1 Executive Summary

More than 25 years after the first discovery of the Antarctic Ozone Hole and the ratification of the Montreal Protocol (signed in 1987 and reinforced several times), significant ozone reductions still occur over both poles in spring. Some details of the dynamic and chemical processes involved were still not completely understood, and the response of the ozone layer to future alterations in temperature, circulation patterns and chemical composition in the stratosphere related to climate change – or possible geoengineering ventures to mitigate climate change – could not be realistically predicted.

Through dedicated laboratory and field measurements, RECONCILE has augmented our understanding of key processes dominating Arctic ozone loss. This improved process understanding has been implemented in global chemical transport models (CTM) and prognostic chemistry climate models (CCM). The main outcomes of RECONCILE are:

- An unprecedented dataset to study processes in the Arctic winter stratosphere has been obtained from airborne measurements in 2010 and two Match campaigns in 2010 and 2011. The data are publicly available under https://www.fp7-reconcile.eu/reconciledata.
- A clear positive signal has been revealed towards recovery of the Antarctic Ozone Hole.
- Ozone levels over the Arctic dropped to values lower than ever before in spring 2011. This was caused by a combination of inhibited dynamic supply of ozone from lower latitudes and substantial chemical ozone loss, both fostered by special meteorological conditions with a very cold and stable polar vortex. A direct link to climate change for this particular winter cannot be established, but climate change may potentially lead to similar conditions in future years. Surface ultraviolet (UV) radiation in the northern hemisphere was elevated in 2011, locally up to a factor of two for short time periods in March and April. However, these increases are smaller than in the Antarctic and do not exceed UV increases related to variability in the meridional transport of ozone from the tropics.
- Laboratory experiments and new field data on the ClOOCl photolysis rate have reduced the uncertainty of this important kinetic parameter to about 20 %. No evidence has been found supporting the existence of yet unknown chemical mechanisms significantly contributing to polar ozone loss.
- Strong evidence has been found for significant chlorine activation not only on polar stratospheric clouds (PSCs) but also on cold binary aerosol.
- PSCs can form more rapidly and at higher temperatures than previously thought by heterogeneous nucleation[1]. This has been unambiguously demonstrated and parameterised, enabling a more realistic representation of denitrification[2] in models. A variety of possible heterogeneous nuclei has been characterised by chemical analysis of the non-volatile fraction of the background aerosol. While meteoritic particles present in the samples were expected, other components including crustal material and particles of anthropogenic origin were not necessarily expected and raise questions on sources and transport pathways of stratospheric aerosols.
- Deficiencies to accurately simulate Arctic vortex composition in spring have been attributed to an unrealistic model initialisation for a vortex perturbed by strong mixing events in the early winter. Good agreement between observed and simulated tracers suggests an appropriate parameterisation of transport and mixing processes in chemistry transport models. However, inaccuracies in trajectory calculations based on meteorological wind fields can lead to amplified uncertainties in ozone loss estimates in inhomogeneous regions within the polar vortex.
- Based on the results on PSC formation and catalytic ozone chemistry, kinetic and photochemical rate constants have been updated and a novel non-equilibrium PSC scheme has been incorporated in the chemistry climate model (CCM) LMDz Reprobus. This more realistic description of processes that are key for polar ozone loss clearly enhances the robustness of the past and future ozone trends calculated by the LMDz CCM. Compared to reference simulations, the new scheme does not change simulated ozone levels dramatically. However, for past ozone trends, the RECONCILE upgrade results in a better fit to observations for Antarctica while for the Arctic, no significant difference between the simulations is discerned from the large dynamic variability.

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[1] The condensation of nitric acid trihydrate (NAT) and ice on existing tiny solid particles.

[2] The permanent removal of nitrogen compounds via sedimentation on large solid PSC particles. The nitrogen compounds would otherwise lead to chlorine deactivation and bring the ozone depletion process to a halt.

2 Project context and the main objectives

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 that 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., 2012, see also Section 3.7 of this report) 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.

RECONCILE aimed at understanding how the physical and chemical processes affecting stratospheric ozone will be influenced by and feedback on climate change with a focus primarily on the Arctic stratosphere. Below, a brief overview of past research and policies on polar stratospheric ozone depletion is given, and the state of knowledge at the beginning of the RECONCILE project is described, followed by the specific project objectives addressing the remaining open questions and knowledge gaps.

The Discovery of the ozone hole and the Montreal Protocol

An illustrated timeline of "historical events" and important research projects with respect to polar stratospheric ozone is given in Figure 1.

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 (Figure 2).

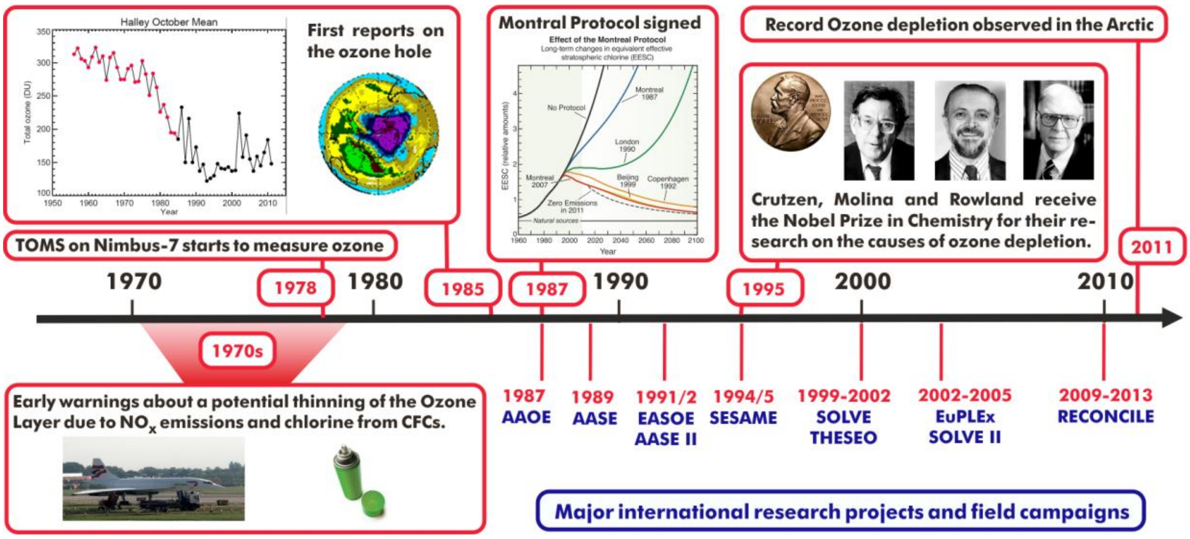

Figure 1 Illustrative timeline of ozone research. More detailed discussions on the history of the discovery of the anthropogenic influence on stratospheric ozone are given by Solomon (1999), Brasseur (2008) and Müller (2009). Parts of this figure are from WMO (2010) and http://www.nobelprize.org, the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

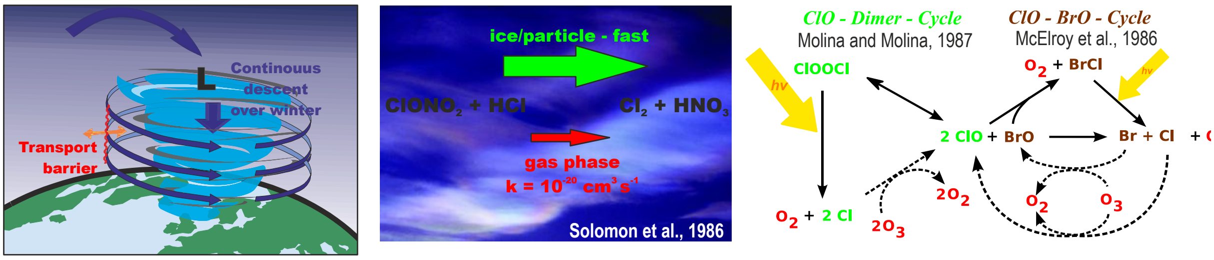

Figure 2 Conditions favouring the formation of an ozone hole in polar regions in spring. (a) The isolation of the air inside the polar vortex inhibits dilution of active chlorine and replenishment of ozone (McIntyre, 1989; Proffitt et al., 1989). (b) Fast heterogeneous reactions on particle surfaces 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).

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 (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 Figure 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).

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., 2006; Rex et al., 2004).

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.

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 Figure 1) carried out during the past 25 years have continuously advanced our understanding of the processes involved in ozone depletion. Many 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 maneuverability 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 Section 3 of this report stand in and continue this fruitful tradition.

RECONCILE objectives

At the beginning of the RECONCILE project in 2009, the lack of understanding in stratospheric dynamics and chemistry was most palpable for issues including the catalytic ClOx/BrOx 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 circumpolar 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, eventually, 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, particularly 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., 2003; 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. Temperature histories along air parcel trajectories, which were calculated using gridded data of numerical weather prediction (NWP) models, were not fully reliable because, for example, effects of stratospheric mountain waves were not taken into account (Dörnbrack et al., 1998; Dörnbrack et al., 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 sulfate 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 σ_{ClOOCl}. In 2007, values of σ_{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.** With the updated CCM, simulations of past and future ozone trends were to be carried out to assess the impact of new process parameterisations compared to "reference simulations". A critical test regime should evaluate the success of the model in reproducing observed ozone trends.

3 Main S & T results/foregrounds

The organisation of this Section stems from the work package structure of RECONCILE.

First, the outcome and data products resulting from the aircraft (WP 5) and Match (WP 6) field campaigns are described. Then, new scientific knowledge and process understanding gained in the areas of catalytic ozone destruction (WP 1), heterogeneous chlorine activation and PSC microphysics (WP 2) and Arctic vortex dynamics and mixing (WP 3) is presented. The development of process parameterizations and their implementation in a global chemistry climate model (WP 4) are described together with the outcome of new CCM simulations on past and future Arctic ozone depletion. Additional outcomes from RECONCILE not directly related to the project objectives are given at the end of the section.

Many of the RECONCILE results described here integrate contributions from external partners (i.e. partners who were not part of the consortium of the official RECONCILE beneficiaries) and projects funded from national and international resources. A complete list and more details of these extremely fruitful collaborations are given in Section 4 of this report.

3.1 The RECONCILE aircraft campaign in spring 2010 (WP 5)

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.

The meteorological situation in the campaign winter

The Arctic winter 2009/10 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. 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) telecommunication pattern in the troposphere, as described by Nishii et al. (2011) and Orsolini et al. (2009). A major sudden stratospheric warming (SSW) at the end of January 2010 concluded the anomalously cold vortex period and prevented an extensive ozone loss in the RECONCILE winter. **A detailed overview of the meteorological situation and evolution of the polar vortex has been given in Dörnbrack et al. (2012)**.

Overview of the RECONCILE flights

During two measurement periods (17 January – 02 February and 27 February – 10 March 2010) to cover early and late winter conditions, 12 RECONCILE flights of the high-altitude aircraft M55-Geophysica to-

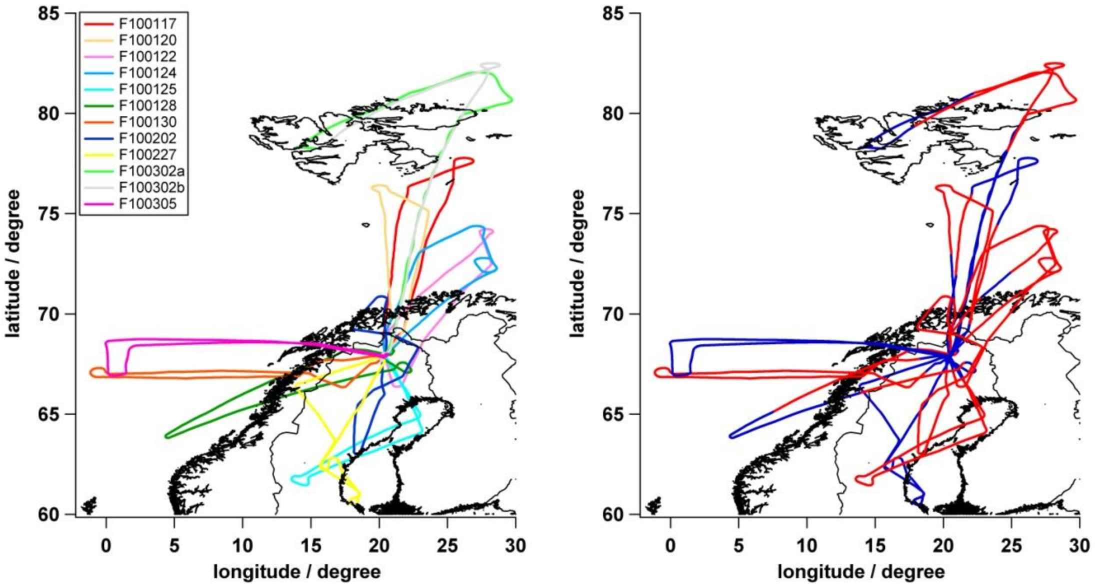

Figure 3 Geophysica flight paths during RECONCILE. In the left panel, all flights are marked by different colours (referring to flight date, yymmdd). In the right panel, flight sections in vortex air are marked red, flight sections outside the polar vortex are marked in blue. Two self-match flights were carried out on 25 and 30 January, a match flight on 2 February (matching the flight on 30 January).

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 Figure 3.

No	date	take off	landing	Scientific targets
F1	17.01.	11:20	15:33	PSC characterization
F2	20.01.	08:13	11:55	PSC characterization and CALIPSO match
F3	22.01.	10:01	13:35	PSC characterization
F4	24.01.	13:30	16:56	PSCs and renitrification
F5	25.01.	05:50	09:19	PSCs, de/renitrification, Cl active., self-match experiment[1]
F6	28.01.	09:00	12:56	vortex survey (dynamics and mixing)
F7	30.01.	06:36	10:15	self-match experiment[1], Cl activation, de/renitrification
F8	02.02.	10:01	13:31	match experiment[2], Cl activation, de/renitrification, O_3 loss
F9	27.02.	12:06	15:41	vortex filament
F10[3]	02.03.	02:51	06:31	vortex survey, Cl deactivation, de/renitrification, O_3 loss
F11[3]	02.03.	09:35	13:35	vortex survey, Cl deactivation, de/renitrification, O_3 loss
F12	05.03.	15:05	18:36	vortex filament
F13[4]	10.03.	07:14	10:51	test of remote sensing instrumentation, O_3 loss

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[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., in preparation*).

[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

talling 57 flight hours were carried out from Kiruna, Sweden. An additional flight was carried out as part 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 Figure 3. An overview of the comprehensive and innovative payload is given in Table 2. **The data are available online at https://www.fp7-reconcile.eu/reconciledata.html.** Particularly noteworthy in this dataset are the PSC observations during the first five flights. The in-situ cloud particle instruments FSSP, CIP and CCP, the airborne lidar MAL and backscatter sonde MAS, and the infrared limb sounders CRISTA-NF and MIPAS, provided, together with the COBALD backscatter sonde data (cf. Section 3.2) as well as ground-based and space-borne lidar observations, **the most comprehensive set of PSC observations to date** was obtained, including the first visual evidence for so-called NAT-rocks (cf. Section 3.4). Additionally, SIOUX measured gas phase and particle NO_y to assess the nitrogen redistribution linked to PSCs, and COPAS measured the smaller particles that serve as condensation nuclei involved in the formation of PSCs. The implementation of an impactor sampler in the inlet of COPAS made possible, **for the first time, a chemical composition analysis of the refractory aerosol inside the polar votex (cf. Section 3.4)**.

Flight planning was based on results of the Integrated Forecast System (IFS) of the European Centre of Medium-Range Weather Forecasts (ECMWF), with **innovative flight planning tools being used from the first time during the RECONCILE aircraft campaign**:

- Different thermo-dynamical 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. Section 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.

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-α	(Zöger et al., 1999)
FLASH	H_2O (gas phase)	8 s	0.2 ppm	6 %	Lyman-α	(Sitnikov et al., 2007)
SIOUX	NO NO_y Particle NO_y	1 s 1 s	10 % 15 %	3 % 5 %	Chemiluminescence, + Au-converter + Subisokinetic inlet	(Voigt et al., 2005)
HALOX	ClO ClOOCl	10 s 30 s	20 % 30 %	5 % 15 %	CCRF + thermal dissociation	(von Hobe et al., 2005)
HAGAR	N_2O, CFC12 CFC 11 CH_4, H_2 SF_6 Halon 1211 CO_2	90 s 90 s 90 s 90 s 90 s 5 s	1.3 % 1.6 % 1.8 % 2.3 % 3.0 % 0.1 %	0.3 % 0.6 % 0.8 % 1.3 % 2.0 % 0.05 %	Gas Chromatography (GC) with electron capture detector (ECD) IR absorption	(Homan et al., 2010; Werner et al., 2010)
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. (0.4-47μm)	1 s	20 %	10 %	Laser-particle spectrometer	(de Reus et al., 2009)
CCP	Cloud particle size distrib. (3-47μm)	1 s	20 %	10 %	Laser-particle spectrometer	
CIP	Cloud particle size distrib. (25-1600μm) 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 isotopologues	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	0,1 K 0.1 m/s	PT100, 5-hole probe	
remote sensing						
MAL 1 &2	Remote Aerosol Profile (2km 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 programme)	T < 1K / vmr typ. 10-15 %	T < 1K / 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 viewing geometry	(Ungermann et al., 2012; Weigel et al., 2010)
MARSCHALS	O_3, H_2O, CO, HNO_3, N_2O				Millimetre Wave spectrometer in limb viewing geometry	(Moyna et al., 2006)

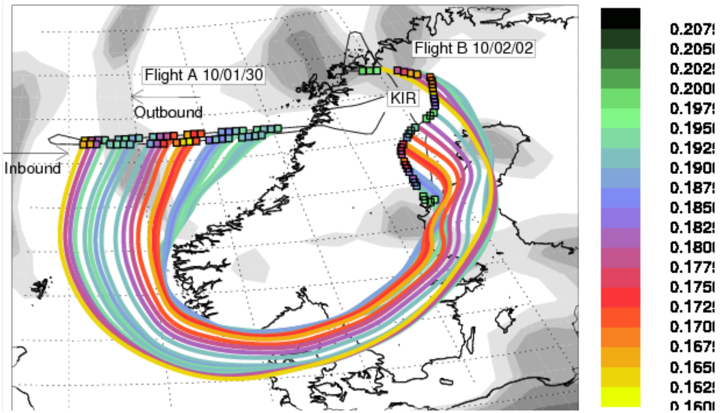

Figure 4 Flight tracks of the second self match flight on 30 January and the match flight on 2 February with HAGAR N_2O measurements along the flight tracks shown by the coloured squares. The trajectories connecting the flights are colour coded according to the N_2O (in ppb) observed during the first flight. The grey contours in the background show PV from ERA-interim at the 425 K level on 30 January 12 utc, with white representing high PV and darker grey representing areas of low PV pointing to a significant contribution of non-vortex-air.

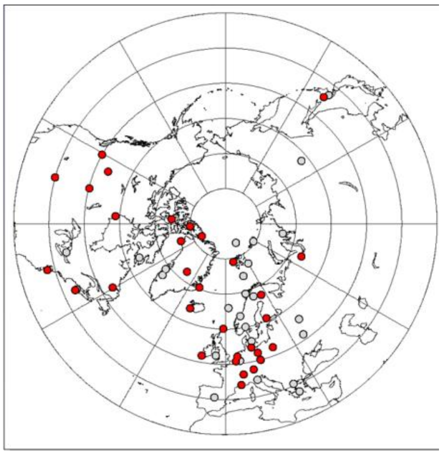

Figure 5 Network of ozone sounding stations that participated in the RECONCILE Match campaign (red). Gray dots indicate stations that were active in Match campaigns in the past but which went out of operation during recent years due to lack of support.

Three innovative aircraft Lagrangian Match experiments[3] **were carried out during the RECONCILE**. 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 (Figure 4). The planning of these Lagrangian flights was carried out using trajectories based on ECMWF data.

3.2 RECONCILE Match campaigns in 2010 and 2011 (WP 6)

The Match campaign in the winter 2009/10 consisted of two elements, ozone sonde matches and COBALD backscatter sonde activities, including water vapour soundings. **An additional ozone sonde Match campaign was carried out in the subsequent Arctic winter 2010/11 that saw record breaking ozone loss (Manney et al., 2011, cf. Section 5)**.

The Match technique to estimate winter ozone loss has been first described by Rex et al. (1999). 31 ozone sounding stations (Figure 5) 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 February 28, 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. The campaign in the winter 2010/11 started on 12 January 2011 and was stopped on 27 April 2011. About 270 ozone sondes were launched from 13 stations actively participating in the campaign.

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., 2013a; Khaykin et al., 2013). 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

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[3] 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. Section 3.2

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.

Complementary to the RECONCILE activities, 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). 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 RECONCILE aircraft flights and the sounding programme at Sodankylä was carried out in two phases, with 18 payloads flown between 17 January and 06 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 ± 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., 2013, manuscript in preparation). 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 night-time 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.

3.3 Results related to parameters driving catalytic ozone loss cycles (WP 1)

Laboratory experiment on ClOOCl

A dual wavelength range spectrometer system (Figure 6) 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. Besides an optically pure gas phase UV spectrum of ClOOCl, the experiment resulted in **the first measurement of the visible spectrum of ClOOCl** (Young et al., in prep.). Both the measured UV and visible spectra of ClOOCl are shown in Figure 7.

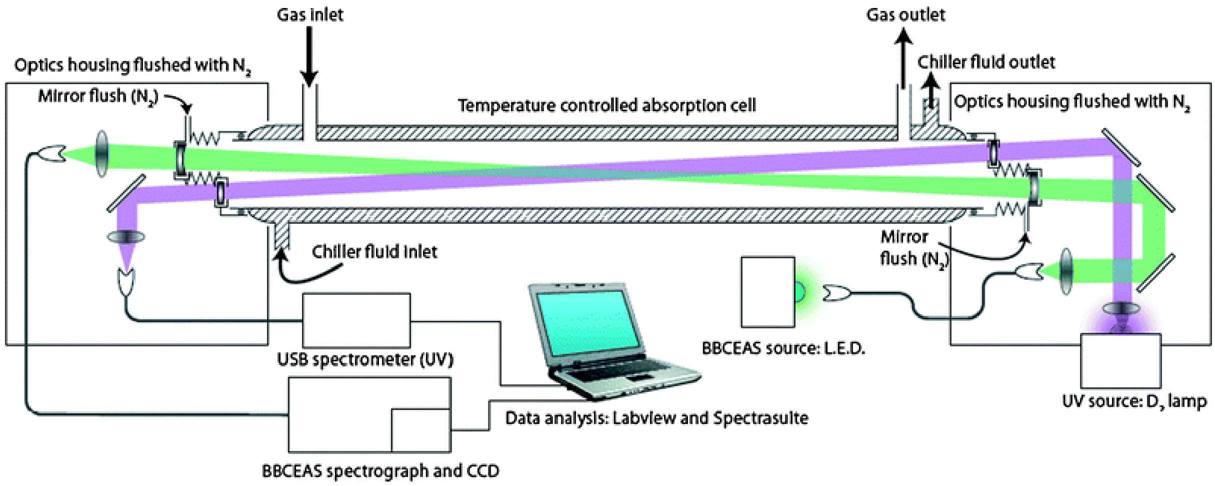

Figure 6 Schematic oft he BBCEAS system used in the ClOOCl experiment. From Young et al. (2011).

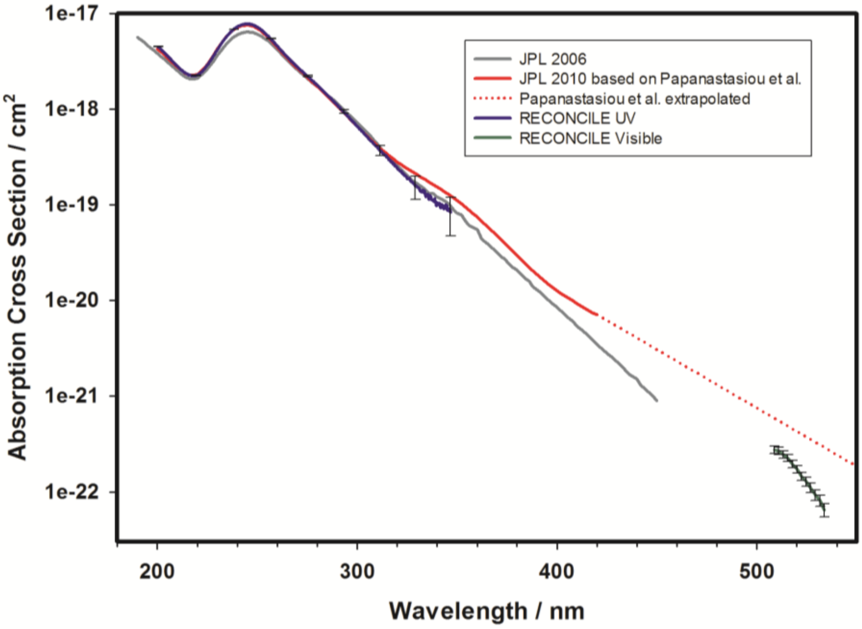

Figure 7 UV and visible absorption cross sections of ClOOCl, provided error bars are 1σ.

Within the reported errors the UV spectrum obtained from the new laboratory experiment 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 first measurement of the much weaker visible spectrum of ClOOCl provides important information, with which to constrain the long wavelength absorption of the ClOOCl and hence ClO_x catalysed polar O_3 loss cycles. It can be seen that the measured visible spectrum of ClOOCl is in close agreement with what was expected from extrapolating the JPL recommendations.

Field observations: the RECONCILE self Match flight

Results from the laboratory experiment are consistent with atmospheric observations made during the self-match flight (cf. Section 3.1) published by Sumińska-Ebersoldt et al. (2012) and shown in Figure 8. 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 (Figure 9).** 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).

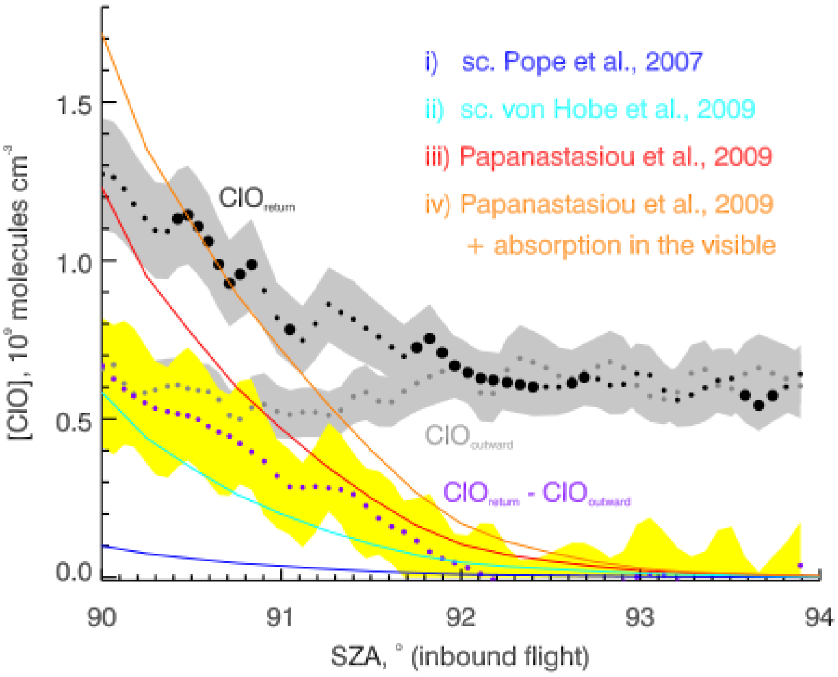

Figure 8 Comparison of the observed rise of ClO concentrations (marked by the purple symbols) with SZA to the integrated ClO produced by simulations using various choices for J_{ClOOCl}. The measured rise is calculated by subtraction of ClO measured on the outbound flight leg (grey symbols) from ClO measured on the inbound flight leg (shown as black symbols with the thicker ones representing strict match pairs). The outbound ClO data are plotted according to the SZA at the matching inbound measurements. Grey and yellow areas show the measurement uncertainties.

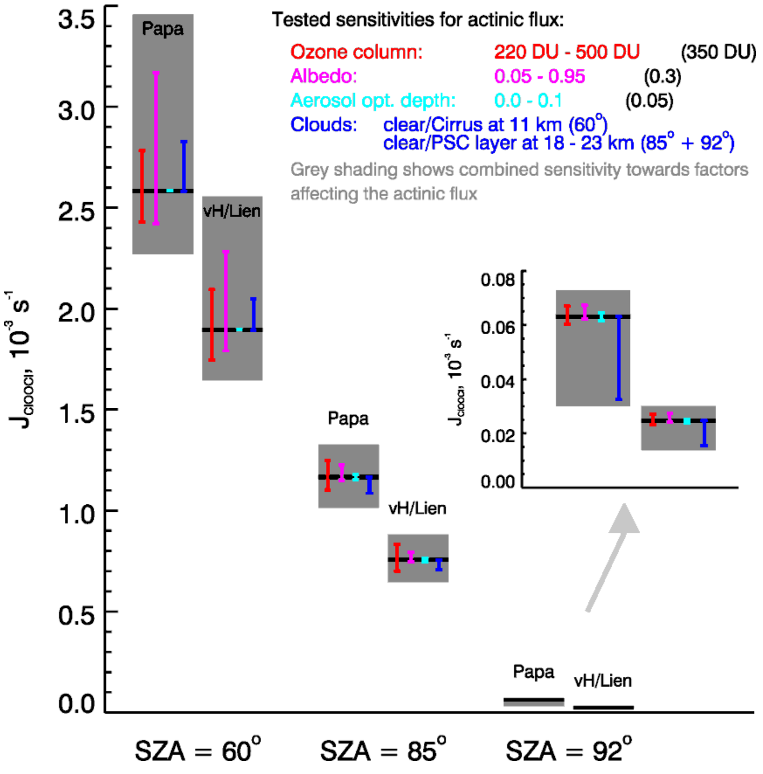

Figure 9 ClOOCl photolysis rate, J_{ClOOCl}, at different solar zenith angles for two different ClOOCl cross sections: Papa: determined by Papanastasiou et al. (2009) and currently recommended by JPL; vH/Lien: spectrum measured by von Hobe et al. (2009) scaled to the absolute measurement by Lien et al. (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, manuscript in preparation). 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).

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 $\boldsymbol{k_f}^1$	(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 $\boldsymbol{K_{eq}}$	(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 $\boldsymbol{K_{eq}}$ (Suminska-Ebersoldt et al., 2012). Again, this is in good agreement with theoretical considerations by von Hobe et al. (2007)
ClOOCl absorption cross sections $\boldsymbol{\sigma_{ClOOCl}}$	(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.

[1] We only consider the low pressure limit for $\boldsymbol{k_f}$, which is more relevant for stratospheric conditions than the high pressure limit

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).

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 Sept. 7, and 8th, 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_{BrONO2} to the three body formation reaction ($BrO + NO_2 + M$) rate coefficient of $BrONO_2$ $k_{BrO+NO2}$, i.e. $J_{BrONO2}/k_{BrO+NO2}$, is by a factor 1.7 (-0.32 to +0.51) 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 $J_{BrONO2}/k_{BrO+NO2}$ ratio as compared to the JPL-2011 recommendations), and (2) a larger $J_{BrONO2}/k_{BrO+NO2}$ ratio may cause a small increase in the bromine-mediated ozone loss in the stratosphere (maximum -0.8%, see Figure 10).

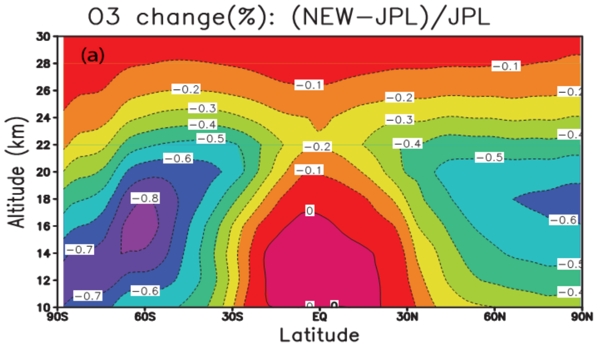

Figure 10 Difference in zonal mean annual mean ozone from a simulation of the SLIMCAT 3-D CTM with scaled J_{BrONO2} (times 1.27) and $k_{BrO+NO2}$ (times 0.75) compared to a run with standard JPL kinetics (from Kreycy et al., 2012).

The trend of total stratospheric bromine (Figure 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 year 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 ± 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 ± 0.25 ppt, where 90% is due to contributions from CH_2Br_2 and $CHBr_3$ (e.g. Brinckmann et al., 2012).

3.4 Processes related to polar stratospheric clouds (PSCs) and aerosols (WP 2)

Heterogeneous activation on cold binaries

As expected, the cold vortex temperatures and extensive PSC coverage in December 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 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 simulation). 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 (Figure 11). 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 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 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.**

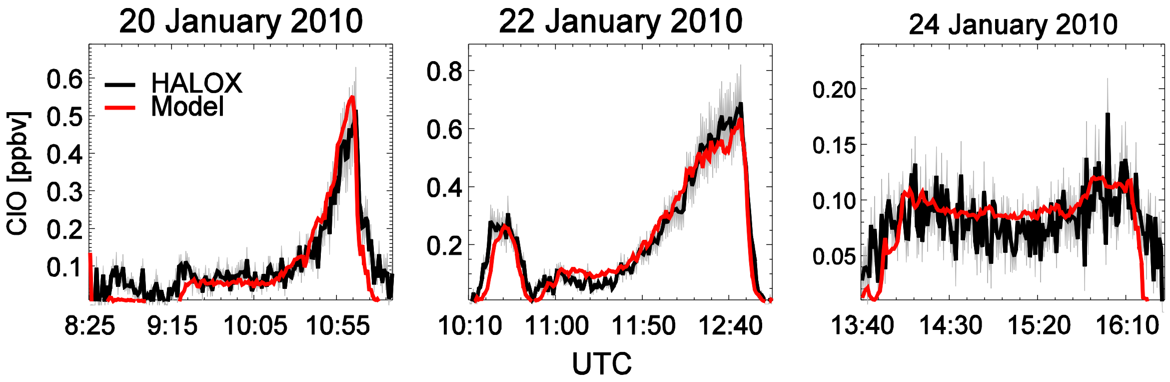

Figure 11 ClO simulated by CLaMS and observed by HALOX. Adapted from Wegner et al. (2012a)

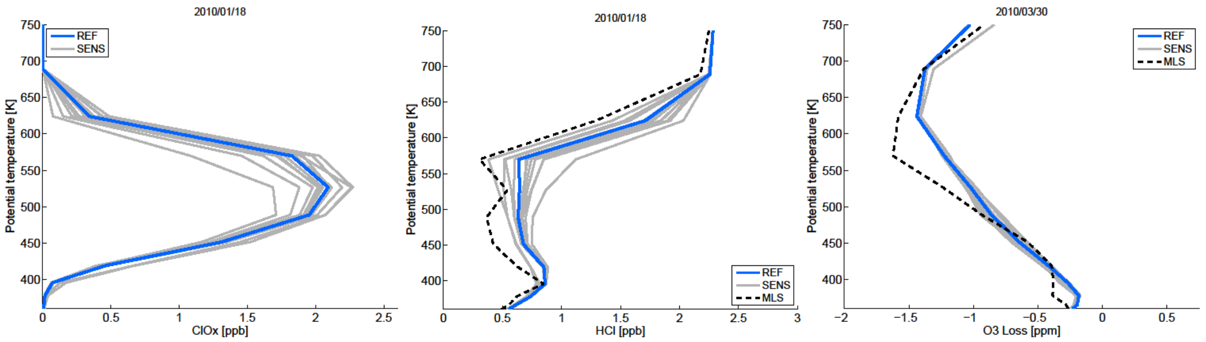

Figure 12 Results of ATLAS model runs, showing the reference (blue) and sensitivity (grey) runs for vortex-averaged ClO_x (left) and HCl (middle) for 18 January 2010, as well as ozone loss for 30 March 2010 as a function of potential temperature. For comparison, HCl observed by MLS and ozone loss inferred 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.

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. (2012b), 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 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 (Figure 12, 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 (Figure 12). Assuming default values for NAT number density and supersaturation, runs with different rates for the heterogeneous reactions $ClONO_2$+HCl and $ClONO_2$+H2O 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).

PSC formation and microphysics

PSC observations in the Arctic winter 2009/10

During the RECONCILE campaign winter, the most comprehensive dataset on PSC properties to date has been obtained 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 Figure 13. The early period from 15–30 December 2009 was characterized primarily by patchy, tenuous low number density liquid/NAT mixtures (Figure 13a). No ice clouds were observed by CALIPSO during this early phase, suggesting 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 (Figure 13b). 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 (Figure 13c). The final period of the PSC season (22–28 January) was characterized by a major stratospheric 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 (Figure 13d). 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 profiles at PSC levels (Figure 14) 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 during 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 penetration of NAT fields during

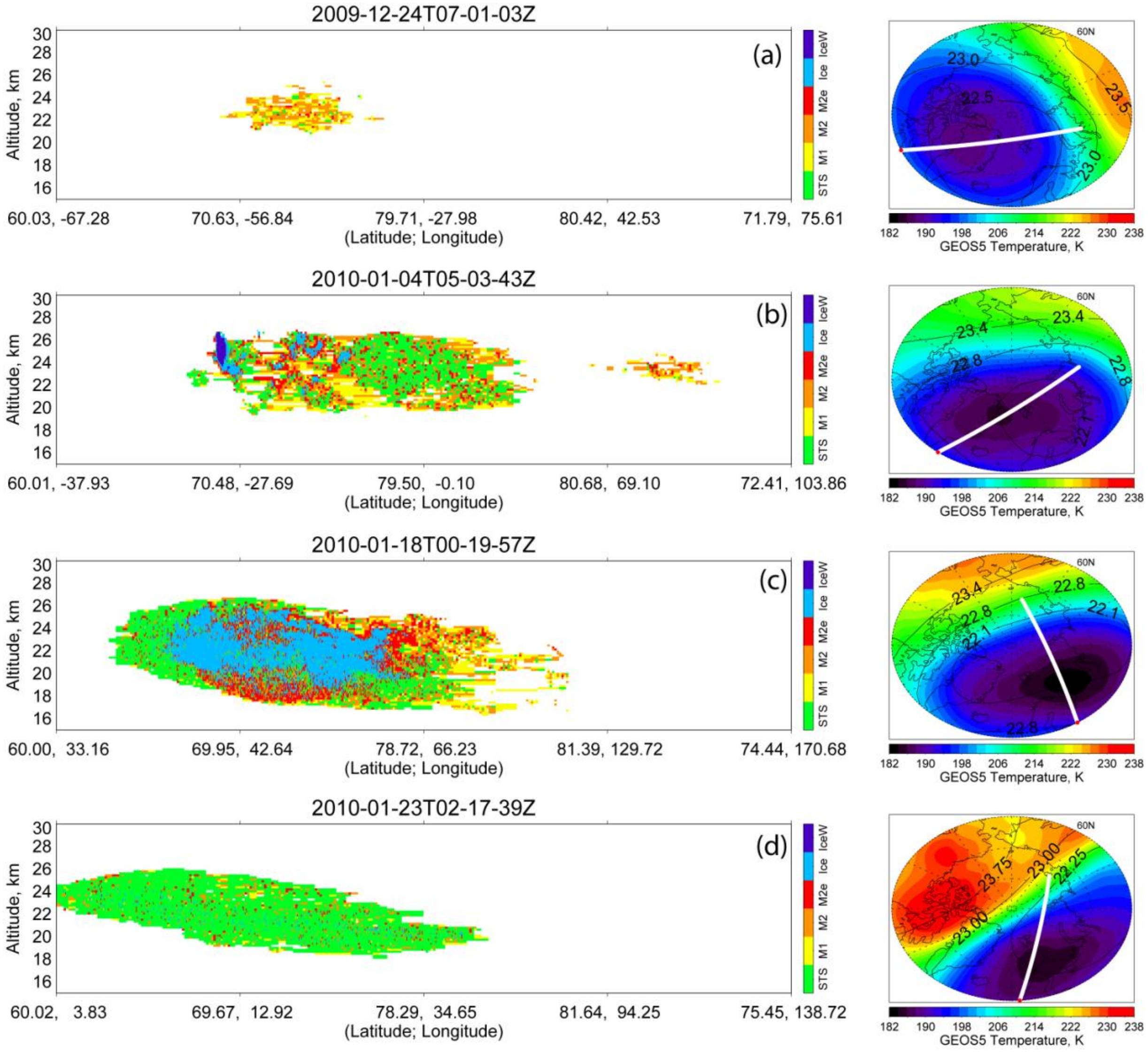

Figure 13 Examples of CALIPSO PSC composition observations during the 2009-2010 Arctic winter. The panels on the left show CALIPSO PSC composition images for representative orbits from four periods during the winter: (a) 15-30 December, (b) 31 December-14 January, (c) 15-21 January, and (d) 22-28 January. 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).

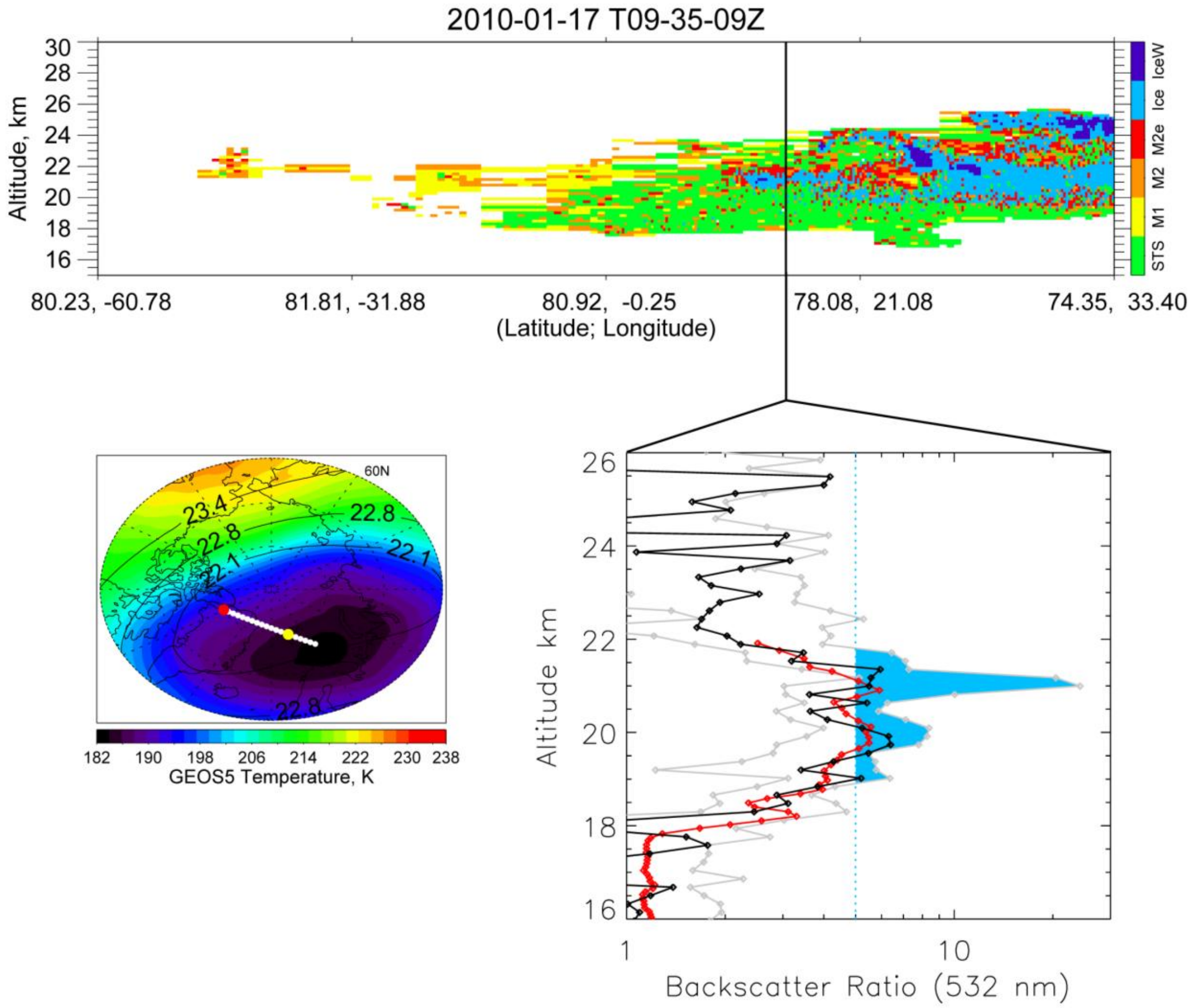

Figure 14 CALIPSO - COBALD comparison on the 17 January 2010. Upper panel: CALIPSO PSC composition observations. The black line denotes the location of the simultaneously performed COBALD sounding. Lower left panel: GEOS5 temperature and geopotential height fields at 30 hPa for 12:00 UTC with the location 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.

a RECONCILE flight was confirmed also by the MAL (Miniature Aerosol Lidar), MAS (Multiwavelength Aerosol Scattersonde), and SIOUX (StratospherIc Observation Unit for nitrogen oXides) instruments (c.f. 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 minutes 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 15 shows a size distribution averaged over a period of 22 minutes 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 0.5-1µm 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).

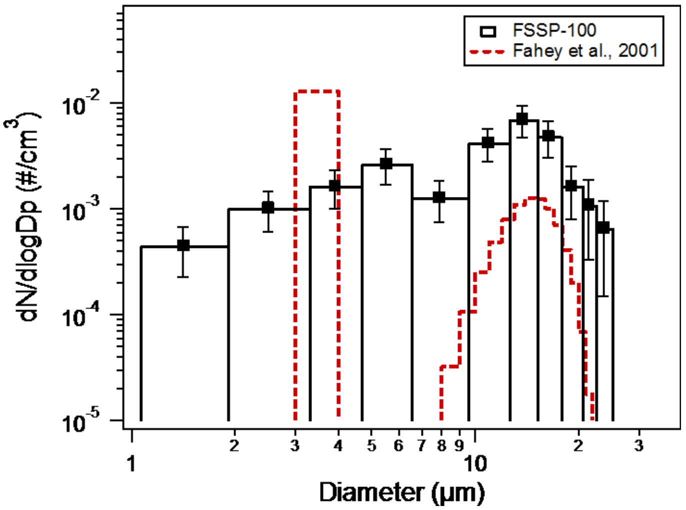

Figure 15 Number size distribution obtained from FSSP-100 measurements during RECONCILE flight on 25 January 2010, averaged over 22 minutes (from 7:58 to 8:20 UTC). For comparison, the size distribution retrieved from NO_y observations in the Arctic winter 2000 by Fahey et al. (2001) is shown in red.

Consistent with the in situ observations, the presence of ice-particles at 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 Figure 15) compare reasonably well with the observed IR limb spectra under the assumption that the large particles consist of NAT (Griessbach et al., Kalicinsky et al., *manuscripts in preparation*). A distinct feature in the spectra suggests that the small particle mode must also contain a significant amount of NAT.

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., 2013). 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. (2013a) using microphysical box modelling.

Characterisation of the 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, manuscript in preparation) present **a detailed chemical analysis of the refractory aerosol material** from 13 (one for each flight) in-vortex particle samples that 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 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 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 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 of PSCs, and furthermore influences the tropospheric cloud formation after the polar vortex has dissolved in the Arctic spring season.

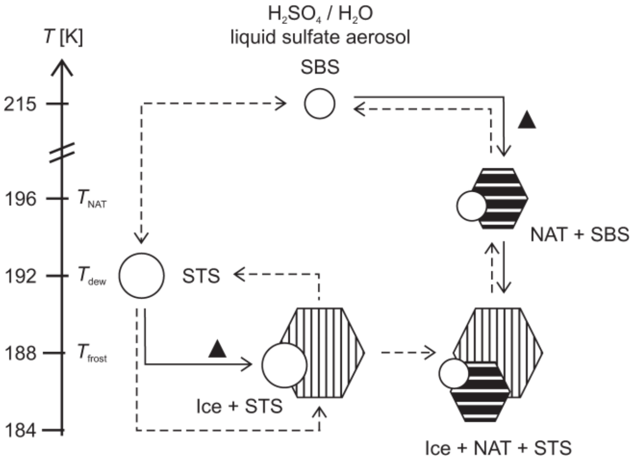

Figure 16 PSC formation pathways as implemented into ZOMM with SBS = Supercooled Binary Solution (H_2SO_4/H_2O), STS = Supercooled Ternary Solution (HNO_3/H_2SO_4/H_2O), NAT = Nitric Acid Trihydrate ($HNO_3 \cdot 3H_2O$), ▲ = solid inclusion, e.g. meteoritic dust. Dashed arrows denote pathways included 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.

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

For every distinct PSC period defined by Pitts et al. (2011) and described above, optical parameters simulated by the Zurich Optical/Microphysical box Model (ZOMM) were compared to selected CALIPSO PSC observations. 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). 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. Figure 16 shows the **new scheme of PSC formation and phase transitions** that is presented and discussed in detail in two recent publications (Hoyle et al., 2013; Engel et al., 2013b).

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). Preliminary results from additional laboratory experiments on meteoritic material 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.

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, a process closely linked to PSC microphysics described above in this Section.

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. Section 3.7). 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 early 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.

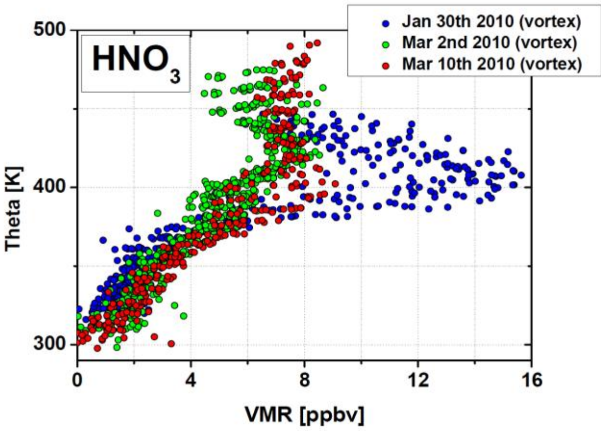

Figure 17 MIPAS-STR results showing vertical redistribution of HNO_3 in Arctic polar winter/spring 2010.

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 $7.8 \cdot 10^{-6}$ 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 a new nucleation rate parameterisation described based on the results obtained with ZOMM (described above), 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 Figure 17 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 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 has been 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 is improved and whether $ClONO_2$ formation is simulated adequately (Woiwode et al., 2012a, manuscript in preparation).

3.5 Vortex dynamics and mixing (WP 3)

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 (Figure 18). 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 (Figure 19, 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 Figure 19). There was a significant drop in T_{MIN} to values as much as 9 K below the climatological mean (Figure 18). 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

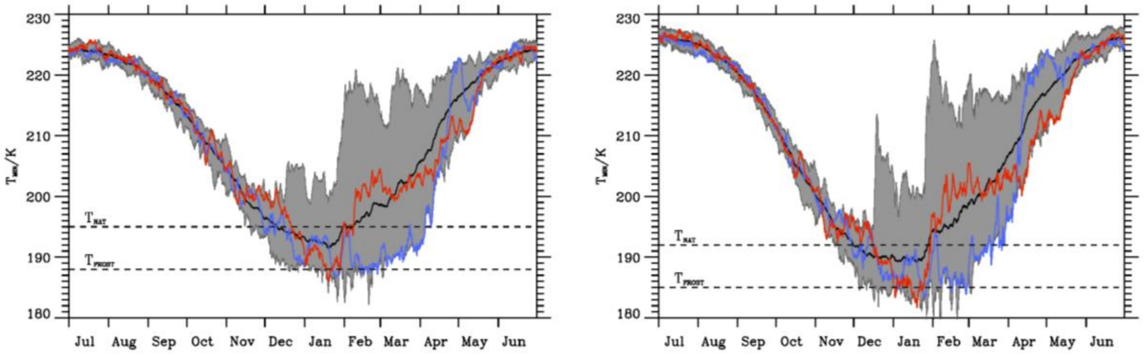

Figure 18 Minimum temperatures T_{MIN} (K) between 65 °N to 90° N on the 50 hPa (left panel) and 30 hPa (right panel) pressure surfaces. Black line: mean value from 1989-2011; red line the T_{MIN} evolution from July 2009 through June 2010, blue line from July 2010 through June 2011; shaded area encompasses 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.

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 Figure 19) 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 (Figure 19**Fehler! Verweisquelle konnte nicht gefunden werden.**, 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 implications for stratospheric ozone trends in the northern hemisphere.

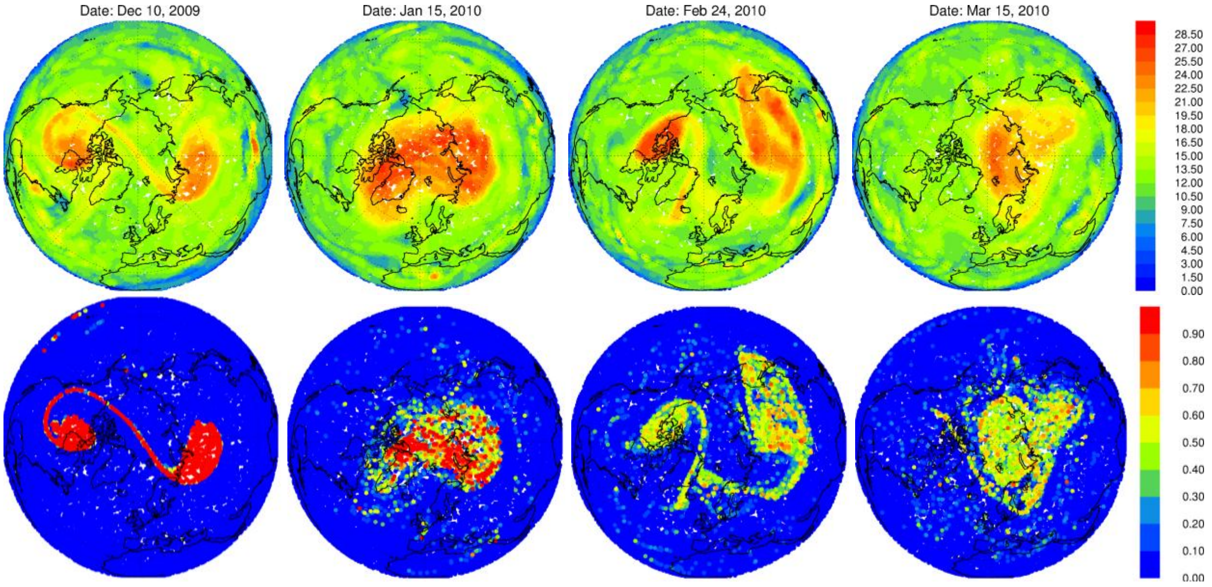

Figure 19 Modified potental vorticity (PV, given in 10^{-6} K m^2 kg^{-1} s^{-1}, upper panel) and inert tracer P4 (indicating the relative contribution of air masses that were inside the vortex core at the time of initialization, lower panel) on 450 K isentropic level during the first vortex split, after re-organization of the vortex, during the second vortex split and at the end of the winter (order from left to right).

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 (Figure 19, left hand column). After a re-organization before January 2010 the vortex stayed coherent with significant mixing only below 500 K (Figure 19, second column). Following the second vortex split enhanced transport of air from lower latitudes into the reformed vortex occurred, (Figure 19, third column), leading to a heterogeneous origin distribution inside the vortex interior during late winter (Figure 19, right hand column). The results from the CLaMS simulations were validated with observations obtained by the CRISTA-NF (CRyogenic Infrared Spectrometers and Telescopes for the Atmosphere) (Kalicinsky et al., 2013) and HAGAR (Hösen et al., 2012, manuscript in preparation) 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, manuscript in preparation), which is also captured by the CLaMS simulations.

Accuracy of trajectories and impact on ozone loss estimates

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, manuscript in preparation). 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 (Figure 4), 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.

Relative role of dynamics and chemistry to the ozone polar deficit in 2010 and 2011

We contrast the ozone evolution and loss during these two winters, and quantify the respective contributions of dynamical mixing and of heterogeneous chemistry to ozone loss during the winters of 2010 and 2011, in comparison to the last 14 years. First, we examine the dynamical and chemical contributions to column ozone variability in the polar cap (latitudes > 60N) in Figure 20. The top panel (a) highlights column ozone in 2010 (black) and 2011 (red) against the background of the last 14 years, and shows the good model agreement with satellite observations (shown in dashed). Noteworthy are the exceptionally low values in 2011 after mid-February, while values in 2010 are the highest among recent years. The panel (b) shows column ozone in model simulations with switched-off ozone chemistry: hence, it reveals that the dynamical replenishment of polar O3 through winter by transport from lower latitudes was anomalously weak (strong) in 2011 (2010) after mid-February. The difference in this polar transport (2011 minus 2010) is plotted on panel (c), along with

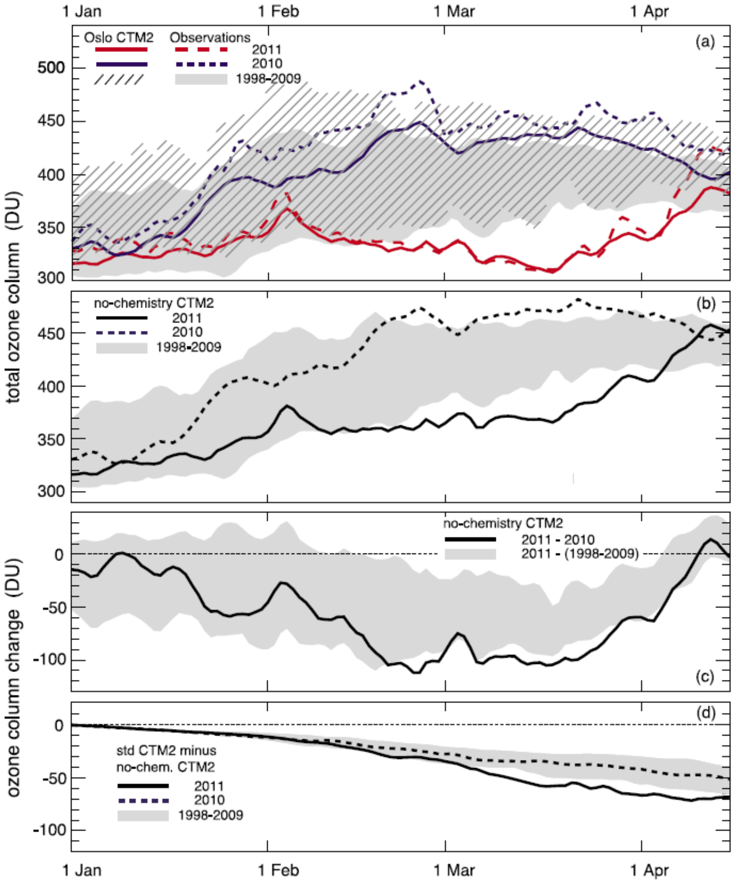

Figure 20 (a) Modeled (full lines) and observed (dashed lines) average column ozone north of 60 N for 2010 (black) and 2011 (red), along with the range of the last 14 years in shading. (b) Same in simulations with no ozone-depleting chemistry north of 60N for 2010 (dashed) and 2011 (full). (c) Relative difference in transport in 2010 compared to 2011 (full), also with range for the last 14 years in shading (d) absolute ozone loss from heterogeneous chemistry in 2010 (dash) vs 2011 (full line). From Isaksen, (2012).

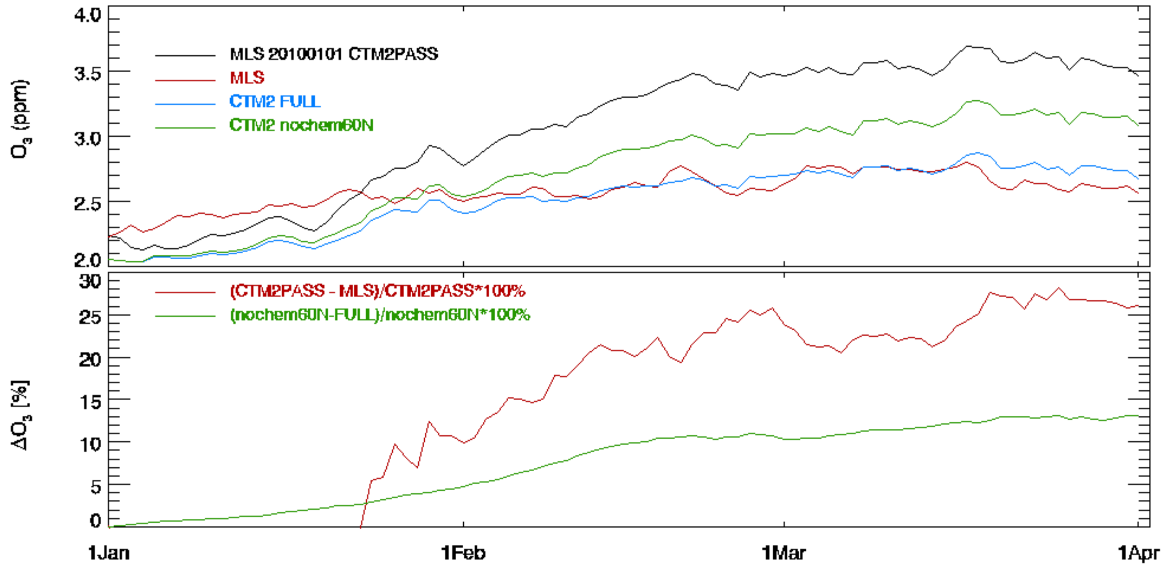

Figure 21 (top) Modeled, vortex-averaged ozone in 2010 at 68 hPa: black line is passive ozone from MLS, blue line is Oslo CTM with full chemistry, green line is Oslo CTM with ozone-depleting chemistry switched-off, and red line is gridded MLS. (bottom) Relative vortex-averaged ozone loss in 2010 at 68 hPa: green line is Oslo CTM, and red line is inferred from MLS observations. Losses (in %) shown as positive.

differences from other years (2011 minus other years, shading). Hence, in 2010, ozone is replenished more efficiently at high latitudes than in 2011.

The rapid increase in the curve for 2010 corresponds to rapid mixing events, like the late January 2010 stratospheric warming. This replenishment 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**. In fact, the lower observed or modelled values in polar regions were mirrored by higher values at low latitudes (not shown), pointing to a more ”sluggish” Brewer-Dobson circulation in 2011compared to 2010. Panel (d) shows model O_3 loss in the polar cap being the strongest in 2011, 70 DU at end of April, compared to the last 14 years, as shown by other recent studies (Balis et al., 2011; Kuttippurath et al., 2012; Manney et al., 2011). While the chemical loss in 2011was much higher than in 2010 or other years, the relative weakness of transport into the very stable and narrow vortex was the dominating factor. **While halogen-induced ozone depletion was record high for the Arctic in 2011, it appears that dynamics was important in setting the scene for the low ozone amount (Balis et al., 2011).**

The above polar-cap averages draw a budget for the Arctic region as a whole, irrespective of the vortex boundary. The modelled vortex-averaged ozone losses are much higher than the polar cap losses of Figure 20d: 92 DU rather than 72 DU at the end of March 2011, and 50 DU rather than 40 DU at the end of March 2010. This is understandable as polar-cap averaging mixes extra-vortex air and vortex air.

At 68 hPa, the model ozone evolution is in good agreement with gridded MLS (Figure 21: model is blue curve, MLS is red curve). To infer ozone loss from observation, we transported the initial MLS ozone on January 1 using the Oslo CTM. The difference between this “passive MLS” (Figure 21: passive MLS is black curve) and the observed gridded MLS (as a proxy for assimilated MLS) is used to infer ozone loss, as in Søvde et al. (2011). At the end of March, this ozone loss is about 1.0 ppm, or a relative ozone loss of 26%, which is in accordance with the estimates derived from the in-situ aircraft observations by HAGAR or by ozonesonde data (Hosen et al., 2013; Rex et al., 2012). The model ozone is estimated by comparing the run with full chemistry with a run with ozone-depleting chemistry switched-off as in Isaksen et al. (2012) (Figure 21: no chemistry is green curve). The model estimate for ozone loss reaches about 0.5 ppm at the end of March 2010, indicating that in warm years, the ozone loss in Oslo CTM is under estimated.

3.6 Integration of results in a chemistry climate model for more reliable predictions of stratospheric ozone and climate (WP 4)

The airborne campaign and laboratory work performed in RECONCILE have contributed to reduce uncertainties in the processes leading to polar ozone loss. An ensemble of chemistry-climate model (CCM) simulations has been performed in order to assess the **impact of the improved knowledge inherited from RECONCILE on the prediction of future stratospheric ozone loss**.

Improvements in the chemistry-climate model

All CCM simulations have been carried out with the LMDz chemistry-climate model coupled to the Reprobus chemical model (Hourdin et al., 2012; Jourdain et al., 2008; Marchand et al., 2012). In its standard version, this model includes a PSC scheme with one small ($r = 0.5$ µm) and one large ($r = 6.5$ µm) mode of NAT particles. This is a simple equilibrium scheme but with a treatment of "NAT rocks" that is usually absent in current CCMs. For RECONCILE, an **improved parameterisation of PSCs** directly inspired from the results obtained in the other work packages has been developed and implemented in the CCM. The new PSC scheme takes into account a tracer keeping the memory of the time spent under T_{NAT}, allowing a more realistic slow growth of NAT particles. Heterogeneous nucleation of NAT is now allowed above the ice frost point, but one week of temperatures continuously below T_{NAT} is required in a given air mass to grow a PSC particle of $r = 7.5$ µm. This is in contrast with the previous equilibrium scheme for which NAT rocks were formed as soon as temperatures dropped below T_{NAT}. Hence, it is expected that the new PSC scheme produces less denitrification than the original equilibrium scheme. Another improvement of the CCM concerns the kinetics of the ClO_x partitioning and ozone loss. The **new kinetics parameters implemented in the CCM** are identical to those summarized in Table 3 and offer the greatest consistency with the experiments and observations made within RECONCILE.

Long-term simulations: experimental setup

External forcings (such as greenhouse gases or ozone depleting substances) used in the simulations are those prescribed in the CCMVal-2 international project of evaluation of chemistry-climate models (SPARC, 2010). Three CCM simulations have been integrated over the period 1960-2050: the reference simulation (REF) with the equilibrium NAT scheme, a "corrected temperature" simulation (TC) with the same equilibrium NAT scheme but with polar temperatures corrected from their past bias relative to ERA-Interim, and a "RECONCILE" simulation (NMP) with corrected temperatures and improved microphysics and kinetics.

Past ozone changes 1960-2005

The springtime total ozone values calculated by the CCM in polar regions from 1970 to 2005 are shown in Figure 22. In the Arctic the strong inter-annual variability of ozone and the small temperature correction make it difficult to identify significant systematic differences between the three runs. There is no clear evidence that one version of the model performs better than the others. The "corrected temperature" simulation (named TC in the Figures) tends however to produce the lowest Arctic ozone values, especially after 2000. This is expected since the TC simulation considers slightly lower temperatures than in the reference run and has a fast denitrification scheme. The RECONCILE run (named NMP in the Figures) has a slower denitrification scheme. It produces winters with Arctic ozone columns generally lower than in the reference run, but is characterized by the strongest inter-annual variability. In the southern hemisphere, polar ozone has a weaker inter-annual variability making it easier to see the expected effect of the increased halogen loading on ozone, especially in October. The temperature correction in the TC simulation reduces the negative temperature bias and results in higher ozone column values, especially after the 1980s, correcting partly the large negative ozone bias of the REF simulation. The RECONCILE run shows Antarctic ozone column values that are close to the values of the "corrected temperature" (TC) simulation during the 1970s. Afterwards, the new PSC parameterisation acts to reduce the strength of the denitrification resulting in significantly higher ozone columns compared to the TC simulation. The large negative ozone column bias during the 1990s and beginning of 2000s seems to have completely disappeared in the NMP simulation. **The RECONCILE simulation reproduces very well the past evolution of the Antarctic ozone column since 1980.**

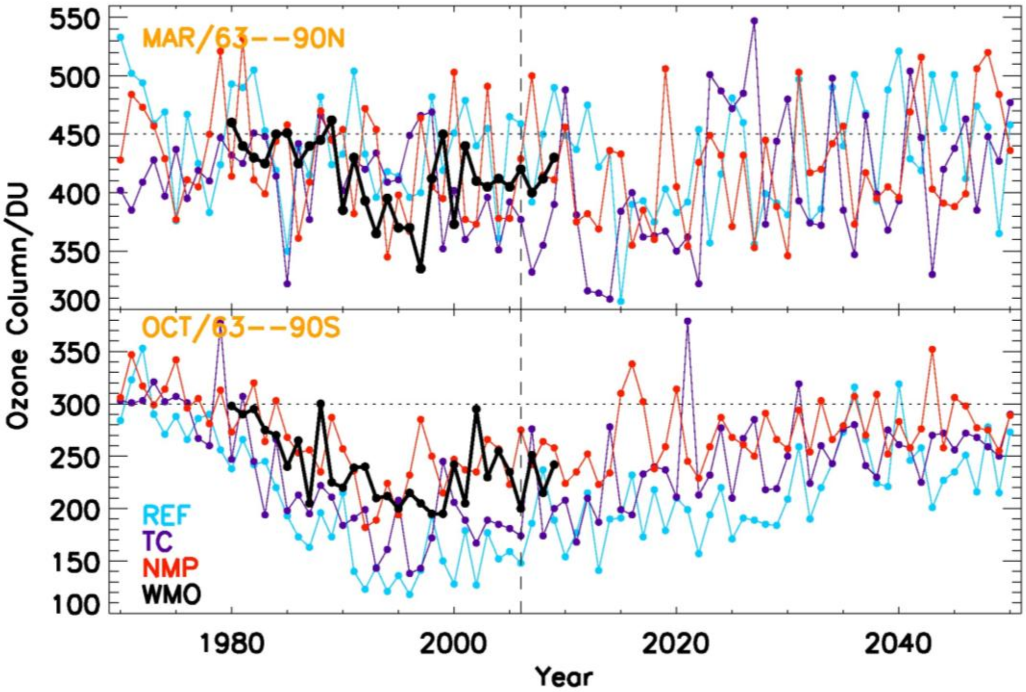

Figure 22 The springtime total ozone values (DU) calculated by the CCM in polar regions from 1970 to 2050. Top: March, northern hemisphere 63°N-90°N. Bottom: October, southern hemisphere 63°S-90°S. Cyan (REF): CCM reference simulation. Purple (TC): CCM corrected-temperature simulation. Red (NMP): CCM Reconcile simulation (NMP). Black (WMO): TOMS/OMI/SBUV merged satellite data.

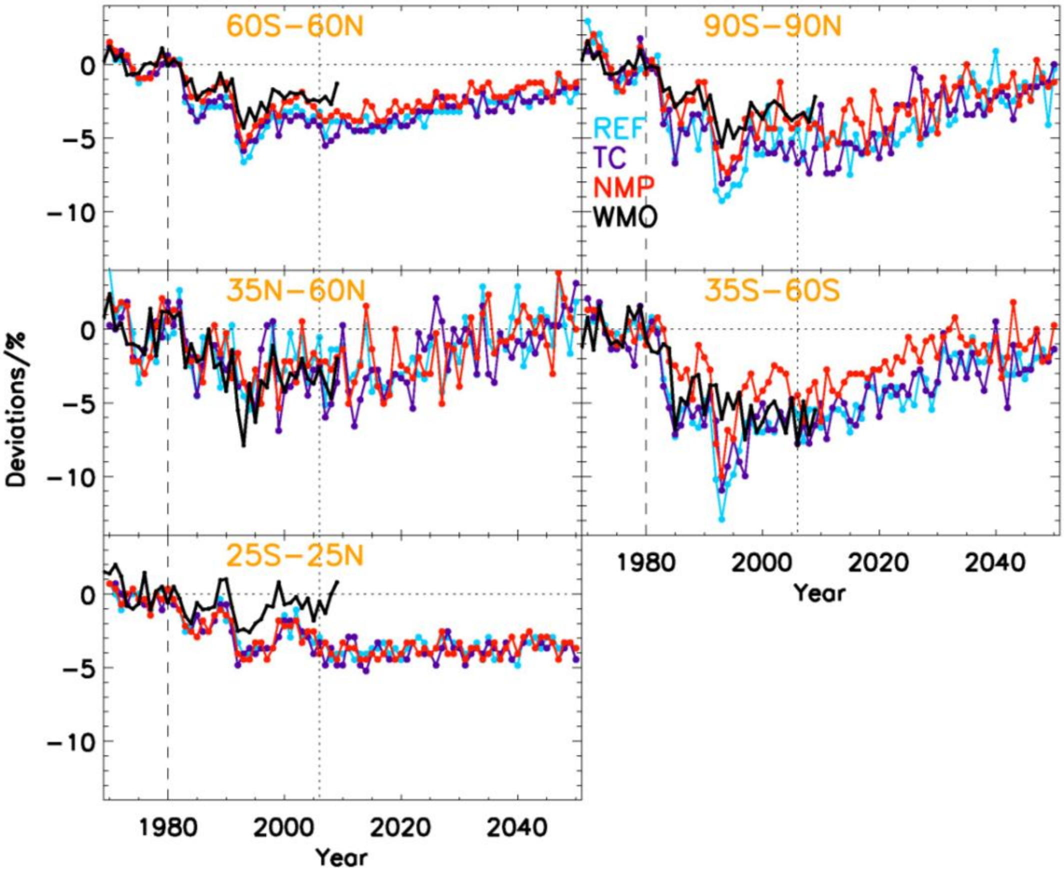

Figure 23 Annual mean area-weighted total ozone deviations (%) from the 1970-1980 mean calculated by the CCM from 1970 to 2050. Different latitude bands are represented. Cyan (REF): CCM reference simulation. Purple (TC): CCM corrected-temperature simulation. Red (NMP): CCM Reconcile simulation (NMP). Black (WMO): TOMS/OMI/SBUV merged satellite data. Each data set is deseasonalized with respect to the period 1970-1980.

The total ozone deviations for various latitude belts are shown in Figure 23. When averaged over the globe, the ozone trends computed by the reference simulation are stronger than observed trends. The temperature correction tends to reduce this bias, but the **new PSC parameterisation seems to improve further the simulation of recent trends**. This better performance of the RECONCILE simulation can be attributed to more realistic amounts in the polar regions, since all simulations perform about equally in the mid-latitudes and the tropics. In the SH hemisphere however, the RECONCILE simulation stands out with column ozone anomalies around -3 or -4% in the last 10 years instead of -6% in the observations, which has to be related to the shallower and less persistent ozone hole produced in that run. All the simulations also show a very large drop in ozone column in the SH mid-latitude following the Mt Pinatubo eruption in 1991, whereas the observations do not show a noticeable anomaly. This major discrepancy has already been pointed out in other studies. Interestingly, the RECONCILE simulation shows much less negative ozone column anomaly following the volcanic eruption of El Chichon in 1982 whereas the other simulations and the observations indicate a drop of about 3% following this eruption. This suggests that the effect of highly enhanced aerosol levels on stratospheric ozone destruction can be masked by other effects, possibly associated with natural variability, not only in reality but also in models.

Future ozone changes 2005-2050

The ozone changes predicted in both polar regions over the period 2005-2050 are shown in Figure 22. In the Antarctic spring, all simulations show a gradual increase in ozone from the last decade of the 20th century at about the same time as the Cl_y abundances have reached their maximum and begin to decline. Projected ozone levels are generally larger in the RECONCILE simulation, which before 2020 gives three occurrences of limited ozone depletion leading to “pre-ozone hole” October values. The difference with the other runs tends however to decrease with time. This is expected in a future atmosphere where PSCs and chlorine should play a lesser chemical role. As a result, the **stable return to pre-ozone hole October values does not occur before 2050 for all simulations**, in good agreement with previous CCM predictions (SPARC, 2010). Results in the Arctic are more difficult to evaluate due to the chaotic nature of the dynamics. There are however occurrences of very low March ozone values (~300 DU) before 2020 in the REF and TC simulations. These episodes of strong ozone depletion are not observed in the RECONCILE run. After 2020, the three simulations show a similar long-term positive O_3 trend. The March ozone level of 1970s is crossed towards 2030. This result is also in line with previous CCM studies (SPARC, 2010).

Future trends for global, midlatitude, and tropical ozone are shown in Figure 23. In a globally-averaged sense the temperature correction and the new PSC microphysics have little impact on the projected ozone trends. The return to the 1970s level occurs a few years after 2050 for the three simulations. The greatest difference between the three experiments is observed in the SH midlatitudes, where the RECONCILE simulation predicts a recovery to the 1970s values about 10 years earlier (2030) than the other simulations. This results from the faster replenishment of the ozone hole in years 2000-2020 (Figure 20). In the tropics, the more intense upwelling by the Brewer-Dobson circulation prevents any significant ozone recovery over years 2005-2050. This effect is caused by the increased concentrations in greenhouse gases and is notably difficult to reproduce by CCMs, which usually show a large spread in projected tropical ozone trends (WMO, 2010).

In summary, our simulations show that **correcting the cold bias in the LMDz CCM is not sufficient to reproduce properly the past ozone loss over the Antarctic. In this region the new RECONCILE PSC and ClO_x parameters have a clear positive impact on the results.** Due to large inter-annual variability this effect is difficult to identify in the Arctic, and further analysis will be needed after the project. Regarding future trends, the RECONCILE parameters have in general little impact on the results. However, it must be noted that **the CCM now includes a more realistic description of processes that are key for polar ozone loss**. For that reason it is clear that **the robustness of the past and future ozone trends calculated by the LMDz CCM has improved with RECONCILE**.

3.7 Additional project outcomes not directly related to original project objective and work packages

Record Arctic ozone loss in spring 2011

Observations and analyses

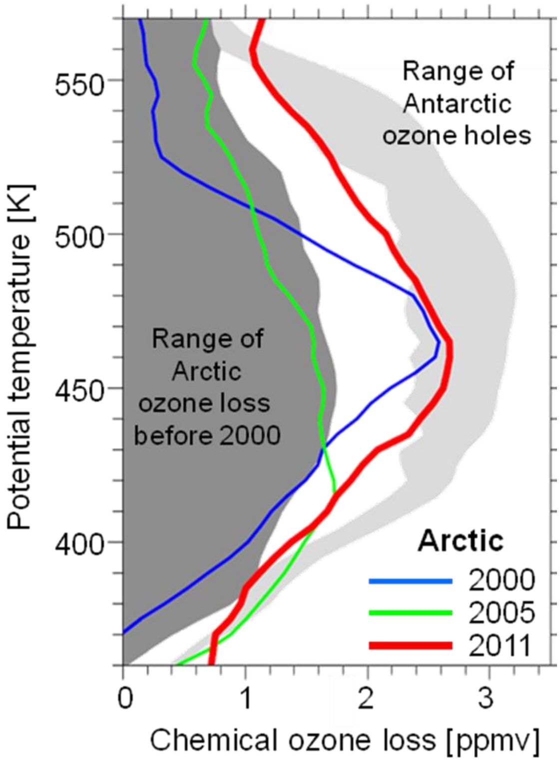

Figure 24 Vertical profile of chemical ozone loss for 2011 and previous Arctic winters derived from Match ozone sonde measurements.

In terms of geographical extent and persistence of PSC conditions, 2010/11 was one of the coldest Arctic winters on record (Figure 18), 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 (Figure 24) . 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. (2012) also discussed the dynamical situation in both winters and extended the analysis of Manney et al. (2011) by describing the contribution of NOx cycle at higher altitudes. An analysis of the relative roles of dynamics and chemistry in the winter 2011 has been described in Section 3.5.

Impact on surface UV radiation

Von Hobe et al. (2012a, manuscript in preparation) carried out an analysis of a 30-year record of surface UV data products from satellite measurements. Representative results for three Arctic and one Antarctic winter are shown in

Figure 25. The study revealed that even 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.

Antarctic Ozone recovery

A long-term assessment of ozone loss and ozone trends in Antarctica has recently been published by Kuttippurath et al. (2013). 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.

ODPs and GHG potential of CFCs and HCFCs

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., 2012a; Laube et al., 2010b; Oram et al., 2012).

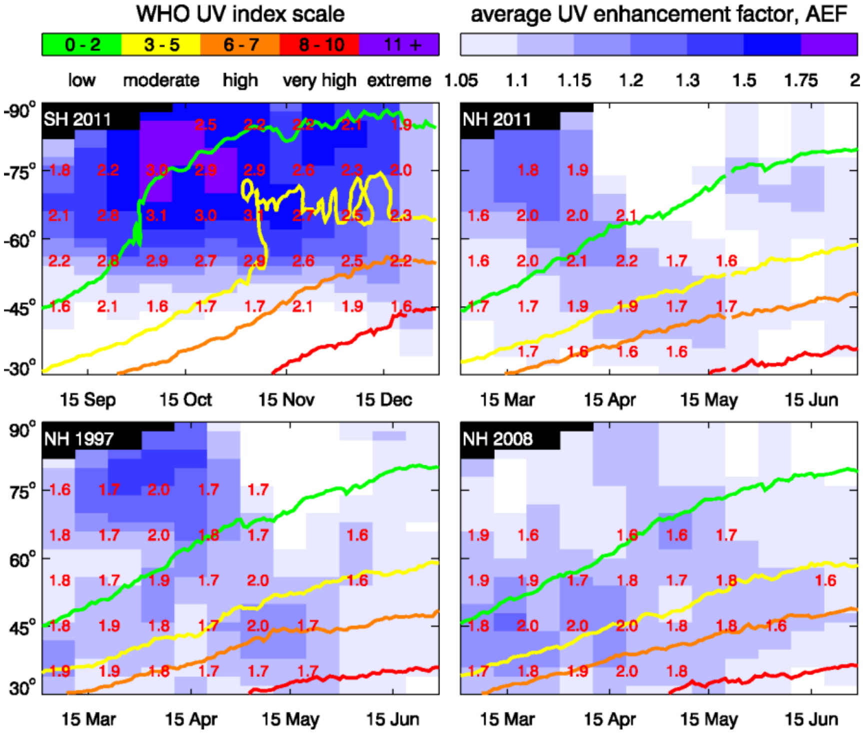

Figure 25 Enhancements in UV radiation over background conditions, determined by normalizing data to 1979-81 averages. Local-noon clear-sky WHO UV index data were taken from a combined MSR/SCIAMACHY dataset (http://www.temis.nl/uvradiation/UVarchive.html) . Area-weighted average UV enhancement factors (AEF) are determined in 2° latitude bands over 10 days. The red numbers show the highest normalized UV values found in 15–day time periods/10° latitude belts (only shown if this exceeds 1.5)

4 Impact

Scientific results, impact and dissemination

Improved process understanding

RECONCILE has substantially advanced our understanding of several key processes in the mechanistic chain that leads to Arctic ozone loss in spring. As described in Section 3 of this report, the project has

- better quantified the rates of chemical reactions in the catalytic ozone loss chains relevant in the polar stratosphere, and essentially removed the last doubts that chlorine – still present in the stratosphere from the anthropogenic CFCs released before the Montreal Protocol – is responsible for this ozone loss.
- corrected and augmented our understanding of PSC formation and microphysics, leading to a more accurate representation of denitrification, one of the key processes determining how much ozone is lost in the Arctic vortex in any particular winter.
- revealed caveats in our understanding of and ability to model transport and mixing across the vortex edge, which has implications for ozone loss estimates and with respect to the export of ozone depleted air to lower latitudes in spring.

Numerous papers on "process understanding" have been – or are still in the process of being – published in the scientific literature. They are to a large extent bundled in a special issue in the EGU (= European Geosciences Union) open access journal *Atmospheric Chemistry and Physics*.

Improved climate models

By completing our understanding of the chemical and dynamical processes affecting polar ozone depletion and transport to lower latitudes, and the translation of this understanding to parameterisations for chemical climate models, **RECONCILE has reduced uncertainties and added further confidence in these models and thus made a significant contribution to improving the predictability of future polar ozone depletion**.

This matches the expected impact of sub-activity 6.1.1.2. in the FP 7 Theme 6 work programme, namely to ***"reduce the uncertainties in the evolution over the next decades of the stratosphere and improve estimates of time scales of ozone layer recovery"*** and to ***"provide information on the interactions of changes in the stratosphere and climate"***.

The improved climate models can now be used together with emission scenarios based on different possibilities of global socio-economic development (a detailed investigation of which was clearly outside the scope of the project) to better constrain the expected evolution of ozone layer recovery, of climate impacts of stratospheric changes, and of the impact this will have on the societies and economies in the mid latitudes.

Contributions on political and societal aspects of ozone depletion

Other studies have also been (are being) published that have a stronger focus on political and societal aspects of polar ozone depletion:

- RECONCILE contributed to studies to better constrain the temporal evolution of ozone depleting and climatically active substances in the stratosphere.
- A study on the first signs of recovery of the Antarctic ozone layer has been published.
- The most severe spring Arctic ozone depletion to date has been detected and characterised.
- The impact of severe Arctic ozone depletion on surface UV radiation has been investigated.

Dissemination with a focus on the general public

Besides the dissemination of results in the scientific literature – the main focus of a process understanding oriented research project such as RECONCILE – a number of measures were taken to address and inform the general public. The most important ones are:

- A project website was installed early in the project period (March 2009) containing information on the project and the consortium, as well as background information on polar stratospheric ozone depletion.

The website has been kept up to date with results, links to scientific papers, and other news, and we will continue to update it regularly in the future.

- Several press releases were issued by the coordinator and by the partners leading to articles in local and regional newspapers and radio coverage
- During the European Geosciences Union (EGU) General Assembly 2011 in Vienna, the RECONCILE team supported and participated in a press conference on the record Arctic ozone depletion in spring 2011.
- A campaign video was created during the second part of the RECONCILE aircraft campaign, describing the campaign work and the scope and motivation of RECONCILE. The video was disseminated via the website of Forschungszentrum Jülich and is still available on the RECONCILE website.
- A television feature for a documentary about climate engineering ("The weather makers", shown in "Abenteuer Wissen in the German ZDF) was also made during the RECONCILE field campaign.
- A "campaign blog" from Kiruna was featured on the RECONCILE website. It presented information on "the scientific campaign work and beyond".

Integrated approach

The RECONCILE project has applied a unique integrated approach using laboratory studies, field experiments, and simulations, drawing together the different communities to collaborate on an extraordinary level. For example, the collaboration between the laboratory and field scientists has aided to resolve the discrepancies related to ClOOCl photolysis and the kinetics of the ClO dimer cycle. The synergetic use of in-situ data, remote sensing observations and modelling tools and the good communication between the scientists involved has been especially fruitful in the context of understanding PSC formation and evolution and the vertical redistribution of denitrification in the vortex. Here, models are now close to reconciling all the – at first look sometimes contradictory – evidence from laboratory and field experiments.

The involvement of a CCM partner and the direct linkage of the process understanding to the chemistry climate modelling has ensured the direct utilisation of the process study results for the overarching goal of ***"improving the predictability of future ozone depletion"***. Publication of the RECONCILE CCM work and presentation on international conferences and meetings is still ongoing. In the coming months and years, a substantial impact and visibility of RECONCILE in international climate change research programmes and reports (cf. below) and in the larger scientific community can be expected.

Positioning in the international context of research

RECONCILE has build on a number of successful past international research projects on polar ozone depletion (e.g. SOLVE-THESEO, VINTERSOL-EuPLEx). By addressing some of questions that had not been fully answered and questions that had only arisen within these past projects, RECONCILE has continued in the successful tradition of "polar stratospheric ozone research made in Europe".

The participation of leading institutions and research groups in their respective fields of expertise and of *the* European airborne platform for stratospheric research, the Russian high altitude research aircraft M55 Geophysica, has added visibility to the project and ensured the international recognition of the project.

Co-operation with various partners and projects was established to bundle activities, use synergies and create a broader database and exploit it more comprehensively. The most important co-operations are listed in Table 4. A particular fruitful collaboration is the one with the NASA scientists Michael Pitts and Lamont Poole on the understanding of processes related to PSCs.

The RECONCILE consortium and external collaborators together represent a significant part of the stratospheric research community, and many of the scientists involved in the project serve as co-authors of the **WMO ozone assessments and IPCC climate reports**. The project results published over the past three years will directly contribute to the 2014 WMO assessment. The corroboration of many cornerstones of our knowledge of ozone depletion and the fine-tuning of those processes where significant uncertainties still existed will help to sustain and enhance the credibility of the ozone layer science and policy, with indirect implications for the credibility of the science and policy on global climate change.

Table 4 IMPORTANT CO-OPERATIONS WITH OTHER PROJECTS AND SCIENTISTS

Group/Project	Contact Person	Short Description of Activities
CALIPSO	Michael Pitts	Polar Stratospheric Cloud analyses from CALIOP spaceborne lidae
	Hideaki Nakajima	Ground based LIDAR observations from Ny Alesund, Spitsbergen
Univ. Stockholm	Farah Khosrawi	Ground based LIDAR observations from Esrange (near Kiruna)
FMI LAPBIAT-II	Rigel Kivi	Ozone sondes, aerosol backscatter sondes, COBALD sondes, hygrometers FLASH-B and CFH from Sodankylä, Finnland
ACE-FTS	Kaley Walker	ACE-FTS satellite observations
AURA-MLS	Michelle Santee	AURA-MLS satellite observations
Univ. Maryland	Ross Salawitch	CTM modelling, in particular photochemistry
Belgian Inst. for Space Aeronomy	Sébastien Viscardy	Developing parameterisations for modelling of PSCs in a CTM
IMAU, Utrecht University	Thomas Röckmann	WAS Measurements (cf. WP 5)
University of East Anglia	Johannes Laube	Analysis of WAS whole air samples (cf. WP 5)
Technical Univ. Darmstadt	Martin Ebert	Laboratory analysis of impactor samples collected during the RECONCILE Geophysica flights (cf. WP 2 and WP 5 in Section 3.2.2)
SHIVA	Klaus Pfeilstiker	Information on the total amount of stratospheric Bry and its trendand the contribution of VSLS species to the stratospheric halogen budget.
GSMA (Reims) / LMD/CNRS	Emmanuel Riviere	Analysis of the PSC occurrence as observed by CALIPSO, detailed study of observed cases using MiPLaSMO, Satellite-Match MIPLASMO - CALIPSO, Simulation of mountain wave activity running the WRF or the RAMS mesoscale model, possibly MiPLASMO simulations along MATCH trajectories. One major goal is the improvement of the NAT scheme in MiPLASMO (proposal within French CHAT-LEFE program)
Univ. Leeds	Martyn Chipperfield	Improve representation of polar processes in TOMCAT/SLIMCAT CTM and resolve differences in chlorine activation, denitrification and O_3 loss compared to observations and other RECONCILE models. Develop a parameterisation scheme for denitrification in CCMs and evaluate/improve representation of all stratospheric chemical processes in new UKCA model. Develop flux-based emissions for all important long-lived halocarbon source gases; use them in future CCM simulations to 2100. Proposal in UK call submitted.
Envival Life	Mark Weber	Use of M55 data (HALOX, ...) for ENVISAT validation
Sharp	Ulrike Langematz	
Oxford University	Anu Dudhia	Retrievals of MIPAS-Envisat data: cloud tops, temp, O_3, N_2O, CH_4, H_2O, CFCs, $ClONO_2$, HNO_3, N_2O_5, NO_2
JEM-SMILES	Yasko Kasai	Provision of JEM-SMILES data of O_3, HCl, ClO, and BrO; use the Geophysica data for JEM-SMILES validation
NCAR	Simone Tilmes	Better constrain exchange processes between upper troposphere (UT) and lower stratosphere (LS) in Polar regions using tracer profiles.
ODIN	Joachim Urban	Satellite Observations: routine two-day SMR data (ClO, O_3, ...)
CU Boulder	Darin Toohey	Cooperation on HALOX ClO/BrO measurements and related science

Technological advance

The M55 Geophysica aircraft is one of the four – and the only non US – aircraft available worldwide for probing the atmosphere above 16 km altitude, and the use of this *European* high altitude research platform was absolutely essential to the RECONCILE science objectives. Via funding of a large Geophysica field mission, **the RECONCILE project, on the other hand, helped to ensure that this unique tool M55 Geophysica continues to be available to the European atmospheric research community for future missions.**

Two technological advances on the instrumentation have moved forward the state of the art in airborne field observations:

- Particles in the sub-micrometer size range were sampled in the stratosphere onto boron substrates (in an impactor probe placed in the COPAS inlet, cf. Section 3.1) and subsequently analysed in the laboratories at the TU Darmstadt using state-of-the-art analysis techniques such as environmental scanning electron microscopy connected to energy dispersive x-ray analysis. This enabled us to perform one of the sparsest samples of stratospheric sub-micron aerosol particles worldwide, particularly in the Arctic Winter vortex to study their morphology and chemical composition to get insights into their origin and their history in the atmosphere. (cf Secction 3.4). The RECONCILE campaign marked the very first deployment of this Particle sampler that is currently being developed further (e.g. to collect multiple samples in one flight frequency) and will be used in future campaigns to gain insight in the processes leading to the formation of stratospheric aerosols and high tropospheric clouds. A German national project with a strong focus on such measurements, SPITFIRE, has received a positive funding decision, and they are also proposed in a large collaborative project, StratoCLIM, in the EC's call ENV-2013 (funding decision pending).
- A range resolving channel was newly implemented and deployed for the first time in the MAS particle instrument, essentially transforming it into a sideward facing lidar. Together with the upward and downward facing MAL instruments, this allows for a powerful, tomographic-like, representation of the PSC field.

Besides the instrumentation flown on the aircraft, the successful operation of the newly developed COBALD optical backscatter sondes also marks a technological advance in terms of atmospheric aerosol and cloud observations. Applying new high-power LEDs and digital lock-in signal processing made the instrument very compact and lightweight so that during the RECONCILE campaign from Ny Alesund, the operational radiosonde/ozonesonde payloads could be complemented with aerosol and cloud particle characterization. The additional in-situ information delivered by COBALD was extremely helpful for the interpretation of the radiosonde data and provided important information on stratospheric aerosol and PSC particles used for microphysical modelling and for validation of the CALIPSO spaceborne lidar data products.

Training Junior Researchers

Numerous junior scientists have been involved in RECONCILE, and the project has had significant impact on their scientific careers and thus on shaping the next generation of European atmospheric scientists. Eleven PhD projects have been carried out or are just short of completion. These are described in some detail in Table 5. In addition, Master students were involved at most of the university partners.

As project coordinator, Dr. Marc von Hobe (JÜLICH), has further developed his leadership abilities, and will continue to play a leading role in the atmospheric science community. By leading work package 4 and co-leading work package 1 respectively, Dr. Marion Marchand (CNRS) and Dr. Francis Pope (UCAM) have also played leading roles in the project and further advanced their scientific careers.

Table 5 PhD students involved in RECONCILE and their projects

Student	Partner	University issuing degree	Date of defense
	Title		
	Description		
Péter Kiss	ELTE	Eötvös Loránd University, Budapest	14.05.2010
	Analysis of the European Wind Power Climatology and the Possible Cosmic Radiation Forcing on Global Lightning Activity		
	The thesis provides a comprehensive statistical analysis of ERA-40 surface wind fields (probability distributions, spectral properties, comparison with tower measurements, etc.). In the second part lightning frequencies are compared with various parameters characterizing the strength of cosmic radiation. (Activities in Reconcile were independent from the subject of this thesis with a large overlap of exploited methodology.)		
Olga Suminska-Ebersoldt	JÜLICH	University of Wuppertal, Physics	05.03.2012
	Stratospheric ClOOCl chemistry at high solar zenith angels		
	The thesis focuses on the chemistry of ClOx cycle, important but still not well parameterized chain of processes leading to the ozone depletion in the polar stratosphere. The data sampled during RECONCILE enabled narrowing of the range of possible ClOOCl photolysis absorption cross sections and of the ClO/ClOOCl thermal equilibrium constants, a significant ClOOCl photolysis longward of 420 nm was investigated and ruled out. Comprehensive sensitivity studies revealed that at high solar zenith angles albedo and tropospheric clouds do not influence the ClOOCl photolysis rate significantly and that the impact of ozone and aerosol is moderate.		
Christoph Kalicinsky	BUW	University of Wuppertal, Physics	19.10.2012
	CRISTA-NF observations in the vicinity of the polar vortex		
	The IR spectra measured by CRISTA-NF during the RECONCILE aircraft campaign allow for the detection of PSCs and the discrimination of PSC particle types (e.g. observation of ice PSC on 17 January 2010). The derived high resolution cross-sections of several trace gases (e.g. CFC-11, O3, ClONO2, HNO3) enable the analysis of very small-scale transport structures (vertical extent even less than 1 km) in the atmosphere and the retrieved CFC-11 and ozone VMRs are used to distinguish between vortex and non-vortex air masses. Comparisons between the CRISTA-NF results and simulations by the CTM CLaMS show the capability of the model to reproduce even small-scale transport structures and demonstrate the usability of a model concept which is based on artificial tracers and can be used to analyse the origin of the observed air masses.		
Ines Engel	ETHZ	ETH Zürich	18.02.2013
	The Role of Heterogeneous Nucleation in Polar Stratospheric Cloud Formation: Microphysical Modeling		
	The thesis revisits the microphysics of PSCs by means of comprehensive modelling studies and leaves little doubt about the importance of heterogeneous processes. To obtain model agreement with observations, NAT and ice particles must form heterogeneously on preexisting solid particles. In addition, small-scale temperature fluctuations are crucial to simulate ice number densities similar to those inferred from the observations.		
Tobias Wegner	JÜLICH	University of Wuppertal, Chemistry	01.03.2013
	Chlorine Activation and Heterogeneous Chemistry in the Polar Stratosphere: Model Simulations, In-Situ and Satellite Observations		
	The thesis investigates the role of heterogeneous chlorine chemistry on cold binary aerosol using in-situ observations, satellite data and the CLaMS CTM. Besides, modifications to the representation of PSCs in the CTM version version of WACCM are described that improve the agreement with observations of PSCs and chlorine compounds. New processes have also been included in WACCM to improve the agreement between simulated and observed HCl in the core of the Antarctic vortex.		
Wolfgang Woiwode	KIT	Karlsruhe Institute of Technology, Chemistry	19.04.2013
	Qualification of the airborne FTIR spectrometer MIPAS-STR and study on denitrification and chlorine deactivation in Arctic winter 2009/10		
	The thesis describes the qualification of the airborne FTIR spectrometer MIPAS-STR (Michelson Interferometer for Passive Atmospheric Sounding-STRatospheric aircraft) and studies on the processes of denitrification and chlorine deactivation in the Arctic winter lower stratosphere in early 2010 during the RECONCILE campaign. Using the MIPAS-STR measurements, HNO$_3$-redistribution trough denitrification as well as chlorine deactivation via the reaction into ClONO$_2$ is analysed for specific flights. Furthermore, a combined study with MIPAS-STR measurements and the CLaMS model was carried out, addressing the properties of large HNO$_3$-containing particles.		
Elisabeth Hösen	BUW	University of Wuppertal, Physics	Exp. May 2013
	Untersuchung von Transport, Mischung und Ozonverlust in der arktischen Polarregion im Winter 2009/2010 basierend auf flugzeuggestützten in-situ Messungen		
	The vortex index, calculated from N$_2$O measurements by HAGAR (along with measurements of CFC12, CFC11, H1211, SF$_6$, CH$_4$, H$_2$ and CO$_2$), allows to denote the characteristic origin of an air mass with respect to the vortex. For the RECONCILE winter 2009/10, an intrusion transported and mixed into the vortex, an extrusion of vortex air and a tropical streamer transported and mixed into the surf zone as well as small scale mixing across the vortex edge have been identified by the vortex index, by analysing the evolution of the F11-N$_2$O correlation and by comparisons with CLaMS simulations. The chemical ozone loss inside the vortex until the beginning of March was derived by analysing the evolution of the O$_3$-N$_2$O correlation taking mixing into account by looking at the temporal evolution of this correlation.		
Sergey Molleker	MPG	Johannes Gutenberg University Mainz	Exp. July 2013
	Charakterisierung von optischen Partikelspektrometern und in-situ Messungen zur Mikrophysik der polaren stratosphärischen Wolken		
	The in-situ optical particle spectrometers used during the Geophysica campaign are characterised and observed size distributions of PSC particles are presented and discussed. A special focus is given to large NO$_y$ containing particles, the so-called NAT Rocks.		
Viktória Homonnai	ELTE	Eötvös Loránd University, Budapest	Exp. end of 2013
	Trends and Correlation Properties of Stratospheric Water Vapor and Ozone Concentrations		
	In the first part of thesis, balloon profiles of water vapour are evaluated over Boulder. Statistically significant trends can be identified in very restricted range of the lower stratosphere. In the second part, a comprehensive spectral and correlation analysis is provided for global total column ozone from satellite measurements and from two climate chemistry models. The result can help to improve both the dynamical core and the chemistry module of the numerical models. (The second part is fully connected to Reconcile.)		
Isla Young	UCAM	Cambridge University	Exp. end of 2013
	Determination of the UV and visible spectra of chlorine monoxide dimer (ClOOCl) and chlorine (Cl$_2$)		
	A novel temperature dependent dual path spectrometer was developed to measure species of atmospheric interest in both the UV and green spectral regions . This apparatus allowed for the measurement of the UV and visible spectra of the chlorine monixide dimer (ClOOCl) and the temperature dependence of the visible chlorine (Cl$_2$) spectrum.		
Sabrina Ludmann	UHEI	University of Heidelberg	Exp. Early 2014
	UV/Vis Limb measurements from aboard the Geophysica aircraft during the project Reconcile		
	The present PhD reports on novel profile measurements of O$_3$, O$_4$, NO$_2$ and BrO performed by means of UV/Vis limb spectroscopy from abord the Geophysica aircraft in the lowermost stratosphere during the Arctic winter 2010. Major characteristics of the method as well the sensitivity of measurements towards the used a priori information, atmospheric and flight attitude parameters are discussed along with information gained with respect to the photochemistry and budget of stratospheric bromine.		

Involvement of researchers from new EU member states

One of the partners in the RECONCILE consortium has been Dr. Imre Janosi from the E.L. University of Budapest in **Hungary**. This group has made valuable scientific contributions to RECONCILE, and through the project has been closely integrated into the European atmospheric science community, where scientists from new member states are still badly underrepresented. Atmospheric research, especially in the stratosphere is quite demanding, and requires many resources missing in the new EU member states. Integrating groups from such countries in large collaborative projects such as RECONCILE helps them to get access to such resources, helps the research community to utilize the often great expertise of these groups, and helps both to achieve objectives that are in their common interests.

In July 2012, the RECONCILE project and the Hungarian partner ELTE were chosen to present at an international press event in Budapest on EC funded research.

Use and dissemination of foreground

As described in Section 3, a ***comprehensive set of observational data*** was acquired from the Geophysica and the Match sonde campaigns carried out within RECONCILE.

All data were initially archived in a **database maintained by the RECONCILE coordinator Forschungszentrum Jülich** and used for scientific exploitation by project beneficiaries and associated partners, leading to many of the scientific results and publications described above. Recently, the database has been transferred to an open access web server linked to the RECONCILE web page. It is thus put at the disposal of the atmospheric research community at large.

A ***special issue*** has been set up in ***Atmospheric Chemistry and Physics (ACP)***. This journal, run by the European Geosciences Union, provides open access to all its articles and currently has the highest impact factor of any journal dedicated to atmospheric sciences.

Although only the LMDZrepro CCM is used immediately within the RECONCILE project, the improved understanding and parameterisations of important atmospheric processes will be published and thus available to other CCM groups. RECONCILE will therefore lead to an ***advance of CCM capabilities to simulate stratospheric processes in general***.

The contribution that RECONCILE will make to future ***WMO assessments and IPCC reports*** has already been mentioned in Section 3.1. We expect publications coming out of RECONCILE to be cited in both these reports, and scientists involved in the project to contribute directly as authors, contributors and reviewers.

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