**Atmospheric
Chemistry
and Physics
Discussions**

# Reconciliation of essential process parameters for an enhanced predictability of Arctic stratospheric ozone loss and its climate interactions

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## Abstract

Significant reductions in stratospheric ozone occur inside the polar vortices each spring when chlorine radicals produced by heterogeneous reactions on cold particle surfaces in winter destroy ozone mainly in two catalytic cycles, the ClO dimer cycle and the ClO/BrO cycle. Chlorofluorocarbons (CFCs), which are responsible for most of the chlorine currently present in the stratosphere, have been banned by the Montreal Protocol and its amendments, and the ozone layer is predicted to recover to 1980 levels within the next few decades. During the same period, however, climate change is expected to alter the temperature, circulation patterns and chemical composition in the stratosphere, and possible geo-engineering ventures to mitigate climate change may lead to additional changes. To realistically predict the response of the ozone layer to such influences requires the correct representation of all relevant processes. The European project RECONCILE has comprehensively addressed remaining questions in the context of polar ozone depletion, with the objective to quantify the rates of some of the most relevant, yet still uncertain physical and chemical processes. To this end RECONCILE used a broad approach of laboratory experiments, two field missions in the Arctic winter 2009/10 employing the high altitude research aircraft M55-Geophysica and an extensive match ozone sonde campaign, as well as microphysical and chemical transport modelling and data assimilation. Some of the main outcomes of RECONCILE are as follows: (1) vortex meteorology: the 2009/10 Arctic winter was unusually cold at stratospheric levels during the six-week period from mid-December 2009 until the end of January 2010, with reduced transport and mixing across the polar vortex edge; polar vortex stability and how it is influenced by dynamic processes in the troposphere has led to unprecedented, synoptic-scale stratospheric regions with temperatures below the frost point; in these regions stratospheric ice clouds have been observed, extending over $> 10^6$ km$^2$ during more than 3 weeks. (2) Particle microphysics: heterogeneous nucleation of nitric acid trihydrate (NAT) particles in the absence of ice has been unambiguously demonstrated; conversely, the synoptic scale ice clouds also

appear to nucleate heterogeneously; a variety of possible heterogeneous nuclei has been characterised by chemical analysis of the non-volatile fraction of the background aerosol; substantial formation of solid particles and denitrification via their sedimentation has been observed and model parameterizations have been improved. (3) Chemistry: strong evidence has been found for significant chlorine activation not only on polar stratospheric clouds (PSCs) but also on cold binary aerosol; laboratory experiments and field data on the ClOOCl photolysis rate and other kinetic parameters have been shown to be consistent with an adequate degree of certainty; no evidence has been found that would support the existence of yet unknown chemical mechanisms making a significant contribution to polar ozone loss. (4) Global modelling: results from process studies have been implemented in a prognostic chemistry climate model (CCM); simulations with improved parameterisations of processes relevant for polar ozone depletion are evaluated against satellite data and other long term records using data assimilation and detrended fluctuation analysis. Finally, measurements and process studies within RECONCILE were also applied to the winter 2010/11, when special meteorological conditions led to the highest chemical ozone loss ever observed in the Arctic. In addition to quantifying the 2010/11 ozone loss and to understand its causes including possible connections to climate change, its impacts were addressed, such as changes in surface ultraviolet (UV) radiation in the densely populated northern mid-latitudes.

## 1 Introduction

The stratospheric ozone layer shields the Earth's surface from UV radiation. Human activities, primarily the release of chlorofluorocarbons (CFCs) and halons in the second half of the 20th century, have led to a thinning of this protective layer which is most particularly marked in polar regions in spring (WMO, 2010). Even though the production of CFCs has been banned by the Montreal Protocol and its subsequent amendments, and the ozone layer is expected to recover by the middle of the century at mid-latitudes and by the end of the century over Antarctica (Eyring et al., 2010; WMO, 2010), climate

change induced changes in stratospheric conditions and their impact on ozone are still not easy to predict. The record-breaking ozone loss that occurred over the Arctic in spring 2011 (Manney et al., 2011; Kuttippurath et al., 2012a) has shown that stratospheric ozone is still a critical issue, and that climate change could bring about ozone hole like conditions in the Arctic as long as stratospheric chlorine levels are still high enough.

With its focus primarily on the Arctic stratosphere, the EU funded project RECONCILE, short for "Reconciliation of essential process parameters for an enhanced predictability of Arctic stratospheric ozone loss and its climate interactions", runs from 2009 to 2013 and aims at understanding how the physical and chemical processes affecting stratospheric ozone will be influenced by and feedback on climate change. In Sect. 2, a brief overview of past research on polar ozone depletion is given, and the motivation and objectives of RECONCILE are stated. The overall strategy as well as the tools and activities adopted are described in Sect. 3, and a summary of the main scientific findings of the RECONCILE project is presented in Sect. 4. Section 5 explains how these results are used to advance our ability to predict the future state of the ozone layer using chemistry climate models (CCMs), i.e. global circulation models (GCMs) that include chemical reactions. Section 6 gives a brief overview on the record Arctic ozone depletion 2011 and its implications.

## 2   Historical background and objectives

Below, the history of research and policies on stratospheric ozone is briefly reviewed and the state of knowledge, including open questions, at the beginning of the RECONCILE project is described. Some key events – from the early warnings of possible dangers to the ozone layer expressed in the 1970s to recent international research projects with a focus on polar stratospheric ozone – are illustrated in Fig. 1.

## 2.1 The discovery of the ozone hole and the Montreal Protocol

In 1985 Joe Farman, Brian Gardiner and Jonathan Shanklin from the British Antarctic survey reported "that the spring values of total $O_3$ in Antarctica have now fallen considerably" (Farman et al., 1985), and others soon confirmed this observation (Chubachi and Kajiwara, 1986; Gernandt, 1987; Stolarski et al., 1986). Even though a number of scientists had expressed concerns that anthropogenic emissions of CFCs (Cicerone et al., 1974; Crutzen, 1974; Molina and Rowland, 1974), nitrous oxide ($N_2O$) from fertilizer use (Crutzen, 1976), and reactive nitrogen compounds from high-flying aircraft (Johnston, 1971; Crutzen, 1971) would potentially lead to faster catalytic ozone removal rates and hence lower stratospheric ozone concentrations, the "Ozone Hole" came as a surprise to atmospheric scientists. But with the research that had already been carried out in the 1970s as a solid foundation, a theory was soon developed that the dramatic ozone destruction over the Antarctic in spring is the result of a complex interplay of atmospheric dynamics, heterogeneous reactions, and catalytic reactions involving chlorine compounds (Fig. 2).

A link was made between these stratospheric chlorine compounds and CFCs, which had been introduced as refrigerating agents and as propellants in spray cans in the middle of the 20th century. Being inert in the lower atmosphere, these compounds are transported to the stratosphere, where they release chlorine upon UV irradiation. We undoubtedly know today that the attribution of CFCs as the ultimate cause of the ozone hole has been correct: Morgenstern et al. (2008) and Newman et al. (2009) recently demonstrated that this first widely visible global environmental threat caused by human activities had the potential for a global catastrophe.

Fortunately, the 1987 Montreal Protocol and its subsequent amendments and adjustments essentially banned CFCs and certain other chlorinated and brominated compounds from being produced. As a result, stratospheric chlorine and bromine levels have started to decline since the late 1990s and will continue to do so over the next decades, although this will be a slow process due to their long atmospheric lifetimes

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(WMO, 2010). In most regions, column ozone is expected to return to 1980 levels (1980 is usually taken as the reference year for the so-called ozone recovery) around the middle of the century (Eyring et al., 2010; WMO, 2010). However, the state-of-the-art CCM simulations used to estimate the recovery dates, clearly show that the decline in ozone depleting substances (ODS) is only part of the story. The alterations to atmospheric dynamics and composition induced by global climate change (see Figs. 3–22 in WMO, 2010) become increasingly important and have a significant impact on stratospheric ozone and consequently on recovery date projections (Eyring et al., 2010; Li et al., 2009; Waugh et al., 2009; WMO, 2010).

## 2.2 Arctic variability and interactions with climate change

Ozone is a greenhouse gas and radiatively heats the stratosphere. The reduced stratospheric heating related to the substantially reduced ozone amounts in Antarctic spring has led to notable changes in atmospheric dynamics in the Southern Hemisphere (Polvani et al., 2010; Gillett and Son, 2012 and references therein) and may be the cause for observed increases in Antarctic sea ice (Turner et al., 2009). Increases in UV radiation caused by ozone depletion may affect ecosystems with possible feedbacks on the carbon cycle.

On the other hand, climate change affects biogeochemical processes as well as atmospheric dynamics, leading to alterations in emissions and transport times of many trace gases that play a role in stratospheric chemistry and affect ozone (Engel et al., 2009 and references therein; Waugh et al., 2009). A major factor in Arctic ozone depletion, the stability of the polar vortex, directly depends on planetary wave forcing, which is also sensitive to climate change (e.g. Shindell et al., 1999; Sigmond et al., 2008; Dameris and Baldwin, 2012 and references therein; McLandress and Shepherd, 2009). A particular susceptibility of the Arctic stratosphere to climate change may manifest in the trend of cold stratospheric winters in the Arctic getting colder, amplifying heterogeneous chlorine activation and ozone depletion (Rex et al., 2004, 2006).

Over longer periods, climate change and possible geoengineering ventures to mitigate climate change (e.g. Crutzen, 2006) may radically alter the temperature, circulation patterns and chemical composition in the stratosphere. It has been argued (e.g. Tilmes et al., 2008) that the artificial injection of sulphur into the stratosphere considered as a geoengineering venture (e.g. Crutzen, 2006) would increase the extent of Arctic ozone depletion in cold winters and would considerably delay the recovery of the Antarctic ozone hole. To realistically predict the response of the ozone layer to these changes and the future evolution of Arctic ozone, a complete and correct representation of all relevant processes is necessary.

## 2.3 Major breakthroughs in understanding polar ozone depletion through previous projects and field campaigns

A large number of international research projects (included on the timeline shown in Fig. 1) carried out during the past 25 yr have continuously advanced our understanding of the processes involved in ozone depletion. Virtually all major breakthroughs (e.g. role of chlorine radicals in ozone destruction by Anderson et al., 1989; detection of large nitric acid trihydrate (NAT) particles by Fahey et al., 2001; heterogeneous nucleation and growth of NAT particles by Voigt et al., 2005) have been based on outstanding experimental results gathered during large scale field campaigns. Some of these results were somewhat unexpected or even surprising and have triggered new research.

Since the Airborne Antarctic Ozone Experiment (AAOE) in 1987 (Tuck et al., 1989), numerous campaigns for the exploration of polar as well as mid-latitude and tropical UTLS (upper troposphere and lower stratosphere) processes have been carried out (e.g. Turco et al., 1990; Anderson and Toon, 1993; Newman et al., 2002; Vaughan et al., 2008; Cairo et al., 2010), employing state-of-the-art instrumentation and elaborate flight strategies that exploit the full horizontal and vertical manoeuvrability of high-altitude research aircraft like the US ER-2 and Russian M55 Geophysica and complemented by parallel balloon-borne and lower flying aircraft measurements and extensive modelling

activities. The RECONCILE project and the Arctic field campaign described in this paper stand in and continue this fruitful tradition.

## 2.4 Objectives of RECONCILE

At the beginning of the RECONCILE project in 2009, the lack of understanding in strato-
spheric dynamics and chemistry was most palpable for issues including the catalytic $ClO_x/BrO_x$ chemistry, chlorine activation on cold stratospheric aerosol, NAT nucleation mechanisms, and mixing and transport of processed air to lower latitudes. Therefore, the following specific objectives were defined for RECONCILE:

– Fully understand and correctly represent in global circulation models the transport
and mixing across the edge of the polar vortex.

The edge of the polar vortex can be defined in several ways (e.g. Dameris et al., 1995; Harvey et al., 2002; Nash et al., 1996; Waugh and Polvani, 2010). Dynamically, it is roughly centred in the polar night jet that develops in the stratosphere over the winter poles in response to seasonal cooling over the respective polar region. The strong cir-
cumpolar westerly winds act as a transport barrier and affect both the extent of ozone depletion and the meridional exchange of ozone depleted air with lower latitudes. Due to the distinct land-sea distribution in the Northern Hemisphere, tropospheric weather regimes and associated planetary wave activity modify the strength of the polar night jet and lead to instabilities. Therefore, the persistence of the polar vortex and, even-
tually, the strength of transport barrier are rather variable during each winter and vary interannually. A misrepresentation of the transport and mixing across the vortex edge in global circulation models affects our ability to reliably predict both quantities in future climate scenarios.

– Elucidate in detail the processes of NAT nucleation leading to polar stratospheric
cloud (PSC) formation, in particular the role of meteoritic dust and other refractory material as condensation nuclei, and the mechanism by which NAT particles grow to rather large sizes forming so-called NAT rocks.

The conventional understanding of PSC formation based on NAT nucleation depending on prior ice formation has been questioned based on in-situ and ground-based lidar observations (Drdla et al., 2002; Pagan et al., 2004; Voigt et al., 2005). However, these in-situ and ground-based lidar observations relied on spatially and temporally limited data and left room for uncertainties. One issue were temperature histories along air parcel trajectories, which were calculated using gridded data of numerical weather prediction (NWP) models. Due to the limited spatial and temporal resolution, these temperature histories were not reliable because, for example, effects of stratospheric mountain waves were not taken into account (Dörnbrack et al., 1998, 1999).

The possible role of meteoritic dust and other refractory material as condensation nuclei has been pointed out (Curtius et al., 2005; Voigt et al., 2005). NAT particles with diameters of tens of microns have been observed and termed "NAT rocks" (Brooks et al., 2003; Fahey et al., 2001; Fueglistaler et al., 2002). When gravitationally sedimenting, these particles efficiently transport and, eventually, redistribute nitric acid, $HNO_3$. Later in the polar winter, this redistribution impacts the potential for further PSC formation and the rate of chlorine deactivation into chlorine nitrate, $ClONO_2$.

– Investigate the importance of chlorine activation on background aerosol.

Katja Drdla (Drdla, 2005; Drdla and Müller, 2012) pointed out that at sufficiently low temperatures, chlorine is activated on the background binary aerosol, and that the actual formation of PSCs is not a prerequisite for chlorine activation. This is of particular importance in the context of enhanced stratospheric water vapour concentrations fostering an enhanced total reactive surface area (Anderson et al., 2012), or of geo-engineering concepts to cool the Earth's surface by deliberately enhancing the stratospheric loading of sulphate aerosol (Crutzen, 2006) or other aerosol compositions (Pope et al., 2012). Clearly, the process of chlorine activation on binary aerosol could have consequences for stratospheric ozone on a global scale.

– Reduce uncertainties and reach an adequate level of confidence concerning the parameters governing the rate of catalytic ozone loss

Uncertainties in various kinetic parameters and the stratospheric bromine budget led to significant uncertainties in model simulations of ozone loss (e.g. Frieler et al., 2006). The single most critical parameter governing the rate of catalytic ozone loss in polar spring is the ClOOCl photolysis rate $J_{ClOOCl}$, i.e. the product of the actinic flux and the ClOOCl photolysis cross sections $\sigma_{ClOOCl}$. In 2007, values of $\sigma_{ClOOCl}$ determined in numerous laboratory experiments differed by up to a factor of 10, and an experiment by Pope et al. (2007) that suggested the lowest absorption cross sections ever published even put into question our qualitative understanding of the catalytic ozone loss (von Hobe et al., 2007; Schiermeier, 2007; von Hobe, 2007) leading to speculations on previously unknown additional ozone loss mechanisms (e.g. Salawitch et al., 2009; Vogel et al., 2006).

Besides investigating these dynamic, microphysical and chemical processes, and thus completing our knowledge and understanding of the processes leading to polar ozone depletion, the central aim of RECONCILE is to develop reliable process parameterisations and implement them in a global CCM. It is worth pointing out that the model spread in stratospheric ozone simulations and projections is particularly wide for the Arctic region (SPARC, 2010). This can be attributed to high natural variability but also to very significant model differences in the representations of polar processes.

## 3   RECONCILE strategy and activities

Figure 3 gives a graphical representation of the RECONCILE overall approach. To address the uncertainties and open questions as named in Sect. 1.4, a comprehensive strategy of laboratory experiments, aircraft and match campaigns, microphysical and chemical transport modelling, and data assimilation is employed. The results are used to produce reliable parameterisations of key processes in Arctic stratospheric ozone depletion, which are implemented in a global CCM. The quality of the simulations is assessed against observations.

### 3.1 Laboratory experiments

#### 3.1.1 The ClOOCl absorption spectrum

A dual wavelength range spectrometer system that can simultaneously perform single pass UV absorption spectroscopy (wavelength range 200–350 nm) and broadband cavity enhanced absorption spectroscopy (BBCEAS, wavelength range 510–560 nm) has been used to investigate the UV and visible absorption cross sections of molecular chlorine ($Cl_2$) and ClOOCl at different temperatures (Young et al., 2011). With this spectrometer, the experiment carried out by Pope et al. (2007) – i.e. purifying ClOOCl by cold trapping prior to spectral analysis – was repeated, but now with the possibility to simultaneously monitor and unambiguously subtract the $Cl_2$ contribution to the spectrum, i.e. the impurity that proved to cause major uncertainties in the previous experiment.

#### 3.1.2 Heterogeneous NAT nucleation

The heterogeneous NAT nucleation rate on meteoritic nuclei was investigated by revisiting earlier experiments of Biermann et al. (1996). Back then, an upper bound had been reported for freezing rates of 6 different ternary $H_2SO_4$/$HNO_3$/$H_2O$-solutions with immersed micrometeorites under stratospheric conditions. Using a Differential Scanning Calorimeter (DSC) (e.g. Koop and Zobrist, 2009; Marcolli et al., 2007; Zobrist et al., 2008), new bulk experiments were carried out on fragments of the meteorites Orgueil and Murchison with radii varying between 100 and 500 nm, provided in a collaboration with CSNSM (Centre de Spectrométrie Nucléaire et de Spectrométrie de Masse, Paris, France).

### 3.2 Field observations

Extensive field measurements to study processes related to transport and mixing within and at the edge of the polar vortex, PSCs, chlorine chemistry and ozone loss were

carried out in the Arctic winter 2009/10. This winter was marked by a sequence of tropospheric disturbances and associated planetary wave activity impacting the formation and evolution of the polar vortex in late November and early December 2009. Afterwards, a period of exceptionally cold temperatures, leading to extensive PSC formation, followed in January 2010 due to the development of a positive phase of the Western Pacific (WP) teleconnection pattern in the troposphere, as described by Nishii et al. (2011) and Orsolini et al. (2009). A major sudden stratospheric warming (SSW) near the end of January 2010 concluded the anomalously cold vortex period and prevented an extensive ozone loss in the RECONCILE winter (cf. Sect. 4.1). A detailed overview of the meteorological situation and evolution of the polar vortex has been given in Dörnbrack et al. (2012). An additional match campaign as well as investigations based on satellite observations were carried out in the subsequent Arctic winter 2010/11 that saw record breaking ozone loss (Manney et al., 2011, cf. Sect. 6).

### 3.2.1 The Geophysica aircraft campaign

During two measurement periods (17 January–2 February and 27 February–10 March 2010) to cover early and late winter conditions, 12 RECONCILE flights of the high-altitude aircraft M55-Geophysica totalling 57 flight hours were carried out from Kiruna, Sweden, complemented by one flight of the PremierEX project funded by ESA (Spang et al., 2011). An overview of all flights with the respective scientific missions is given in Table 1 and the flight tracks are shown in Fig. 4. An overview of the instruments deployed onboard the M55-Geophysica during the RECONCILE campaign is given in Table 2. The data will be made available online at https://www.fp7-reconcile.eu/reconciledata.html.

Flight planning was based on results of the Integrated Forecast System (IFS) of the European Centre of Medium-Range Weather Forecasts (ECMWF). Different thermodynamical fields and derived quantities from the deterministic run were visualized on a web-site as part of the mission support system being developed at DLR (Deutsches Zentrum für Luft und Raumfahrt, Rautenhaus et al., 2012). Chemical forecasts were

made using a CLaMS (Chemical Lagrangian Model of the Stratosphere, cf. Sect. 3.5) simulation that started at 1 December 2009 and was run using up-to-date ECMWF analyses and ECMWF forecasts 3 to 4 days into the future.

Air mass trajectories for Lagrangian match experiments were also based on ECMWF forecasts. The idea of the match method consists in probing an individual air parcel twice in order to obtain information on the temporal evolution of the concentration of chemical species (cf. Sect. 3.2.2). During the RECONCILE aircraft campaign it was applied to three flights. During two self-match flights air parcels probed during the outbound flight leg were sampled again during the return flight leg to investigate the temporal evolution of the concentration of chlorine species during sunrise. During one match flight air parcels probed during one flight were sampled again three days later to investigate the transport and mixing processes in the stratosphere (Fig. 5).

### 3.2.2  Match sonde and balloon activities

A match campaign in the winter 2009/10 consisted of two elements, ozone sonde matches and COBALD backscatter sonde activities, including water vapour soundings.

The match technique to estimate winter ozone loss has been first described by Rex et al. (1999). 31 ozone sounding stations (Fig. 6) participated in the 2010 match campaign and 573 ozone sondes were launched from 29 of these stations. The ozone sonde match campaign started on 5 January and was ended on 28 February, after a major warming had occurred and the meteorological conditions for continuing the campaign deteriorated. The factors that led to the decision to terminate the campaign included large shear and deformation in the flow, expected small scale filaments in the ozone field and the position of the remnants of the polar vortex which were over Eastern Siberia at that time, out of reach of the network of match stations. A vortex remnant returned to Canada and later to the European sector of the Arctic during mid-March. Coordinated sondes were launched into this remnant to assess whether ozone loss had continued after the end of the match campaign.

An additional ozone sonde match campaign was carried out in the winter 2010/11 with about 270 ozone sonde launches from 13 stations actively participating in the campaign. The campaign started on 12 January 2011 and was stopped on 27 April 2011.

The recently developed light-weight, high-power-LED-based aerosol backscatter sonde COBALD (Compact Optical Backscatter and AerosoL Detector, Wienhold, 2012) was successfully employed during the RECONCILE campaign under Arctic winter conditions (Engel et al., 2012a; Khaykin et al., 2012). During the PSC phase in January 2010, 8 COBALD launches with PSC observations were carried out from Ny-Ålesund, with the advantage of reaching altitudes higher than the M55-Geophysica ceiling altitude. Matches (planned and confirmed using trajectory calculations) with the Geophysica aircraft and with the Cloud-Aerosol Lidar (CALIPSO) satellite (see below) were carried out to allow for a comparison of different particle backscatter measurements and for the observation of cloud changes.

A series of balloon-borne soundings has been carried out in the framework of LAPBIAT-II (Lapland Atmosphere-Biosphere Facility) campaign held at Sodankylä, Finland (360 km East of Kiruna, Sweden) under the premises of Finnish Meteorological Institute's Arctic Research Center (FMI-ARC). A total number of 172 individual balloon instruments including radio and water vapour sondes, aerosol and ozone sensors were flown during the LAPBIAT-II campaign. The balloon flights were coordinated with the aircraft flights and the sounding programme at Sodankylä was carried out in two phases, with 18 payloads flown between 17 January and 6 February 2010 and 15 payloads flown between 10 and 24 March 2010. The stratospheric water vapour measurements were conducted using two types of hygrometers, the Cryogenic Frostpoint Hygrometer (CFH, Vömel et al., 2007) and the Fluorescence Lyman-alpha Stratospheric Hygrometer (FLASH-B, Yushkov et al., 1998). The intercomparison of the data obtained by these two instruments during LAPBIAT-II shows good agreement of $0.73 \pm 5\%$ and consistency in reproducing vertical structures, allowing for the use of FLASH-B and CFH water vapour measurements as interchangeable data series

(Khaykin et al., 2012). During the first phase of LAPBIAT-II the CFH instrument was flown on board of 15 payloads, 11 of which included COBALD backscatter sondes, 5 payloads included FLASH-B sondes and 2 payloads included the backscatter sondes of the University of Wyoming (Rosen and Kjome, 1991).

In coordination with the RECONCILE Geophysica flights, a balloon flight with a remote sensing payload consisting of the instruments MIPAS-B (Michelson Interferometer for Passive Atmospheric Sounding – Balloonborne), TELIS (TErahertz and submillimeter LImbSounder) and mini-DOAS was carried out from Esrange, Kiruna, Sweden on 24 January 2010 (de Lange et al., 2012; Wetzel et al., 2012) to study the diurnal variation of reactive chlorine and nitrogen oxides inside the Arctic vortex from nighttime photochemical equilibrium until local noon covering the full vertical extent of the activated region. Along with $O_3$, $H_2O$ and tracers the complete nitrogen and chlorine families were measured, allowing to study the partitioning and the budgets of these ozone-controlling substances from the tropopause up to about 34 km.

### 3.2.3 Satellite observations

Observations made by a number of satellite instruments have been used extensively to support the flight planning during the field campaigns and in scientific process studies. In the following, the satellite instruments and data products that have been of particular importance for the RECONCILE project are briefly described.

PSCs have been observed since June 2006 by the lidar system on the CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation) satellite, which provides extensive daily measurement coverage up to 82 degrees latitude in both hemispheres. CALIPSO PSCs are detected at 180-m vertical resolution and at horizontal resolutions ranging from 5 km to 135 km, and are classified by composition (supersaturated ternary solution STS, liquid/NAT mixtures, ice, and wave ice) based on the measured 532-nm scattering ratio and aerosol depolarization ratio (Pitts et al., 2009, 2011).

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The Microwave Limb Sounder (MLS) is an instrument on the EOS (Earth Observing System) Aura satellite, which is in a polar orbit of 705 km at an inclination of 98°, and has provided continuous measurements since August 2004 (Waters et al., 2006). MLS provides about 3500 profiles each day from Earth's surface to $\sim 90$ km altitude between 82° N and 82° S. We use observations of gas-phase $HNO_3$ (Santee et al., 2007a), $H_2O$ (Lambert et al., 2007), HCl (Froidevaux et al., 2008), ClO (Santee et al., 2007b), $O_3$ (Froidevaux et al., 2008) and temperature from retrieval version 3.3 (Livesey et al., 2012). In the stratosphere, the vertical resolution and precision of the observations are 3.5–4.5 km and 0.7 ppb for $HNO_3$, 3 km and 15 % for $H_2O$, 3 km and 0.2–0.4 ppb for HCl, 3–4.5 km and 0.1 ppb for ClO, 3 km and 0.04–0.1 ppm for $O_3$, and 4 km and 1 K for temperature

The Atmospheric Chemistry Experiment–Fourier Transform Spectrometer (ACE-FTS) is a solar occultation instrument on SCISAT-1, flying in a circular orbit at 650 km at an inclination of 74° and has made measurements since 2004 (Bernath et al., 2005). ACE-FTS provides daily vertical profiles for up to 15 sunrises and 15 sunsets with latitudinal coverage exhibiting an annual cycle between 85° S to 85° N. We use observations of $ClONO_2$, $N_2O_5$ (Wolff et al., 2008) and HCl (Mahieu et al., 2005) from retrieval version 3 with a vertical resolution of 3–4 km.

The Sub-Millimetre Radiometer (SMR) onboard the Odin satellite observes the thermal emission from the Earth limb and provides vertical trace gas profiles including $O_3$, $N_2O$, and $HNO_3$. Stratospheric mode measurements are generally performed in the altitude range from 7 to 70 km with a resolution of 1.5 km below 50 km tangent altitude and 5.5 km above, normally covering the latitude range between 82.5° S and 82.5° N (Urban et al., 2009; Murtagh et al., 2002).

To assess the impact of the 2011 Arctic ozone hole on surface UV radiation and compare the results to previous Arctic and Antarctic winters (Sect. 6), local-noon clear-sky UV index data products from a combined MSR/SCIAMACHY (Scanning Imaging Absorption Spectrometer for Atmospheric CHartographY) dataset (Eskes et al., 2005; van der A et al., 2010; Allaart et al., 2004) were averaged as a function of day-of-

year and latitude (von Hobe et al., 2012). The comparison of clear-sky data isolates the ozone effect on UV-radiation, i.e. interannual variability due to changes in albedo (snow cover), clouds and aerosol loading is excluded in the analysis.

## 3.3 Microphysical modelling

To understand STS, NAT, and ice formation in the Arctic stratosphere, the Zurich Optical/Microphysical box Model (ZOMM) is used. ZOMM has been described previously by Luo et al. (2003) and Meilinger et al. (1995) and was applied very recently in detailed cirrus modelling (Brabec et al., 2012). Along the lines of previous understanding of PSC formation (Peter, 1997) – kinetic growth of STS by simultaneous uptake of $H_2O$ and $HNO_3$, homogeneous nucleation of ice in these solution droplets (Koop et al., 1995) and then heterogeneous nucleation of NAT on pre-existing ice particles (Carslaw et al., 1998) along air parcel trajectories – the pre-RECONCILE version of the model focussed on simulating exactly these processes. RECONCILE observations triggered the implementation of additional new heterogeneous formation pathways for NAT and ice. To this end, solid nuclei were implemented by assuming the presence of non-volatile particles with a radius of 20 nm and a uniform number density of $7.5\,\mathrm{cm}^{-3}$ throughout the Arctic stratosphere. These assumptions on size and number density reflect the in-situ measurements by the heated condensation nuclei counter COPAS on board of Geophysica during RECONCILE and EUPLEX (cf. Sect. 4.2.2).

These heterogeneous nuclei for NAT and ice formation (Hoyle et al., 2012; Engel et al., 2012b) are then allowed to compete with the accepted pathway of NAT formation via homogeneous nucleation of ice, which typically requires cold localized pools in mountain-wave-driven temperature fluctuations (Carslaw et al., 1998; Peter, 1997). In the absence of better knowledge of the nucleation properties of these nuclei, immersion freezing of NAT and ice was assumed to occur on active sites much like heterogeneous ice nucleation on Arizona test dust demonstrated in previous laboratory experiments (Marcolli et al., 2007). This approach still needs to be reconciled with the laboratory experiments of Biermann et al. (1996), who established upper limits of NAT nucleation

rates and classified direct heterogeneous NAT nucleation on pre-existing solid particles as relatively inefficient.

## 3.4 Chemistry transport modelling

The Chemical Lagrangian Model of the Stratosphere (CLaMS, Konopka et al., 2004; McKenna et al., 2002a,b; Grooß et al., 2005) is a hierarchy of models ranging from a box model to a 3-D Chemistry Transport Model (CTM) based on a Lagrangian transport concept, in which the chemical evolution within individual air parcels is simulated along trajectories determined from ECMWF temperature data and wind fields. Stratospheric chemistry in CLaMS is an update of that described in McKenna et al. (2002a) and now includes reactions of importance in the upper stratosphere. 143 reactions of 45 variable chemical species now explicitly contain reactions involving H radicals as well as $N_2O$ and CFCs. Chemical reaction rates and absorption cross sections are based on JPL 2011 (Sander et al., 2011) except when explicitly noted otherwise. Heterogeneous reaction rates are parameterised as described in Wegner et al. (2012a), and a particle based denitrification scheme (Grooß et al., 2005) is implemented.

For RECONCILE, CLaMS has been used extensively on all spatial, temporal and complexity scales to investigate individual physical and chemical processes using field observations and to develop mathematical parameterisations for use in global prognostic models (cf. below). Hemispheric simulations have been carried out with horizontal resolutions up to 70 km. For the mixing parameterisation (McKenna et al., 2002b; Konopka et al., 2004), a time step of 24 h and a critical Lyapunov coefficient of 1.5 day$^{-1}$ were used (see also Riese et al., 2012). To study transport and mixing processes, and to distinguish between dynamic and chemical processes acting on the concentrations of certain trace gases, dynamical tracers that are not influenced by chemistry such as passive ozone, passive $NO_y$, or artificial air mass origin tracers are used. The evolution of the polar vortex composition during the winter 2009/10 has been investigated utilizing a suite of 9 inert tracers defined by PV thresholds based on an algorithm by Nash et al. (1996). The tracer fields were initialized on 1 December 2009 according

to the initial position of the air masses in the modified potential vorticity (PV) space (see Günther et al., 2008 and references therein). To further quantify the in-mixing of air masses from lower latitudes into the polar vortex during the vortex split in mid December 2009, an additional simulation starting at the beginning of January 2010 with re-initialized tracers was performed.

ATLAS is another global Chemistry Transport Model with a focus on the stratosphere (Wohltmann et al., 2010; Wohltmann and Rex, 2009), based on the same Lagrangian approach as CLaMS. Transport and chemistry are driven by ECMWF ERA Interim data for RECONCILE. The model includes a stratospheric chemistry module that comprises 47 species and more than 180 reactions, detailed heterogeneous chemistry on Polar Stratospheric Clouds and a particle-based Lagrangian denitrification module. For RECONCILE, ATLAS has been used for global modelling studies of the 2009/10 winter (Wohltmann et al., 2012).

## 3.5 Data assimilation

A data assimilation scheme to estimate chemical ozone loss in the Arctic winter stratosphere based on a 3D-Var version of the operational Met Office assimilation system (Jackson, 2007) has been developed by Jackson and Orsolini (2008) to address some potential shortcomings of vortex-averaged and other methods used to estimate ozone loss, namely, the definition of the vortex edge, and estimates of mixing across the vortex edge. Jackson and Orsolini's results showed that the data assimilation method is very promising and can lead to potentially more accurate ozone-loss estimates than other, more established methods.

EOS MLS and SBUV/2 ozone observations are assimilated, together with dynamical observations from satellites, aircraft, radiosondes and surface stations. A realistic ozone field is produced by running the ozone assimilation (referred to as ASSIM) for one week. The ASSIM run is continued for several more weeks, while a reference run (referred to as REF) is also started using the same initial conditions, but continued without further ozone observations being assimilated. Hence in REF ozone is initialized

with EOS MLS and SBUV/2 observations and thereafter transported passively by as-similated winds. The one week initialization step ensures that the reference ozone is realistic and shares the same potential observational biases as the assimilated ozone in the ASSIM run. To estimate chemical ozone loss, REF ozone is subtracted from the
5 ASSIM ozone field. The dynamical fields in both runs are near-identical and differences between the assimilated and reference ozone fields can be attributed to chemistry. In the middle and high-latitude winter lower stratosphere, it is likely that these chemical changes will be overwhelmingly due to heterogeneous PSC-related chemistry. Apply-ing this method for the Arctic winter 2005 between early February and mid March at
10 450 K, Jackson and Orsolini (2008) reported chemical ozone loss of 0.6 ppm, at the lower edge of the 0.6–1.2 ppm range for ozone loss estimates for this period based on other methods.

## 3.6 Global modelling

Global models play a crucial role in understanding the chemical and dynamical pro-
15 cesses in the atmosphere. CCMs provide the only tool for predictions and projections, therefore their validation and the evaluation and improvement of the performance of parameterizations are important parts of the continuous improvement.

   CCM activities within RECONCILE are carried out with the LMDz-Reprobus model (Marchand et al., 2012; Jourdain et al., 2008) that couples the LMDZ general circu-
20 lation model (Hourdin et al., 2006, 2012; Le Treut et al., 1998; Sadourny and Laval, 1984) and the Reprobus stratospheric chemistry package (Lefèvre et al., 1994, 1998). The performances of LMDz-Reprobus simulations of stratospheric ozone and climate have been evaluated against a wide range of observations and against other chemistry-climate models (Austin, 2010; Jourdain et al., 2008; Gettelman et al., 2010; Hegglin,
25 2010). In RECONCILE, the configuration of LMDz (LMDZ5) is identical to that em-ployed for the Coupled Model Intercomparison Project Phase 5 simulations (CMIP5, Hourdin et al., 2012): the model extends from the ground up to 65 km on 39 hybrids pressure vertical levels, and the horizontal resolution is 3.75° in longitude and 1.875° in

latitude. The chemical package provides a comprehensive description of 55 chemical species of stratospheric interest, by means of the usual set of gas-phase and heterogeneous reaction rates recommended by the JPL evaluation (Sander et al., 2011). In its pre-RECONCILE version, the CCM scheme employed for the treatment of polar stratospheric clouds is a simple equilibrium scheme that predicts the existence of liquid (binary $H_2O/H_2SO_4$ or ternary $H_2O/H_2SO_4/HNO_3$ solutions) and solid (NAT or $H_2O$ ice) aerosols from the local thermodynamic conditions in each model grid box. When saturation is reached, solid particles are assumed to form instantaneously at a fixed radius (two modes of 0.5 and 6.5 μm for NAT, 10 μm for water-ice) and only their number density (particles cm$^{-3}$) is variable. As for all equilibrium schemes, the scheme has no memory of the temperature history of the air mass and the particles are not transported separately from one time step to the other. This usual treatment of PSCs in CCMs is computationally cheap but cannot represent properly the slow growth of large solid particles (such as "NAT rocks") and their consequences on denitrification and ozone loss.

The main objective of the global modelling activity in RECONCILE is to assess the impact of the results from the process studies related to the field and laboratory measurements on predictions of polar stratospheric ozone loss. This involves an ensemble of long-term transient simulations covering the period 1960–2050 with different parameterisations of polar stratospheric clouds. The reference simulations of LMDz-Reprobus for the past and future evolution of the ozone layer use the standard reaction rates recommended by JPL 2011 and the pre-RECONCILE PSC equilibrium scheme described above. These simulations also include the same scenarios for the emissions of greenhouse and ozone-depleting gases as those published in the latest WMO assessment (WMO, 2010). On the other hand, the improved version of the CCM takes full benefit of the results of RECONCILE obtained in the laboratory, in the field, and by CTM modelling. In this version, a coherent set of kinetic parameters for ClO$_x$ chemistry that is consistent with observations (Table 3 and Sect. 4.3) is adopted, and a more

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realistic parameterisation of PSCs has been developed based on the results described in Sect. 4.2.

The improved version of LMDz-Reprobus will be integrated over the same timescales as the reference simulation and the impact of the new parameterisations developed from RECONCILE will be evaluated against the observed ozone trends since the 1960s and for the future period. A full analysis of the CCM runs carried out within the RECONCILE framework will be presented in a future paper.

While the last big cross-validation project CCMVal-2 (Eyring et al., 2010; Gillett et al., 2011; SPARC, 2010) provided comprehensive bias estimates for several models, we extended the methodology by comparing dynamical features of empirical data and time series obtained by the LMDZrepro simulations. The main focus was on a global analysis of spectral properties, two-point correlations determined by detrended fluctuation analysis (DFA), and detecting higher order correlations by the spectral surrogate method (see Sect. 5.2).

# 4   Processes in the Arctic vortex

In this section, we summarize the key results from RECONCILE in terms of improved understanding of dynamic, microphysical and chemical processes relevant to polar ozone loss. The field measurements in 2010 provided good opportunities for process studies, even though 2009/10 was a moderate winter in terms of ozone loss. From the match campaign, maximum loss rates on the order of $60\,\mathrm{ppb\,day^{-1}}$ in mid-February and an overall column ozone loss of about 66 DU inside the polar vortex were determined.

## 4.1 Vortex dynamics and mixing

### 4.1.1 The evolution of the 2009/10 Arctic vortex and factors influencing vortex stability

The cooling of the air inside the polar vortex generally followed the 21-yr ECMWF climatological mean from summer until mid-November 2009, when the minimum temperature ($T_{MIN}$) at the 50 hPa pressure surface had dropped to about 200 K (Fig. 7). From this time until mid-December 2009, the minimum temperatures inside the vortex were well above the climatological mean. The period was characterized by significant planetary wave activity (see Fig. 8 in Dörnbrack et al., 2012). As a consequence of these disturbances, the polar vortex split into two unequally strong lobes during the first ten days of December 2009 (Fig. 8, left hand column). The two lobes rejoined, including some mid-latitude air in the process, and the new vortex cooled gradually through mid-January 2010 (second column in Fig. 8). There was a significant drop in $T_{MIN}$ to values as much as 9 K below the climatological mean (Fig. 7). These coldest conditions in the entire winter occurred during a brief period between two sudden stratospheric warmings (SSWs). Indeed, the polar stratospheric cooling and vortex strengthening during this period were a response of the weakened planetary wave activity (Dörnbrack et al., 2012). The formation of such an exceptionally cold vortex corresponds to the development of a positive phase of the Western Pacific (WP) teleconnection pattern in the troposphere, as described by Nishii et al. (2011) and Orsolini et al. (2009). The anomalously cold vortex period was concluded near the end of January 2010 by the onset of a major SSW that was also related to tropospheric features (Ayarzaguena et al., 2011). In early February, the vortex again broke apart into two lobes (third column in Fig. 8) which rejoined again in early March with patches of mid-latitude air that were included in the process. The new single vortex remained intact throughout March (Fig. 8, right hand column). A climatological analysis revealed the surprising result that the 2009/10 winter was the third warmest winter in the 21 yr period from 1989 to 2009 measured by the polar cap temperatures at 50 hPa (see Tables in Dörnbrack et al., 2012).

A comprehensive analysis of SSWs in past Arctic winters also suggests profound tropospheric forcing for the observed major SSWs (Kuttippurath and Nikulin, 2012). The authors also present a statistical analysis for the past 17 Arctic winters showing that ozone loss closely relates to the intensity and timing of major warmings in each winter and argue that a particularly high frequency of major warmings observed in recent Arctic winters could have significant implications for stratospheric ozone trends in the Northern Hemisphere.

### 4.1.2   Mixing within the polar vortex and across its edge

According to the CLaMS artificial tracer simulations, 60 % of vortex air were affected by mixing as a result of the vortex split in December 2009 (Fig. 8, left hand column). After a re-organization before January 2010 the vortex stayed coherent with significant mixing only below 500 K (Fig. 8, second column). Following the second vortex split enhanced transport of air from lower latitudes into the reformed vortex occurred (Fig. 8, third column), leading to a heterogeneous origin distribution inside the vortex interior during late winter (Fig. 8, right hand column). The results from the CLaMS simulations were validated with observations obtained by the CRISTA (CRyogenic Infrared Spectrometers and Telescopes for the Atmosphere) (Kalicinski et al., 2012) and HA-GAR (Hösen et al., 2012) instruments. Analysis of tracer-tracer correlations observed by HAGAR also indicate ongoing irreversible mixing of the intruded air masses inside the vortex in March 2010 (Hösen et al., 2012), which is also captured by the CLaMS simulations.

A detailed case study of model uncertainties in reproducing observed tracer concentrations in a region of strong gradient was carried out for the RECONCILE match flight (Wegner et al., 2010). The comparison of mixing ratios of the tracer $N_2O$ at the start and end points of trajectories connecting the two match flights on 30 January and 2 February revealed that the matches were not always as successful as planned (Fig. 5), and that the determination of matching air masses on the timescale of several days solely by trajectory calculations is insufficient. The mean uncertainty of the trajectory

calculations over three days was 65 km, even when they were based on high resolution ECMWF wind fields. This could potentially impair ozone loss estimates by the match technique in an inhomogeneous vortex.

### 4.1.3 Data assimilation

Issues related to mixing in and at the edge of the polar vortex were also investigated for the assimilation-based method. This was part of a broader study where ozone loss estimates made using the assimilation-based method were compared to ozone loss estimates obtained by running the University of Oslo CTM (Sovde et al., 2011). The focus was on the winter of 2006/07, and the assimilation used 3D-Var, while the CTM constituents were advected using ECMWF analyses produced by 4D-Var. The results indicated that the assimilation-system vertical transport is found to be too fast, which affects the ozone reference field used for the loss estimation, and thus points to an underestimation in the ozone loss estimate. However, horizontal transport errors in the assimilation system were found to be small compared to the vertical transport errors. Improving the ozone reference field, by advecting the Met Office ozone field by the ECMWF 4D-Var winds instead of the Met Office 3D-Var assimilated winds, produced results in better agreement with those produced by the CTM. These results are in agreement with other studies that indicate that stratospheric transport is improved when meteorological analyses are produced using 4D-Var instead of 3D-Var.

## 4.2 PSC evolutions and microphysical processes

### 4.2.1 PSC observations

A large and complementary dataset on PSC properties has been obtained during the RECONCILE winter by ground-based, aircraft and space-borne measurements. The evolution of PSCs during the 2009–2010 Arctic winter is detailed in Pitts et al. (2011). During this unusual winter, more PSCs were observed by CALIPSO than in the

previous three Arctic seasons combined. Examples of CALIPSO PSC observations during the season are shown in Fig. 9. The early period from 15–30 December 2009 was characterized primarily by patchy, tenuous low number density liquid/NAT mixtures (Fig. 9a). No ice clouds were observed by CALIPSO during this early phase, suggest-ing that the NAT clouds were formed through a non-ice nucleation mechanism. From 31 December 2009–14 January 2010, there were frequent mountain wave ice clouds that nucleated widespread NAT particles throughout the vortex (Fig. 9b). Synoptic-scale temperatures in the Arctic fell below the frost point during the period from 15–21 January 2010, which led to a rare outbreak of widespread ice PSCs (Fig. 9c). The final period of the PSC season (22–28 January) was characterized by a major strato-spheric warming that distorted the vortex, displacing the cold pool from the vortex centre. CALIPSO observed primarily STS PSCs during this final period, although NAT particles may have been present in low number densities, but masked from detection by the more abundant STS droplets at colder temperatures (Fig. 9d). The evolution of PSCs in January 2010 observed by CALIPSO was also seen by ground based lidar observations near Kiruna (Khosrawi et al., 2011). PSCs have also been observed by COBALD in every flight within the period from 17 to 25 January 2010. Closely matched measurements between CALIPSO and COBALD agree well in their backscatter pro-files at PSC levels (Fig. 10) and also the agreement between COBALD and the Ny Ålesund lidar is convincing.

The in situ optical particle spectrometers onboard the Geophysica FSSP (Forward Scattering Spectrometer Probe) and CIP (Cloud Imaging Probe) observed PSCs dur-ing the first five RECONCILE flights. Measurements with these instruments show the existence of so-called NAT-rocks up to sizes of roughly 25 µm in diameter, confirming earlier observations (Brooks et al., 2003; Fahey et al., 2001; Northway et al., 2002). The probably first images of NAT-rocks recorded by the greyscale CIP (optical array probe) provide an additional proof of their existence with an optical detection technique other than forward scattering. Those images were observed exclusively when the pen-etration of NAT fields during a RECONCILE flight was confirmed also by the MAL

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(Miniature Aerosol Lidar), MAS (Multiwavelength Aerosol Scattersonde), and SIOUX (StratospherIc Observation Unit for nitrogen oXides) instruments (cf. Table 2). The CIP greyscale image data showed mainly 2–4 pixel features which correspond to particles of roughly 15 µm to 30 µm in diameter. Larger particles were not detected. The images recorded during a PSC penetration on 25 January 2010 over a time period of 30 min amounts to a number concentration of about $10^{-3}$ cm$^{-3}$, which is in the same range as measured by the FSSP. The NO$_y$ instrument (SIOUX) capable of resolving large NO$_y$–containing particles supports the optical data, including the assumed composition of the particles and consequently the assumption of the refractive index of NAT for the processing of FSSP data. Overall, the high number concentration of large NAT particles amounts to condensed phase HNO$_3$ concentrations approaching the theoretical limit of available NO$_y$, raising the question whether the composition of these particles is exclusively NAT. Alternatively, non-spherical shapes of the NAT particles might contribute to a certain overestimation in the particle size and therefore in the total volume of the particle phase. Figure 11 shows a size distribution averaged over a period of 22 min that was measured by the FSSP-100 instrument in one of the dense NAT fields. This size distribution is compared to the theoretical size distribution that Fahey et al. (2001) retrieved from their NO$_y$ measurements made in 2000.

Beside the large NAT particles, the FSSP-300 detected a particle mode around 0.5–1 µm which can be attributed to STS particles. It shows volume to temperature behaviour close to the one measured (Dye et al., 1992) and modelled for STS (Peter, 1997). Throughout all proven PSC penetration events, the particles larger than about 1 µm in diameter were detected only at ambient temperatures below the NAT equilibrium temperature ($T_{NAT}$), but mostly well above the frost point ($T_{ICE}$) so that pure ice particles can most likely be excluded. Only at a few short parts along some of the flight tracks ambient air temperature just fell down to $T_{ICE}$. There was no indication for the presence of ice from the observations of gas phase and total water by FLASH (FLuorescent Airborne Stratospheric Hygrometer) and FISH (Fast In situ Stratospheric Hygrometer). Consistent with the in situ observations, the presence of ice-particles at

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or below the flight altitude is not supported by the spectral signatures seen by CRISTA-NF. Spectra simulated for in situ measured particle size distributions (similar to the one shown in Fig. 11) compare reasonably well with the observed IR limb spectra under the assumption that the large particles consist of NAT (Griessbach et al., 2012; Kalicinsky et al., 2012).

The series of balloon-borne measurements of water vapour and aerosol acquired within the LAPBIAT-II campaign in Sodankylä during the second half of January revealed an unprecedented evidence of water redistribution in the Arctic stratosphere as a consequence of ice PSC formation and concurrent irreversible dehydration by up to 1.5 ppm within 20–24 km altitude range followed by a rehydration in a 2 km thick layer below. The source region and the spatiotemporal evolution of the dehydrated air masses were well established using AURA MLS observations (Khaykin et al., 2012). For the first time, an active phase of ice PSC formation was captured by the simultaneous balloon measurements of water vapour and aerosol on 17 January, providing a unique high-resolution snapshot of repartitioning of water vapour into ice particles. This observation, bearing important implications for the PSCs formation thresholds, is analyzed in detail by Engel et al. (2012a) using microphysical box modelling.

### 4.2.2 Characterisation of background aerosol

The measurements with the COndensation PArticle counting System (COPAS, Weigel et al., 2009) during the RECONCILE winter agree well with the data from Curtius et al. (2005). While outside the polar vortex, the fraction of non-volatile (at 250 °C) particles remains nearly constant with altitude, it increases noticeably with altitude inside the vortex, reaching up to 80 % of detected sub-micron refractory particles deeply inside the Arctic vortex ($\Theta \approx 470$ K).

Nevertheless, the hypothesis by Curtius et al. (2005) that the enhanced non-volatile aerosol in the polar vortex mainly consists of meteoritic smoke particles needs further refinements. Weigel et al. (2012) present a detailed chemical analysis of the refractory aerosol material from 13 (one for each flight) in-vortex particle samples that

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were collected using a miniaturized impactor system placed downstream of the COPAS aerosol inlet, generally at $\Theta > 410$ K. Subsequently, the samples are processed by using offline individual particle analysis with Environmental Scanning Electron Microscopy (ESEM) and Energy Dispersive X-ray (EDX) analysis. Many of the samples contained

5  spheres and aggregates of globules (like "a bunch of grapes", Bigg et al., 1970) in the size range 500 nm–2 µm, chemically dominated by metallic species such as Al, Cr, Mn, Fe or Ni. The chemical composition and morphology indicates that these particles most likely originate to a large extent from space debris. Apart from these metallic particles, also fractured alumo-silicate particles, showing the character of crustal material, were

10  found. In many samples for smaller particles, i.e. $d_p < 500$ nm, also a high number of soot and lead-bearing particles were found, indicating anthropogenic origin. Based on these results, metallic particles (from meteoritic or space debris ablations) are not the major compound of the vortex refractory aerosol material. In particular the fact that surface aerosol material and even anthropogenic particles were found in the subsiding

15  air masses over the Arctic in winter is an essential new finding. It indicates efficient transport pathways for aerosol particles to reach elevated atmospheric altitude, i.e. the stratosphere or even in the mesosphere, to re-enter the lowermost stratosphere due to the developing polar vortex. It is conceivable, that the abundance of non-volatile particles (up to 80 %) in the Arctic winter stratosphere significantly aids the formation

20  of PSCs, and furthermore influences the tropospheric cloud formation after the polar vortex has dissolved in the Arctic spring season.

### 4.2.3   New insights into particle nucleation and PSC formation

The CALIPSO observations during the RECONCILE winter were analyzed in detail to investigate the heterogeneous nucleation pathway of NAT and ice on pre-existing

25  solid particles. The presence of an ample supply of such nuclei in the polar vortex is justifiable as described in Sect. 4.2.2.

For every distinct PSC period defined by Pitts et al. (2011) and described in Sect. 4.2.1, optical parameters simulated by ZOMM (cf. Sect. 3.3) were compared to

selected CALIPSO PSC observations. The best agreement between model results and observations is achieved by (1) allowing NAT and ice to nucleate heterogeneously, and (2) superimposing small scale temperature fluctuations onto the trajectories (which otherwise remain unresolved along the ECMWF trajectories). Only the combination of both effects can provide good agreement in particle types and captures the backscatter and depolarization ratios measured by CALIPSO (Hoyle et al., 2012; Engel et al., 2012c). Figure 12 shows the new scheme of PSC formation and phase transitions that will be presented and discussed in detail in two upcoming papers (Hoyle et al., 2012; Engel et al., 2012b).

At first sight these results appear to be at odds with the previous laboratory-based understanding of NAT nucleation, namely that ice would be the first solid, nucleating homogeneously within liquid aerosol particles (Koop et al., 1995), and that NAT nucleated only subsequently on ice (Carslaw et al., 1998). In these days direct heterogeneous NAT nucleation on pre-existing solid particles was thought to be unlikely (Biermann et al., 1996). Within RECONCILE we presently undertake additional laboratory experiments on meteoritic material. Preliminary experiments corroborate the early results of Biermann et al. (1996), but also seem to suggest that the heterogeneous NAT nucleation rate may indeed reach the upper limits specified by Biermann and colleagues. With today's new insights into low number density NAT clouds, as observed by CALIPSO in December 2009, and NAT-rocks, described by Fueglistaler et al. (2002), nuclei consisting of coagulated meteoritic smoke particles or micrometeorites become favourable candidates to explain those observations.

### 4.2.4 Vertical redistribution of $NO_y$

One of the RECONCILE objectives is to understand the processes causing the vertical $NO_y$-redistribution, i.e. denitrification above about 19 km altitude and a renitrification below.

In 2009/10, Odin/SMR observations showed the strongest denitrification since the start of the Odin measurement period in 2001. However, this was exceeded in the

2010/11 Arctic winter (cf. Sect. 6). Box model simulations together with Odin/SMR, Aura-MLS and CALIPSO observations of $HNO_3$, water vapour and PSC particles respectively show that the strong denitrification observed in mid January 2010 is linked to an unusually strong synoptic cooling event in mid January with ice particle formation on NAT as a possible formation mechanism. The denitrification in the beginning of January was more likely caused by the sedimentation of NAT particles that formed on mountain wave ice clouds (Khosrawi et al., 2011).

During the aircraft campaign, $NO_y$ compounds were observed in situ by the SIOUX instrument and also by remote sensing using MIPAS-STR and CRISTA-NF (Woiwode et al., 2012b; Ungermann et al., 2012). $HNO_3$ observations from the ACE-FTS satellite instrument are also available for the entire winter.

One critical parameter determining the rate and extent of vertical $HNO_3$ redistribution in the polar vortex is the nucleation rate for NAT particle formation. A CLaMS simulation of denitrification was performed in a Lagrangian way using particle trajectories that follow individual NAT particles. Previous simulations (Grooß et al., 2005) that assumed globally constant nucleation rates for all locations with $T < T_{NAT}$ produced reasonable results for the winter 2002/3 using a nucleation rate of $7.8 \times 10^{-6}\,cm^{-3}\,h^{-1}$. Using the same nucleation rate, the observed vertical $NO_y$ redistribution in the winter 2009/10 was underestimated, suggesting faster NAT nucleation. In CLaMS simulations with the new nucleation rate parameterisation described in Sect. 3.3, the vertical redistribution of $NO_y$ compares better to the observations, but still underestimates the observed denitrification in early March.

Significant $HNO_3$ enhancements attributed to renitrification processes were observed by MIPAS-STR and CRISTA-NF in the polar vortex after the end of the PSC phase in the Arctic winter 2009/10. Standard retrieval results from MIPAS-STR showing vertical distributions of $HNO_3$ versus potential temperature are presented in Fig. 13 for the flight on 30 January and for two flights in March (vortex air identified by measured tracer distributions). A significant $HNO_3$ maximum peaking around 405 K is identified for the flight in January directly after the PSC phase of this winter. The redistribution of

$HNO_3$ as a source for $NO_2$ is closely linked to the deactivation of active chlorine species into $ClONO_2$. A dedicated study is carried out involving refined MIPAS-STR retrievals and CLaMS simulations considering the new temperature-dependent NAT nucleation parameterisation to verify whether the representation of $HNO_3$ redistribution in CLaMS
[5] is improved and whether $ClONO_2$ formation is simulated adequately (Woiwode et al., 2012a).

## 4.3 Stratospheric halogen chemistry and catalytic ozone depletion

### 4.3.1 Heterogeneous chlorine activation

As expected, the cold vortex temperatures and extensive PSC coverage in December
[10] 2009 and January 2010 led to substantial chlorine activation. A broad activated layer of enhanced ClO between 16 and 26 km altitude was detected by MIPAS-B on 24 January (Wetzel et al., 2012), corresponding nicely to the vertical extent of PSCs/cold temperatures observed by MIPAS-B in the same data set, as well as CALIPSO (Pitts et al., 2011) and ground based lidars (Khosrawi et al., 2011) in the time period preceding the
[15] flight. $ClONO_2$ observations indicate no or very little activation in the air masses below and above this layer.

The RECONCILE flights on 20, 22 and 24 January showed temperatures where chlorine activation would be expected and were analysed by CLaMS simulations along seven-day backward trajectories (chemistry was initialised from a hemispheric CLaMS
[20] simulation, cf. Sect. 3.4). Up to 1 ppb of additional chlorine activation was produced, and the simulated $ClO_x$ values are consistent with HALOX (HALogen Oxide monitor) observations along the corresponding flight tracks (Fig. 14). Virtually no difference in chlorine activation exists between simulations employing the full PSC scheme with the aerosol surface area density (SAD) increasing with STS, NAT and ice formation and
[25] simulations keeping the SAD at background levels, demonstrating that at sufficiently low temperatures heterogeneous chemistry on the cold binary background aerosol is sufficient to produce the same amount of active chlorine as the full PSC scheme for the

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case considered (Wegner et al., 2012a). The same paper describes another CLaMS investigation for an episode in the Arctic winter 2004/5 constrained by aircraft and satellite data. Significant chlorine activation was produced by CLaMS along 96 h trajectories leading up to a flight on 7 March 2005 (von Hobe et al., 2006) with the resulting $ClO_x$ levels comparing well to the observations. Temperatures were low enough for chlorine activation to occur but significant denitrification probably prevented the formation of NAT and STS, so that chlorine activation occurred to a very large extent on cold background binary aerosol.

Wegner et al. (2012a) corroborate the conclusions of their CLaMS investigations by an analysis of Aura-MLS observations for seven Arctic winters (2004/5–2010/11), focusing on HCl as an indicator for chlorine activation and $HNO_3$ as an indicator for the formation of PSC particles. Below 550 K potential temperature, the vortex average removal rates of gas-phase HCl and $HNO_3$ are not correlated. By 1 January, the vortex average shows that for all considered winters HCl has been removed from the gas-phase to a similar extent while $HNO_3$ shows great interannual variability. This indicates that at these altitudes the additional surface area provided by PSCs does not directly influence the rate of chlorine activation. However, at higher altitudes, where the background aerosol begins to thin out, ice PSCs may play an important role. Between 600 and 650 K, the maximum HCl depletion is observed in the winter 2009/10, which was unique in terms of occurrence of ice PSCs. Other than in the Arctic, HCl depletion cannot be used as an indicator for chlorine activation in the cold vortex core of the Antarctic, where condensation of HCl into STS presents a viable pathway to sequester HCl from the gas-phase. This has been demonstrated by Wegner et al. (2012c) who argue that during the polar night a major part of HCl is dissolved in STS particles, because heterogeneous reactions, especially with $ClONO_2$, cannot explain the observed loss of gas-phase HCl due to shortage of $NO_x$ even if the photolysis of condensed phase $HNO_3$ is taken into account as a potential additional $NO_x$ source.

A comprehensive set of sensitivity runs to examine the impact of known uncertainties in heterogeneous chemistry and PSC microphysics was carried out using the ATLAS

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model (Wohltmann et al., 2012). The simulations were compared to MLS and ACE-FTS observations and in-situ measurements onboard the Geophysica during the aircraft campaign. Results show that ozone loss is a surprisingly robust quantity with regard to changes in the microphysical assumptions with column ozone loss never varying more than 10 % in all sensitivity runs (Fig. 15, right panel). In agreement with Wegner et al. (2012a), chlorine activation on liquid ternary aerosols alone and even on the binary background aerosol was sufficient to explain the observed order of magnitude and morphology of the abundances of chlorine, reservoir gases and ozone (Fig. 15). Assuming default values for NAT number density and supersaturation, runs with different rates for the heterogeneous reactions $ClONO_2 + HCl$ and $ClONO_2 + H_2O$ on liquid aerosols (Hanson and Ravishankara, 1994; Shi et al., 2001) and on NAT (Carslaw and Peter, 1997; Carslaw et al., 1997; based on laboratory studies by Abbatt and Molina, 1992; Hanson and Ravishankara, 1993) produce very similar results. Only if an appreciable NAT surface area density is assumed and the reactions are not dominated by liquid aerosols, the rate and degree of chlorine activation becomes significantly smaller with the rates based on Abbatt and Molina (1992).

Figure 16 summarizes the rate of the most important heterogeneous chlorine activation reaction, $HCl + ClONO_2 \rightarrow Cl_2 + HNO_3$ on different particle types as a function of temperature. Above about 192 K, binary aerosol can activate chlorine just as fast as NAT. Below 192 K, the activation rate increases along a smooth transition from binary to ternary aerosol. Below the frost point, ice PSCs can increase the activation rate again by about a factor of 10.

### 4.3.2  $ClO_x$ catalysed ozone destruction

The UV/Vis spectrum (Young et al., 2012) obtained from the laboratory experiment described in Sect. 3.1 agrees well with the one reported by Papanastasiou et al. (2009) that has been adopted for the most recent JPL recommendation (Sander et al., 2011). Furthermore it is in close agreement with all the ClOOCl spectra which appeared subsequent to Pope et al. (2007), namely: Chen et al. (2009), Lien et al. (2009), Wilmouth

et al. (2009), Jin et al. (2010) and the spectrum reported by von Hobe et al. (2009) which is scaled to the Lien et al. (2009) measurement. It is clearly not in agreement with the Pope et al. (2007) results which likely suffered from subtracting too much $Cl_2$ from the measured composite $Cl_2$/ClOOCl spectrum. The measurement of the much weaker visible spectrum of ClOOCl is the first time this spectrum has been measured. It provides important information to constrain the long wavelength absorption of ClOOCl and hence the impact of this absorption on $ClO_x$ catalysed polar $O_3$ cycles.

Results from the laboratory experiment are consistent with atmospheric observations made during the self-match flight (cf. Table 2) published by Sumińska-Ebersoldt et al. (2012). The observed increase of ClO after sunrise is consistent with the Papanastasiou et al. (2009) cross sections, and essentially rules out ClOOCl absorption cross sections lower than the spectrum published by von Hobe et al. (2009) scaled to absolute cross sections near the peak of the spectrum (246 nm) observed by Lien et al. (2009). The field measurements do not support an additional ClOOCl absorption band of atmospheric significance in the visible region of the spectrum. Remaining uncertainties in the ClOOCl cross sections influence $J_{ClOOCl}$ approximately on the same order of magnitude as parameters influencing the actinic flux (Fig. 17). The presence of PSCs can have a particularly strong influence on the actinic flux and photochemistry. Wetzel et al. (2012) showed that, during the balloon flight on 24 January, the onset of ClO production from ClOOCl photolysis was delayed by about 45 min after sunrise in the stratosphere due to the occurrence of stratospheric clouds.

The thermal equilibrium constant $K_{EQ}$ reported by Plenge et al. (2005) is supported by HALOX ClO and preliminary ClOOCl observations made before sunrise during two RECONCILE Geophysica flights (Stroh et al., 2012). The higher $K_{EQ}$ currently recommended by JPL (Sander et al., 2011) would imply unrealistically high total stratospheric chlorine ($Cl_y$) around 5 ppb to be consistent with the ClO observations (Suminska-Ebersoldt et al., 2012).

Table 3 provides parameterisations for $ClO_x$ kinetics that consistently and reasonably accurately simulate atmospheric observations of $ClO_x$ partitioning and ozone loss. No

indication was found for alternative $ClO_x$ chemistry that was suggested during the time of the RECONCILE proposal (e.g. Salawitch et al., 2009; Vogel et al., 2006).

### 4.3.3 Bromine chemistry

The bromine budget and photochemistry – important for the rate of the ClO–BrO–cycle – was also investigated by aircraft and balloon observations during RECONCILE. First on 7 and 8 September 2009 the stratospheric photochemistry of bromine was probed for the high latitude fall circulation turn-over, where in the stratosphere major reactive bromine species are at daytime BrO and at night time $BrONO_2$. By observing the transition of BrO into its night-time reservoir gas $BrONO_2$ at dusk and dawn with direct sun solar occultation and Limb spectroscopy and by radiative transfer and photochemical modelling, Kreycy et al. (2012) found that the ratio of the photolysis rate of $BrONO_2$ $J_{BrONO_2}$ to the three body formation reaction ($BrO + NO_2 + M$) rate coefficient of $BrONO_2$ $k_{BrO+NO_2}$, i.e. $J_{BrONO_2}/k_{BrO+NO_2}$, is by a factor $1.69 \pm 0.04$ larger than recommended by the most recent JLP-2011 compilation (Sander et al., 2011). Major consequences of the findings for the stratosphere are that (1) recent assessments of total stratospheric bromine ($Br_y$) using the inorganic method for high stratospheric $NO_x$ loadings may have overestimated the necessary correction for the BrO to $Br_y$ ratio (for this flight, $Br_y$ should be 1.4 ppt or 6.8 % of the total lower using the revised $J_{BrONO_2}/k_{BrO+NO_2}$ ratio as compared to the JPL-2011 recommendations), and (2) a larger $J_{BrONO_2}/k_{BrO+NO_2}$ ratio may cause a small increase in the bromine-mediated ozone loss in the stratosphere (maximum −0.8 %, see Fig. 18).

The trend of total stratospheric bromine (Fig. 1.21 in WMO, 2010) was also continued with data collected during RECONCILE, auxiliary field campaigns performed for example in 2008 in the tropics, at high latitudes in fall 2009, in winter 2010 and spring 2011, and lately during the EU project SHIVA (Stratospheric Ozone: Halogen Impacts in a Varying Atmosphere) in the Western Pacific in fall 2011. The collected data indicate that from its peak in the early 2000s, $Br_y$ continues to decrease by about 10 % per

decade and may now have reached about 19 ppt in 4 yr old air. Together with the finding of Kreycy et al. (2012) the collected field data, in particular during SHIVA, indicate that the contribution of the so-called very short-lived species (VSLS) inferred using the inorganic stratospheric method may be lower than stated in WMO (2010), i.e. $4 \pm 2.5$ ppt instead of 6 (range 3–8) ppt. The former estimate is also in better agreement with past and recent estimates of VSLS based on the organic bromine method, which indicate [VSLS] = $2.25 \pm 0.25$ ppt, where 90 % is due to contributions from $CH_2Br_2$ and $CHBr_3$ (e.g. Brinckmann et al., 2012).

### 4.3.4 Halogen source gases

A number of important long-lived brominated and chlorinated source gases (i.e. the three major CFCs and HCFCs, the two major halons, $CCl_4$ and $CH_3CCl_3$) were sampled by the Whole Air Sampler (WAS) onboard the Geophysica and subsequently analysed in the laboratory. Together with age-of-air measurements inferred from $SF_6$, the results have been used for a re-evaluation of the stratospheric lifetimes of these compounds as well as their fractional release (i.e. the fraction that has released its halogens) and Ozone Depletion Potentials (ODPs), all of which are policy-relevant quantities. As outlined in Laube et al. (2012b) there are indications for considerably smaller fractional release for some of these gases. Alongside with possibly longer lifetimes this would not only lead to smaller ODPs in many cases, but also has the potential to significantly delay the recovery of the ozone layer. WAS measurements of other climate-relevant trace gases have improved the knowledge of their atmospheric distributions (e.g. Laube et al., 2010b, 2012a; Oram et al., 2012).

## 4.4 Studies of the Antarctic ozone hole

### 4.4.1 Minimum ozone mixing ratios

Although the main focus of RECONCILE is on Arctic ozone loss, the same processes are at play in the Antarctic, and there is still a lot to be learned from studies looking at the much more severe and annually reproducible Antarctic ozone hole. An investigation was carried out with CLaMS to explain the extremely low ozone mixing ratios below about 10 ppb observed by ozone sondes in Antarctic spring (Solomon et al., 2005). While in principle the mechanisms of ozone depletion are largely known, it was not clear what controls the ozone and chlorine chemistry causing such low ozone concentrations and what determines the lowest ozone values that can be reached. The ozone observations since the 1990s show a very similar behaviour from year to year, that is a minimum value changing with the time of year. This lower envelope of the observations reaches a minimum in late September and increases again in October and November. The complex interaction between gas-phase and heterogeneous chemistry under these conditions is described in detail by Grooß et al. (2011). To correctly simulate the behaviour of the ozone observations, it is necessary to include the oxidation chains of CO and $CH_4$, especially the reaction of the methyl peroxy radical $CH_3O_2$ with ClO (Crutzen et al., 1992) is important here.

### 4.4.2 Ozone trends

A long-term assessment of ozone loss and ozone trends in Antarctica has recently been published by Kuttippurath et al. (2012b). It reveals a slow but significant increase of the September–November average column ozone since the late 1990s, indicating that ozone recovery may have indeed started in the Antarctic.

## 5 Progress in global chemistry climate modelling

### 5.1 Amendments to the model

Based on the results described in Sect. 4, the following updates were implemented in the LMDZ-Reprobus CCM:

- A new PSC scheme is used with an improved parameterization of the growth of NAT particles. This new scheme includes a tracer keeping the memory of the time spent under the theoretical temperature of NAT formation, allowing a more realistic slow growth of NAT particles, rather than the instantaneous formation of large particles previously used in the so-called equilibrium scheme of the CCM. Maximum radius and number density of liquid and solid PSC particles are tuneable parameters.

- Based on the results described in Sect. 4.3.3., no new reactions driving catalytic ozone loss were included. $ClO_x$ parameterisations from Table 5 were adopted as opposed to the reference model that uses parameterisations from Sander et al. (2011).

In addition to LMDZ, scientific findings from the RECONCILE project have been incorporated in the Whole Atmosphere Community Climate Model (WACCM). PSC processes have been improved in WACCM to give a better representation of mixed PSCs and the formation threshold for ice PSCs has been modified to agree with observations (Wegner et al., 2012b).

### 5.2 Model evaluation

Before carrying out long-term CCM simulations, the new PSC scheme developed during the project was tested with the CTM version of REPROBUS driven by ECMWF analysis for the Arctic winter 2009/10. The results obtained with this new microphysical parameterisation show a clearly improved $HNO_3$ distribution when compared to Aura

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MLS measurements, with less denitrification than with the reference model. The impact of this effect on ozone and climate over long timescales now needs to be investigated with the CCM version.

The scanning algorithms necessary for data analysis on global scales and the de-trended fluctuation analysis (DFA) intended to reveal the correlation properties of to-tal column ozone (TO) time series were implemented successfully and the perfor-mance was cross-checked by reproducing the earlier results on satellite measure-ments (Kiss et al., 2007). While the original analysis used mostly the records of two instruments (Nimbus-7 and Earth Probe), the longer and continuous database (daily TO time series between 1978 and 2011) compiled by Bodeker Scientific (http://www.bodekerscientific.com/) was exploited in the present validation (Struthers et al., 2009). DFA runs on the CCMVal-2 outputs from the LMDZrepro reference simulation (1960–2006) indicate that the two-point correlation properties of model data are almost fully consistent with the measurements (Fig. 19a, b).

As for the spectral analysis, various regions exhibit different behaviours. Three signif-icant peaks show up characterizing a half-year, annual and quasi-biennial oscillations (QBO) with different amplitudes. In order to estimate the importance of these modes, the spectral weight of the peaks were determined by integrating the area under the given peak and normalizing by the total power (total area of the frequency spectrum). As an example, a comparison of the spectral weight distribution of the semi-annual peak for the empirical and simulated data is shown in Fig. 19c, d. Interesting dynamical features in the simulations are the apparent shift and increased amplitude at the equa-torial Pacific region, and a semi-annual oscillation above the Tibetian Plateau missing from the observations.

The QBO along the Equator is apparent in empirical TO data. However it is well known that numerical models can hardly reproduce this dynamical mode at the usual spatial and temporal resolutions. In an extended analysis we compared simulated time series of a CCM model, which is able to reproduce QBO (the Japanese Meteorological

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Research Institute model, MRI, in the CCMVal-2 data bank), but we could easily detect other anomalies in the correlation and spectral properties in the MRI time series.

## 6 Arctic ozone depletion in 2011

In terms of geographical extent and persistence of PSC conditions the winter 2010/11 was one of the coldest Arctic winters on record (Fig. 7), and measureable ozone loss rates persisted longer into spring than in any previous winter and exceeded the maximum rates previously measured in the Arctic. Based on Aura Satellite observations (OMI and MLS) as well as results from the 2010/11 match campaign, Manney et al. (2011) have shown that the vertical loss profile is mostly within the range of Antarctic ozone losses and far outside the range of previous ozone loss in the Arctic (Fig. 20). More than 80 % of ozone was lost at the surface subsiding to 460 K by late March. Manney et al. (2011) also provided an in depth investigation of the processes leading to the record ozone loss, highlighting the remarkable degrees of chlorine activation and denitrification. In addition, they presented a comprehensive comparison to the winter 1996/97, which was among the coldest previous Arctic winters. Kuttippurath et al. (2012a) also discussed the dynamical situation in both winters and extended the analysis of Manney et al. (2011) by describing the contribution of $NO_x$ cycle at higher altitudes.

Balis et al. (2011) show that dynamics and in particular weak transport from lower latitudes were important in setting the scene for the record low Arctic ozone in 2011. The respective roles of transport and chemical loss during 2010 and 2011 in simulations with the University of Oslo CTM (Sovde et al., 2011) are compared in Fig. 21. The upper panel shows that in 2010, ozone is replenished more efficiently at high latitudes than in 2011, which results from both isentropic mixing and from transport by the Brewer–Dobson circulation. This indicates that the lack of transport into the high latitudes played an important role in the ozone deficit in 2011. The second panel compares the large relative difference in transport to high latitudes in 2010 vs. 2011 (dotted

line) to the absolute contribution of heterogeneous chemistry to ozone loss in 2011 (full line, signed reversed). While the chemical loss in 2011was much higher than in 2010, the relative weakness of transport into the very stable and narrow vortex was a stronger contributor to the column ozone deficit poleward of 60° N.

Von Hobe et al. (2012a) carried out an analysis of a 30-yr record of surface UV data products from satellite measurements and could show that for Arctic winters with strong ozone depletion including 2011, increases in surface UV radiation did not reach proportions like in the Antarctic. The enhancements were most significant at high latitudes (> 60° N) in March and early April, when absolute UV levels are still moderate. At lower latitudes, where UV increases pose a more serious threat to human health and ecosystems, low column ozone due to transport from the subtropics has a stronger effect on surface UV than spring Arctic ozone loss.

## 7  Conclusions

RECONCILE has substantially advanced our understanding of the processes destroying ozone in the polar vortex. Three particular achievements are (1) the much improved elucidation of the NAT nucleation process and its impact on denitrification and thus ozone loss in the polar vortex, (2) the gathering of substantial evidence for the importance of heterogeneous chlorine activation on cold binary aerosol, and (3) the reconciliation of kinetic parameters governing $ClO_x$ catalysed polar ozone loss. It should be mentioned that many other substantial contributions besides RECONCILE have been made in the past few years, e.g. a recent SPARC initiative (Kurylo and Sinnhuber, 2009) and related laboratory studies to investigate ClOOCl photolysis (Lien et al., 2009; Papanastasiou et al., 2009; von Hobe et al., 2009). The representation of polar ozone depletion and related processes in global models has also been improved (WMO, 2010, and references therein).

But besides helping to reach closure with respect to some existing uncertainties, some of the RECONCILE results also raised new questions. The observation of the

coldest Arctic winter on record in 2010/11 prompting an unprecedented degree of Arctic ozone loss (Manney et al., 2011) and related increases in UV radiation (Bernhard et al., 2012; von Hobe et al., 2012a), and the lack of prognostic tools that could have predicted such an event, made it evident that efforts to understand exactly what drives the stability and interannual variability of the Arctic polar vortex and the development of predictive capabilities are imperative. The finding of a large number of non-volatile particles with sometimes surprising chemical composition inside the Arctic vortex challenges some of our presumptions on how material, and especially aerosols, are transported to and distributed through the stratosphere. This is particularly important if we want to understand and model how climate change might affect the composition of the stratosphere in the future.

These questions concern the preconditions for Arctic ozone depletion rather than the actual ozone destruction process. The answers lie, to a large extent, in the tropics. The tropical tropopause is the main "gate to the stratosphere" (e.g. Fueglistaler et al., 2009) for trace gases and particles, which are then transported to higher latitudes via the Brewer–Dobson (BD) circulation. Changes in the chemical composition of the stratosphere related to climate change are likely, given the impact of climate on tropospheric sources and sinks as well as chemical processing of gases and particles, on convective transport into the tropical tropopause region, and on transport through the stratosphere via the BD. For example, it has been shown by Zhou et al. (2012) that the upwelling branch of the BD circulation is strongly influenced by tropical and subtropical wave activity. Also the turnaround latitudes of the BD circulation are likely governed by an interplay of wave activity at midlatitudes and the tropics/subtropics. Since tropical and subtropical wave activity is expected to increase in a changing climate, this is expected to have an important strengthening influence on the BD circulation (Butchart and Scaife, 2001; Fomichev et al., 2007) with possible influences on ozone depletion at high latitudes. Possibly even more important for ozone loss in the Arctic than potential changes in chemical composition, Naoe and Shibata (2012) pointed out that climate change may lead to changes in the Holton–Tan-mechanism that links the stability of

the polar vortex to the QBO (Garfinkel et al., 2012; Holton and Tan, 1980). Future investigations that address these questions will be needed to complement the work done in RECONCILE and further advance our understanding of Arctic ozone depletion.

Some of these data were available through the special project "Effect of non-hydrostatic gravity waves on the stratosphere above Scandinavia" by one of the authors (A.D.).

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**Table 1.** Overview of RECONCILE flights. All flights were carried out in 2010, take off and landing times are in UTC. The corresponding flight tracks are shown in Fig. 4.

| No | date | take off | landing | Scientific targets |
|---|---|---|---|---|
| F1 | 17 Jan | 11:20 | 15:33 | PSC characterization |
| F2 | 20 Jan | 08:13 | 11:55 | PSC characterization and CALIPSO match |
| F3 | 22 Jan | 10:01 | 13:35 | PSC characterization |
| F4 | 24 Jan | 13:30 | 16:56 | PSCs and renitrification |
| F5 | 25 Jan | 05:50 | 09:19 | PSCs, de/renitrification, Cl active., self-match experiment[1] |
| F6 | 28 Jan | 09:00 | 12:56 | vortex survey (dynamics and mixing) |
| F7 | 30 Jan | 06:36 | 10:15 | self-match experiment[1], Cl activation, de/renitrification |
| F8 | 2 Feb | 10:01 | 13:31 | match experiment[2], Cl activation, de/renitrification, $O_3$ loss |
| F9 | 27 Feb | 12:06 | 15:41 | vortex filament |
| F10[3] | 2 Mar | 02:51 | 06:31 | vortex survey, Cl deactivation, de/renitrification, $O_3$ loss |
| F11[3] | 2 Mar | 09:35 | 13:35 | vortex survey, Cl deactivation, de/renitrification, $O_3$ loss |
| F12 | 5 Mar | 15:05 | 18:36 | vortex filament |
| F13[4] | 10 Mar | 07:14 | 10:51 | test of remote sensing instrumentation, $O_3$ loss |

[1] The self match flight is a particularly useful flight pattern to constrain kinetic parameters. Air masses are sampled twice during the same flight (Schofield et al., 2008). The two RECONCILE self match flights were carried out over sunrise to constrain the ClOOCl photolysis rate $J$ and the ClO/ClOOCl thermal equilibrium constant $K_{EQ}$ (Suminska-Ebersoldt et al., 2012).

[2] Three days after the second self-match flight, the air masses that had been sampled came within reach of the Geophysica again, and a match flight was carried out to test the accuracy of trajectory calculations and to check the consistency of observed and simulated chlorine activation and ozone loss (Wegner et al., 2012).

[3] Flights F10 and 11 were carried out as relay flights with an intermediate stop in Spitsbergen.

[4] Flight 13 was dedicated to test remote sensing instrumentation under the PremierEx project funded by ESA.

**Table 2.** Instrumental payload of M55 Geophysica during the RECONCILE field campaign.

| instrument | parameter | time res. | accuracy | precision | technique | references |
|---|---|---|---|---|---|---|
| **in situ** | | | | | | |
| FOZAN | $O_3$ | 1 s | 0.01 ppm | 8 % | Dye chemiluminescence+ECC | (Ulanovsky et al., 2001; Yushkov et al., 1999) |
| FISH | $H_2O$ (total) | 1 s | 0.2 ppm | 4 % | Lyman-$\alpha$ | (Zöger et al., 1999) |
| FLASH | $H_2O$ (gas phase) | 8 s | 0.2 ppm | 6 % | Lyman-$\alpha$ | (Sitnikov et al., 2007) |
| SIOUX | NO | 1 s | 10 % | 3 % | Chemiluminescence, | (Voigt et al., 2005) |
| | $NO_y$ | 1 s | 15 % | 5 % | + Au-converter | |
| | Particle $NO_y$ | | | | + Subisokinetic inlet | |
| HALOX | ClO | 10 s | 20 % | 5 % | CCRF | (von Hobe et al., 2005) |
| | ClOOCl | 30 s | 30 % | 15 % | + thermal dissociation | |
| HAGAR | $N_2O$, CFC12 | 90 s | 1.3 % | 0.3 % | Gas Chromatography (GC) | (Homan et al., 2010; |
| | CFC 11 | 90 s | 1.6 % | 0.6 % | with electron capture detec- | Werner et al., 2010) |
| | $CH_4$, $H_2$ | 90 s | 1.8 % | 0.8 % | tor (ECD) | |
| | $SF_6$ | 90 s | 2.3 % | 1.3 % | IR absorption | |
| | Halon 1211 | 90 s | 3.0 % | 2.0 % | | |
| | $CO_2$ | 5 s | 0.1 % | 0.05 % | | |
| COLD | CO | 5 s | 9 % | 1 % | TDL | (Viciani et al., 2008) |
| COPAS | Condensation nuclei (CN-total, CN-non-volatile) | 1 s | 10 % | 5 % | 2-channel CN counter, one inlet heated | (Weigel et al., 2009) |
| FSSP | Cloud particle size distrib. (0.4–47 μm) | 1 s | 20 % | 10 % | Laser-particle spectrometer | (de Reus et al., 2009) |
| CCP | Cloud particle size distrib. (3–47 μm) | 1 s | 20 % | 10 % | Laser-particle spectrometer | |
| CIP | Cloud particle size distrib. (25–1600 μm) Particle Images | 1 s | 20 % | 10 % | Laser-particle spectrometer | (Baumgardner et al., 2001) |
| MAS | Aerosol optical properties | 10 s | 5 % | 5 % | Multi-wavelength Scattering | (Buontempo et al., 2006) |
| WAS | Long lived trace gases and isotopo-logues | minutes | 5 % | < 2 % isotopol: 0.1–2 ‰ | Whole air sampling with lab GC and MS analysis | (Kaiser et al., 2006; Laube et al., 2010a) |
| Rosemount probe (TDC) | $T$, $P$ horizontal wind | 0.1 s 0.1 s | 0.5 K 1 m s$^{-1}$ | 0.1 K 0.1 m s$^{-1}$ | PT100, 5-hole probe | |
| **remote sensing** | | | | | | |
| MAL 1 & 2 | Remote Aerosol Profile (2 km from aircraft altitude) | 30–120 s | 10 % | 10 % | Microjoule-lidar | (Matthey et al., 2003) |
| miniDOAS | BrO, OClO | | | | DOAS | |
| MIPAS-STR | Cloud Index, $T$, $HNO_3$,$O_3$,$ClONO_2$, CFCs, $H_2O$ and minor species | typ. 2.4/3.8 min for 1 profile (depends on sampling pro-gramme) | $T$ < 1 K/ vmr typ. 10–15 % | $T$ < 1 K/ vmr typ. 4–12 % | FTIR limb sounder | (Woiwode et al., 2012b) |
| CRISTA-NF | $H_2O$, $HNO_3$, PAN, $ClONO_2$, CFCs, $O_3$, $CCl_4$ | | | | MIR-Emission in limb view-ing geometry | (Ungermann et al., 2012; Weigel et al., 2010) |
| MARSCHALS | $O_3$, $H_2O$, CO, $HNO_3$, $N_2O$ | | | | Millimetre Wave spectrome-ter in limb viewing geometry | (Moyna et al., 2006) |

**Table 3.** Kinetic and photochemical parameters for the ClO-dimer cycle that are most consistent with the experiments and observations made within RECONCILE. Clearly, it does not represent a comprehensive evaluation of kinetic parameters determined in laboratory experiments such as given by Sander et al. (2011).

| | | |
|---|---|---|
| ClOOCl production rate constant $k_f{}^*$ | (Nickolaisen et al., 1994) $2.1 \times 10^{-32} \cdot (T/300)^{-3.01}$ | This is somewhat lower than the current JPL recommendation (Sander et al., 2011), but gives the best fit to field measurements of ClO made during the RECONCILE field campaign in 2010 (Suminska-Ebersoldt et al., 2012) and can be rationalized based on theoretical chemistry calculations (von Hobe et al., 2007) |
| ClO/ClOOCl equilibrium constant $K_{EQ}$ | (Plenge et al., 2005) $1.92 \times 10^{-27} \cdot e^{8430/T}$ | Field measurements of ClO before sunrise in air masses that were shown to be in thermal equilibrium rule out parameterizations that give higher values for $K_{EQ}$ (Suminska-Ebersoldt et al., 2012). Again, this is in good agreement with theoretical considerations by von Hobe et al. (2007) |
| ClOOCl absorption cross sections $\sigma_{ClOOCl}$ | (Papanastasiou et al., 2009) | These published cross sections are consistent with the ones measured in the laboratory as part of RECONCILE and also with ClO observations made during the RECONCILE aircraft campaign |

$^*$ We only consider the low pressure limit for $k_f$, which is much more relevant for stratospheric conditions than the high pressure limit.

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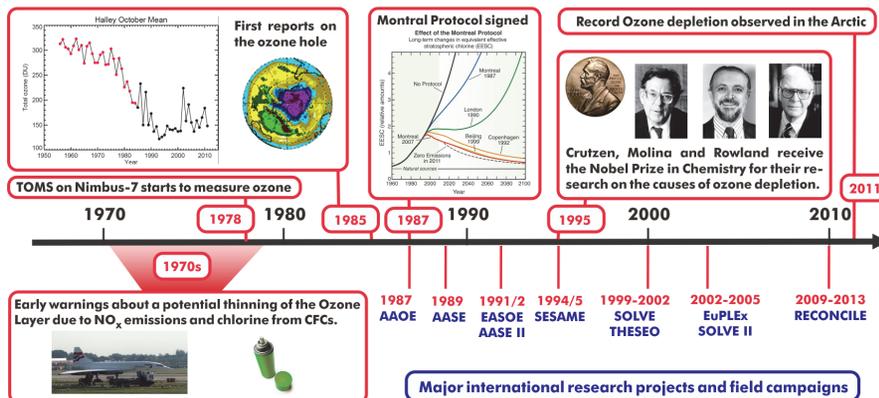
**Fig. 1.** Illustrative timeline of ozone research. More detailed discussions on the history of the discovery of the anthropogenic influence on stratospheric ozone are given by Solomon (1999), Brasseur (2008) and Müller (2009). Parts of this figure are from WMO (2010) and http://www.nobelprize.org, the Concorde photo is courtesy of K.-H. Nogai.

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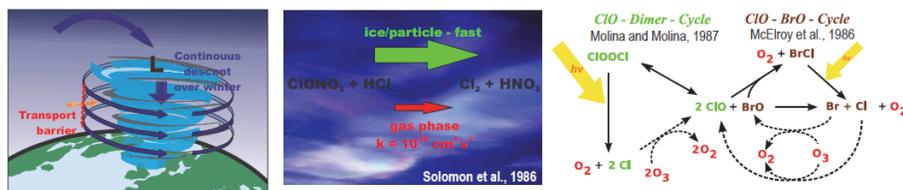
**Fig. 2.** Conditions favouring the formation of an ozone hole in polar regions in spring. **(a)** The isolation of the air inside the polar vortex inhibits dilution of active chlorine and replenishment of ozone (McIntyre, 1989; Proffitt et al., 1989). **(b)** Fast heterogeneous reactions on particle surfaces activate chlorine from reservoir gases (Solomon et al., 1986). **(c)** Ozone is destroyed by catalytic cycles that are particularly efficient under cold temperature/high solar zenith angle conditions (McElroy et al., 1986; Molina and Molina, 1987).
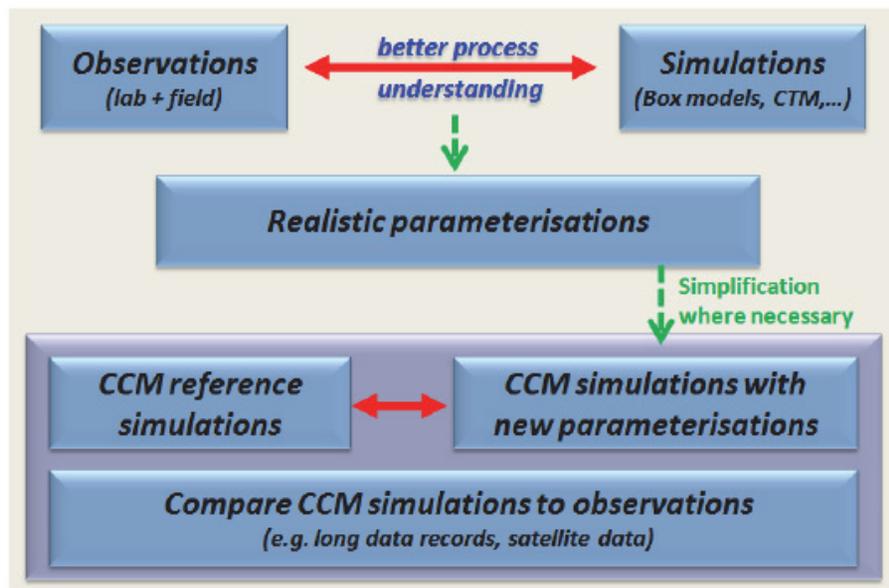
**Fig. 3.** In RECONCILE, a fully integrated approach is adopted that joins detailed process studies including laboratory and field experiments with global chemistry-climate model (CCM) studies.

**Fig. 4.** Geophysica flight paths during RECONCILE. In the left panel, all flights are marked by different colours (referring to flight date, yymmdd). In the right panel, flight sections in vortex air are marked red, flight sections outside the polar vortex are marked in blue. Two self-match flights were carried out on 25 and 30 January, a match flight on 2 February (matching the flight on 30 January).

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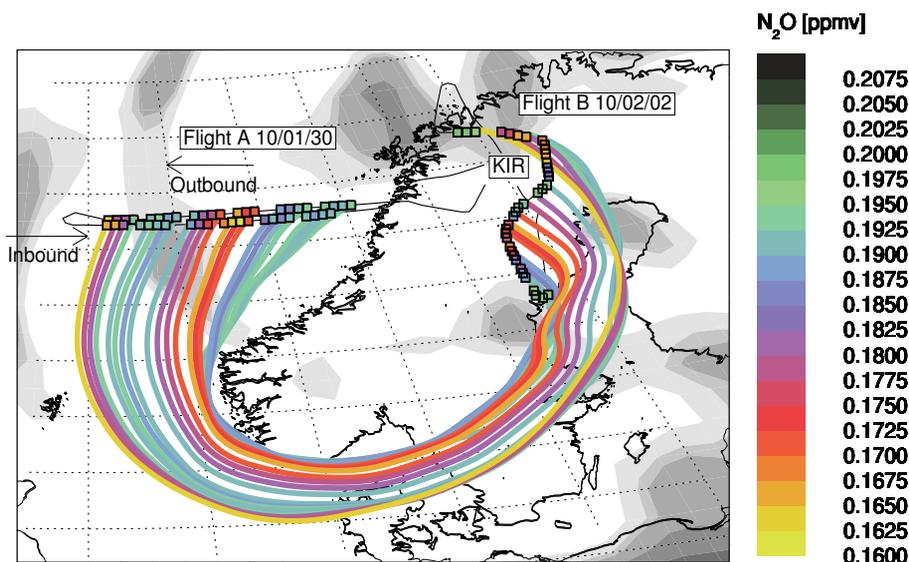
**Fig. 5.** Flight tracks of the second self match flight on 30 January and the match flight on 2 February with HAGAR $N_2O$ measurements along the flight tracks shown by the coloured squares. The trajectories connecting the flights are colour coded according to the $N_2O$ (in ppb) observed during the first flight. The grey contours in the background show PV from ERA-interim at the 425 K level on 30 January, 12:00 UTC, with white representing high PV and darker grey representing areas of low PV pointing to a significant contribution of non-vortex-air.

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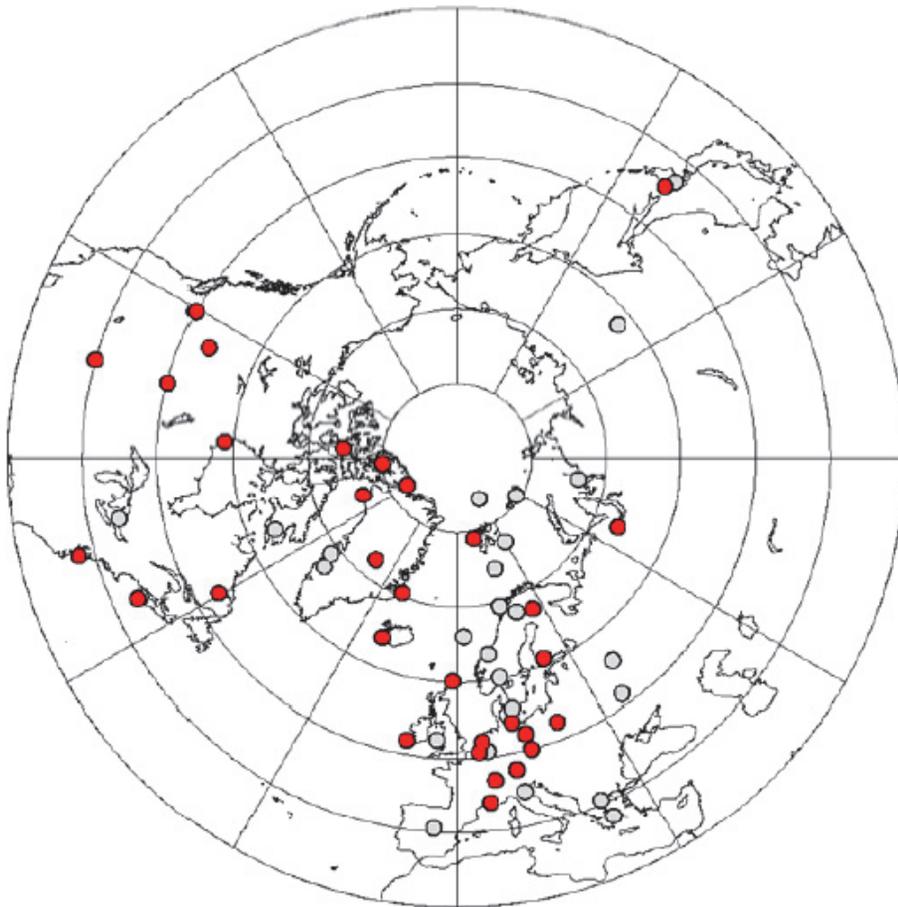
**Fig. 6.** Network of ozone sounding stations that participated in the RECONCILE match campaign (red). Gray dots indicate stations that were active in match campaigns in the past but which went out of operation during recent years due to lack of support.

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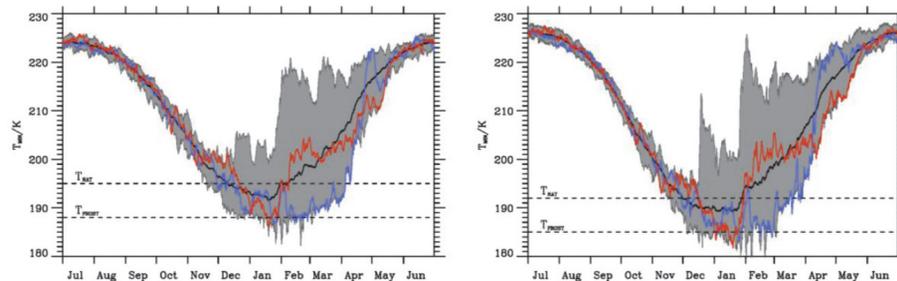
**Fig. 7.** Minimum temperatures $T_{MIN}$ (K) between 65° N to 90° N on the 50 hPa (left panel) and 30 hPa (right panel) pressure surfaces. Black line: mean value from 1989–2011; red line the $T_{MIN}$ evolution from July 2009 through June 2010, blue line from July 2010 through June 2011; shaded area encompasses the minimum/maximum $T_{MIN}$ between 1 January 1989 and 30 June 2012. Source: ECMWF reanalyses interim (ERA Interim) data provided at 6 hourly temporal resolutions, see: http://www.ecmwf.int.
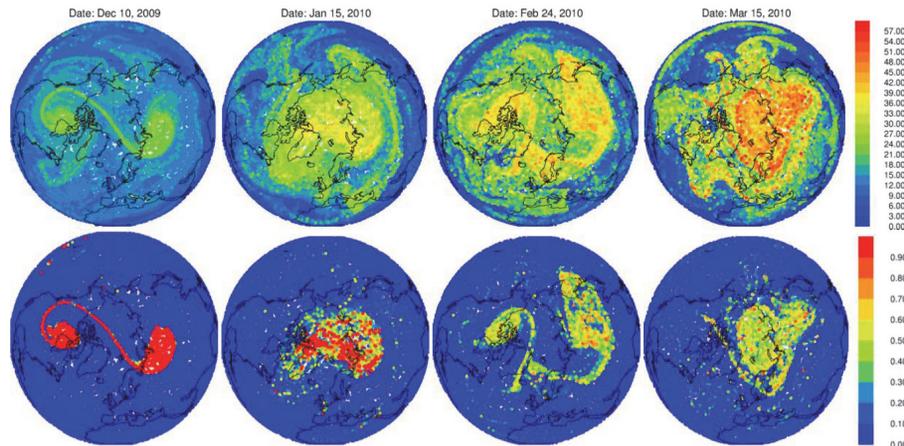
**Fig. 8.** Modified potental vorticity (PV, given in $10^{-6}\,\mathrm{K\,m^2\,kg^{-1}\,s^{-1}}$, upper panel) and inert tracer P4 (indicating the relative contribution of air masses that were inside the vortex core at the time of initialization, lower panel) on 450 K isentropic level during the first vortex split, after re-organization of the vortex, during the second vortex split and at the end of the winter (order from left to right).
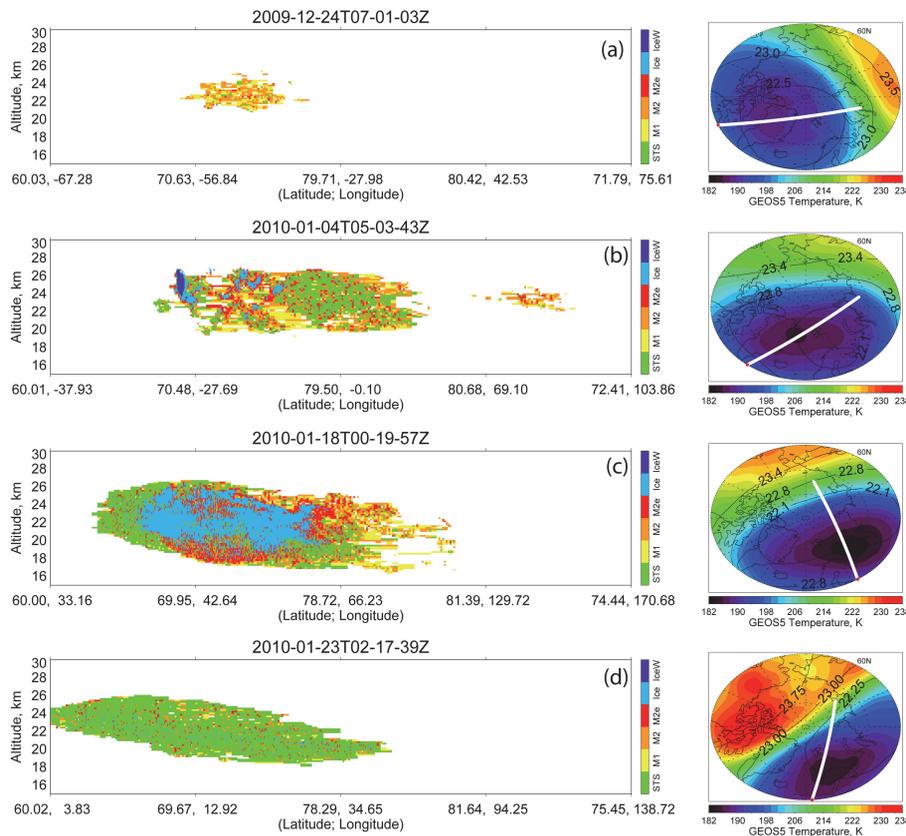
**Fig. 9.** Examples of CALIPSO PSC composition observations during the 2009–2010 Arctic winter. The panels on the left show CALIPSO PSC composition images for representative orbits from four periods during the winter: **(a)** 15–30 December, **(b)** 31 December–14 January, **(c)** 15–21 January, and **(d)** 22–28 January. GEOS5 temperature and geopotential height fields at 30 hPa for 12:00 UTC are shown at the right of each panel with the location of the corresponding CALIPSO orbit track indicated by the white line (red dot marks orbit track start).

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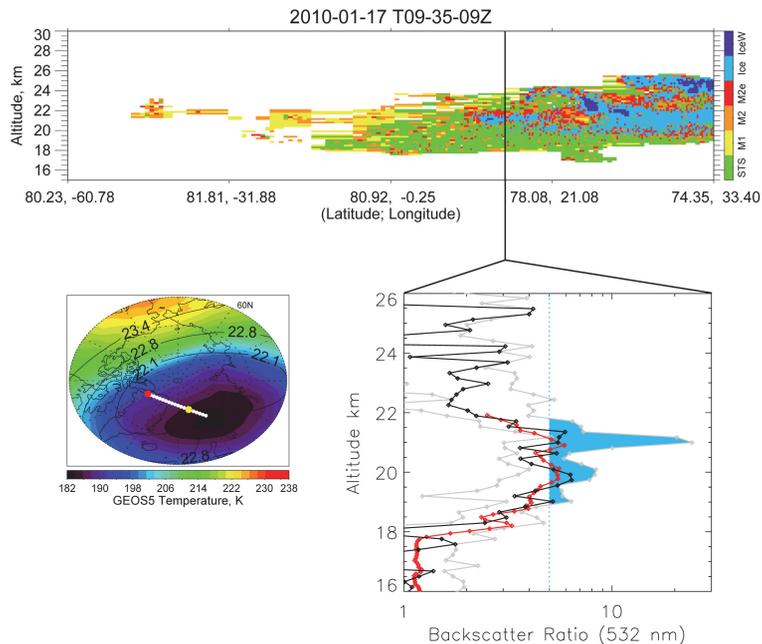
**Fig. 10.** CALIPSO–COBALD comparison on the 17 January 2010. Upper panel: CALIPSO PSC composition observations. The black line denotes the location of the simultaneously performed COBALD sounding. Lower left panel: GEOS5 temperature and geopotential height fields at 30 hPa for 12:00 UTC with the location of the corresponding CALIPSO orbit track indicated by the white line and the position of the COBALD sonde by the yellow dot. Lower right panel: profiles of backscatter measurements from COBALD (red) and CALIPSO (black). According to the composition classification by Pitts et al. (2011) and the definition of ice, backscatter ratios larger than 5 are highlighted in blue. Grey profiles show maximum and minimum backscatter values from CALIPSO within a range of ±25 km around the closest profile indicating the uncertainty between both measurements at 21 km altitude.
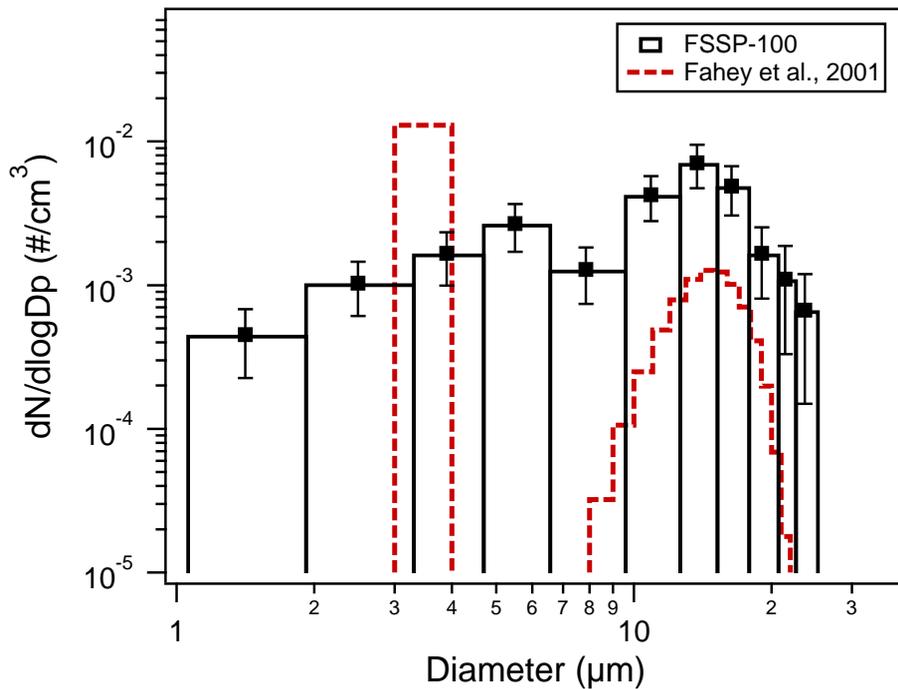
**Fig. 11.** Number size distribution obtained from FSSP-100 measurements during RECONCILE flight on 25 January 2010, averaged over 22 min (from 7:58 to 8:20 UTC). For comparison, the size distribution retrieved from $NO_y$ observations in the Arctic winter 2000 by Fahey et al. (2001) is shown in red.
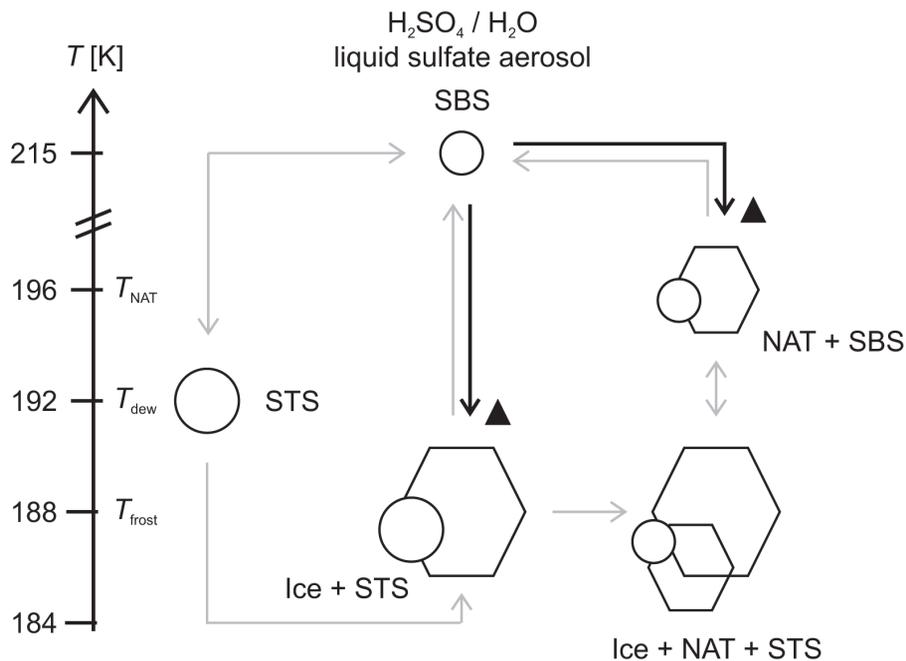
**Fig. 12.** PSC formation pathways as implemented into ZOMM with SBS = Supercooled Binary Solution ($H_2SO_4/H_2O$), STS = Supercooled Ternary Solution ($HNO_3/H_2SO_4/H_2O$), NAT = Nitric Acid Trihydrate ($HNO_3 \cdot 3\,H_2O$), ▲ = solid inclusion, e.g. meteoritic dust. Grey arrows denote pathways included in previous schemes describing PSC formation pathways (going back to Peter, 1997). Black arrows show the heterogeneous nucleation pathways of NAT and ice on pre-existing solid particles supported by the new laboratory and field observations.
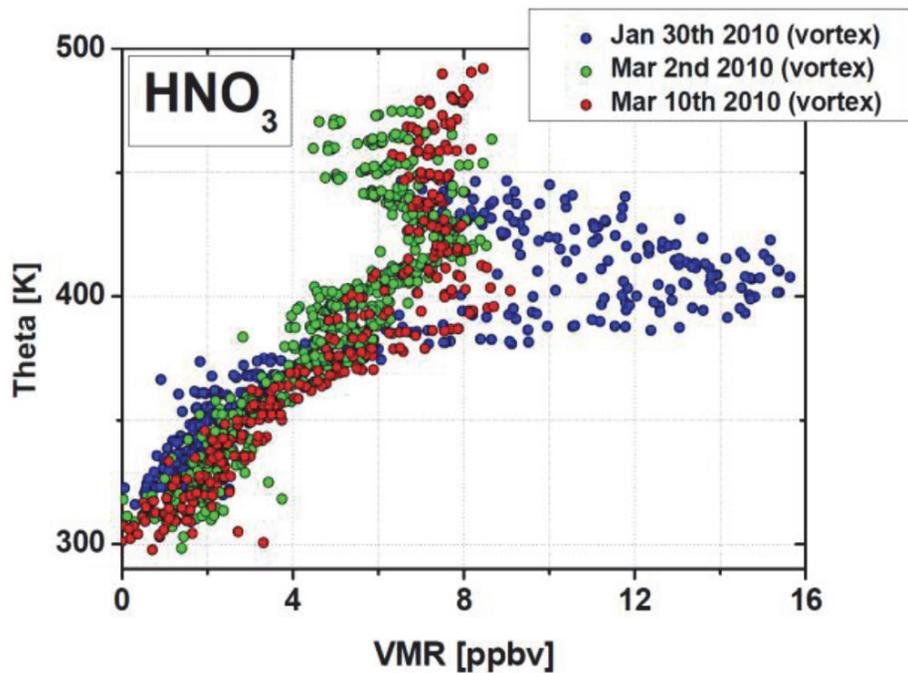
**Fig. 13.** MIPAS-STR standard retrieval results showing vertical redistribution of $HNO_3$ attributed to de/renitrification in Arctic polar winter/spring 2010.
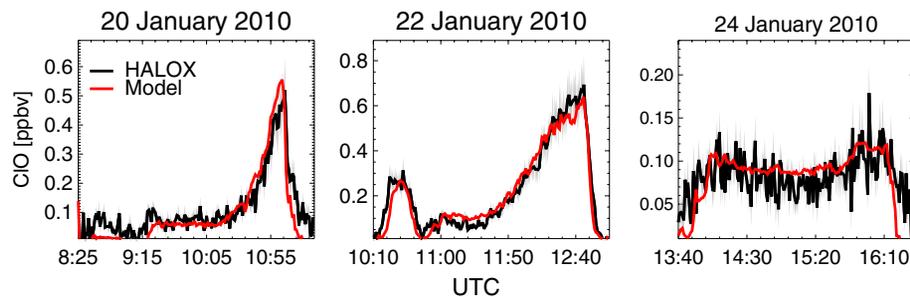
**Fig. 14.** Comparison of ClO simulated by CLaMS and observed by HALOX. Adapted from Wegner et al. (2012a).
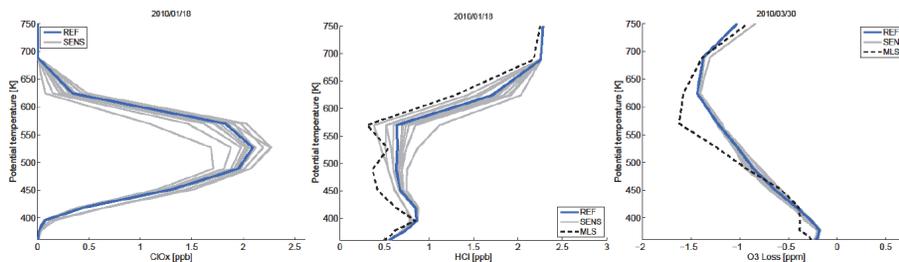
**Fig. 15.** Results of ATLAS model runs, showing the reference (blue) and sensitivity (grey) runs for vortex-averaged ClO$_x$ (left) and HCl (middle) for 18 January 2010, as well as ozone loss for 30 March 2010 as a function of potential temperature. For comparison, HCl observed by MLS and ozone loss inferred from MLS measurements (subtracting observed ozone measurements from the passive ozone tracer) are shown as black dashed lines in the respective panels. See Wohltmann et al. (2012) for details.
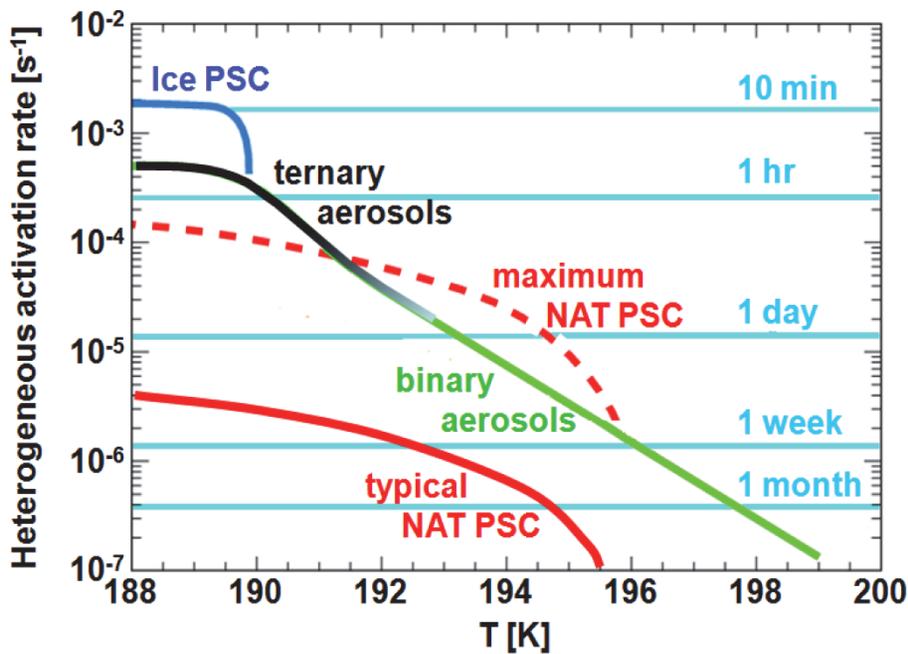
**Fig. 16.** Rates for the reaction $Cl^- + ClONO_2 \rightarrow Cl_2 + NO_3^-$ in/on the condensed phase (liquid or solid) calculated for pressure level 50 hPa and mixing ratio 1 ppb HCl, 10 ppb $HNO_3$ and 5 ppm $H_2O$. Rates for aerosols, ice clouds and "maximum NAT PSC" refer to NAT clouds characteristic for the Arctic, where the low NAT particle number densities with 2 orders of magnitude lower surface area densities hinder the gas phase to equilibrate. Transition from binary $H_2SO_4/H_2O$ aerosols to ternary $HNO_3/H_2SO_4/H_2O$ aerosols is indicated by the smooth transition from light green to black. Short time scales on the order of 1 day for chlorine activation appear to be possible by processing on binary aerosols, not requiring the formation of any PSCs (based on Biele et al., 2001; Carslaw et al., 1997; Dameris et al., 2007; Tsias et al., 1999).

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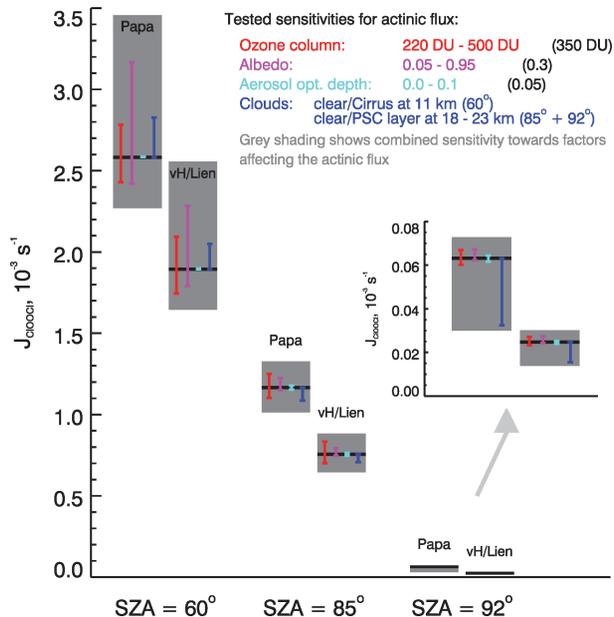
**Fig. 17.** ClOOCl photolysis rate, $J_{ClOOCl}$, at different solar zenith angels for two different ClOOCl cross sections: Papa: determined by Papanastasiou et al. (2009) and currently recommended by JPL; vH/Lien: spectrum measured by von Hobe et al. (2009) scaled to the absolute measurement by Lien et al. (2009) at 248 nm wavelength. The grey area represents uncertainty due to variability in the actinic flux, with the black lines denoting a baseline scenario and the coloured error bars showing the sensitivities towards individual parameters affecting the actinic flux. This plot summarizes von Hobe et al. (2012b). Actinic fluxes were calculated using the radiative transfer model MYSTIC (Monte Carlo code for the phYsically correct Tracing of photons in Cloudy atmospheres, Mayer, 2009) in fully spherical geometry (Emde and Mayer, 2007) with the ALIS (Absorption Lines Importance Sampling) method to perform the high spectral resolution calculations efficiently (Emde et al., 2011). MYSTIC is part of the libRadtran radiative transfer package (Mayer and Kylling, 2005; see also http://www.libradtran.org).

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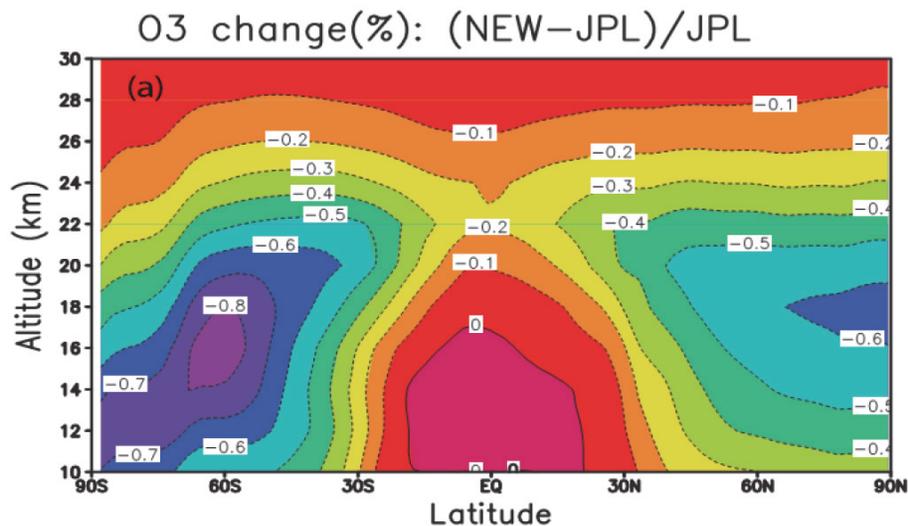
**Fig. 18.** Difference in zonal mean annual mean ozone from a simulation of the SLIMCAT 3-D CTM with scaled $J_{BrONO_2}$ (times 1.27) and $k_{BrO+NO_2}$ (times 0.75) compared to a run with standard JPL kinetics (from Kreycy et al., 2012).
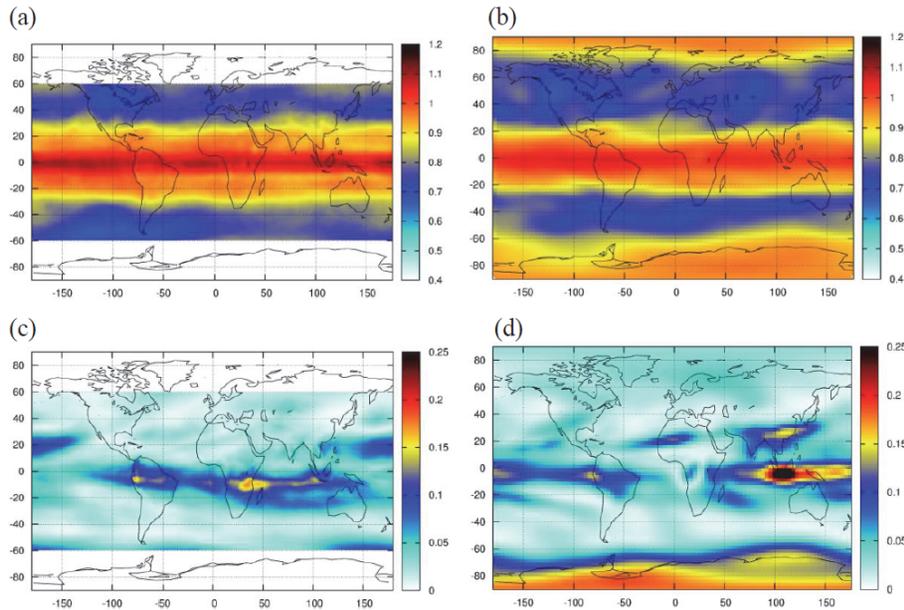
**Fig. 19. (a)** Global geographic distribution of the DFA2 correlation exponent determined for the total column data base produced by Bodecker Scientific, and **(b)** for the LMDZrepro simulations. The reproduction of empirical correlation properties is very accurate in the overlapping region, spectroscopic measurements are not available over the poles. **(c)** Geographic distribution of the spectral weight of semiannual oscillations for the total column data base produced by Bodecker Scientific, and **(d)** for the LMDZrepro simulations. The agreement is satisfactory, however the simulations exhibit anomalously strong oscillations over the Indian ocean and the Tibetan plateau. Semiannual oscillations over the Antarctic is a well known feature in many atmospheric variables.
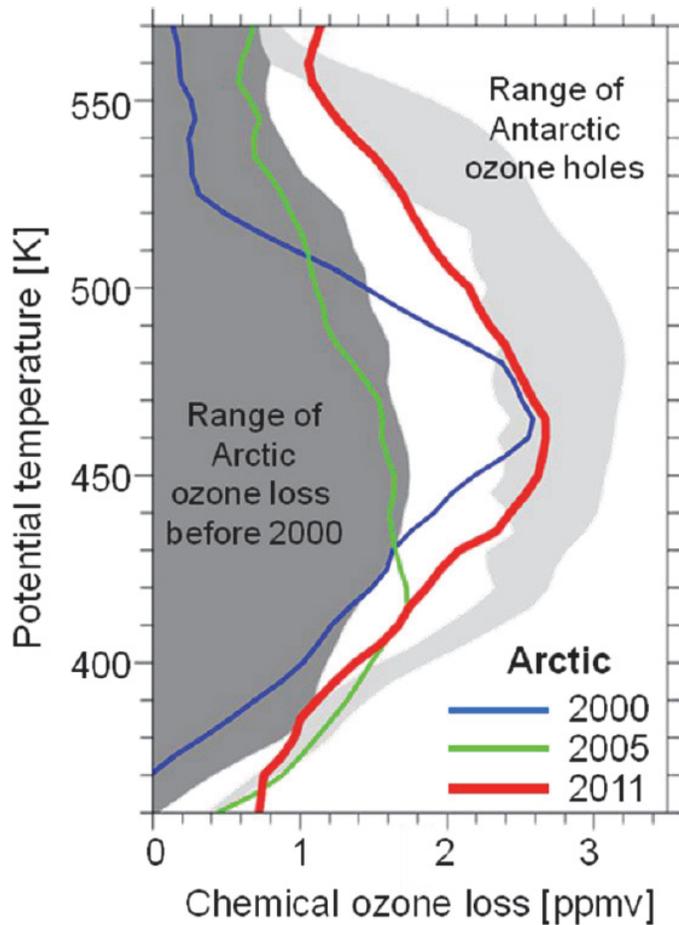
**Fig. 20.** Vertical profile of chemical ozone loss for 2011 and previous Arctic winters derived from match ozone sonde measurements.

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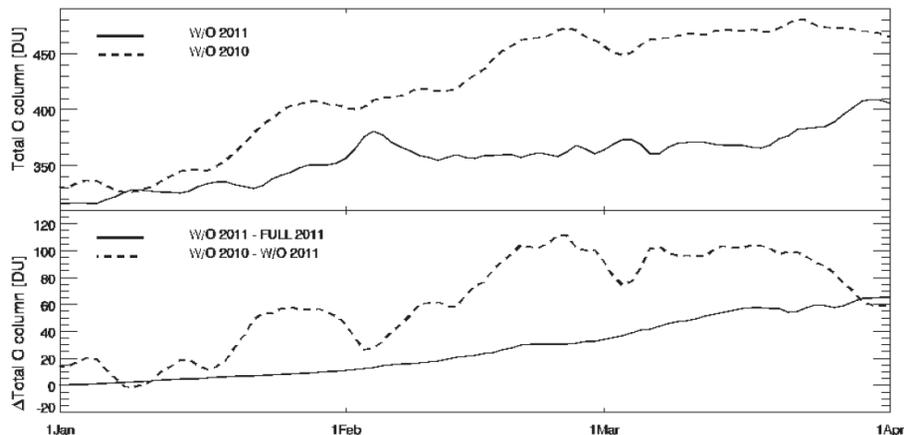
**Fig. 21.** Modelled average column ozone north of 60° N for January, February and March 2010 (upper panel, dotted line) and 2011 (upper panel, full line) in simulations with no ozone-depleting chemistry north of 60° N. Relative difference in transport in 2010 compared to 2011 (lower panel, dotted line), and absolute ozone loss from heterogeneous chemistry in 2011 (lower panel, full line). Figure adapted from Isaksen et al. (2012).