampler was installed in February 2003 at Kohnen Station, the deep drilling location in Dronning Maud Land (75°S, 0°E, 2892 m a.s.l.) and will be in operation for the next years. Due to the fact that Kohnen Station is, in contrast to EPICA Dome C, only a summer camp and not a wintering-over station, the installation of an automated aerosol sampler was required. To our knowledge, we succeeded for the first time in running a stand-alone aerosol sampler in continental Antarctica throughout a year. Nevertheless, automated air chemistry observatories are generally restricted to very few and particular measurements and can definitively not substitute observatories which are daily attended and whose sophisticated experiments are critically assessed every day by skilled scientific staff.

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