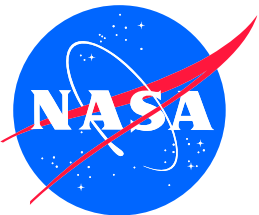# Combining Measurements and Models to Advance Understanding of Polar Processes

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- ✦ An overarching goal of the atmospheric science community is to develop a reliable and accurate predictive capability so that we can assess changes in stratospheric ozone in response to anticipated reductions in chlorine loading and possible climate change, particularly in the Arctic.
- ✦ Fundamental to achieving this goal are process studies addressing specific photochemical, microphysical, or dynamical questions, through which current understanding can be critically tested and potentially important new processes discovered.
- ✦ As more and better atmospheric observations become available and models become increasingly sophisticated, the power of using measurements and models together in process studies also grows.
- ✦ In these lectures we will examine a number of case studies exemplifying how models have been used to aid in the interpretation of measurements in polar process studies.
- ✦ Caveat: This is in no way intended to be a complete survey of the work done using models and measurements together on any of these topics!!

- ✦ Outline:
  - ✧ Defining Polar Processes
  - ✧ Advantages and Disadvantages of Satellite and Aircraft Measurements
  - ✧ Examples of Studies on Stratospheric Chlorine Partitioning
  - ✧ Examples of Studies on Polar Stratospheric Cloud Formation
  - ✧ Examples of Studies on Denitrification
  - ✧ Influence of Meteorological Analyses on Polar Process Studies
  - ✧ Remaining Unresolved Polar Process Questions Requiring Further Measurements/Modelling
  - ✧ EOS Aura Advertisement

The term "**Polar Processes**" encompasses several related topics:

✦ The winter polar vortex as a "containment vessel"
  ✧ vortex development and dissipation, and the timing and variability of these processes
  ✧ vortex strength, morphology, and isolation/permeability
  ✧ unmixed diabatic descent of vortex air
✦ Polar stratospheric clouds (PSCs)
  ✧ phase and composition of PSC particles
  ✧ PSC formation mechanisms
  ✧ extent, duration, and frequency of occurrence
✦ Denitrification and dehydration
  ✧ physical mechanisms leading to these processes, and their interhemispheric differences
  ✧ extent to which they are coupled or independent processes
  ✧ sensitivity of these processes to changes in stratospheric temperatures and/or water vapor
✦ Chlorine activation and deactivation
  ✧ quantitative assessment of chlorine activation/recovery under different aerosol loading conditions
  ✧ degree to which the chlorine budget balances
  ✧ relative importance of continued processing on PSCs or sulfate aerosols in maintaining elevated levels of active chlorine into spring
✦ Chemical ozone destruction
  ✧ quantification of ozone loss throughout the winter in both the Arctic and the Antarctic
  ✧ relative contributions of dynamical/transport and chemical processes to long-term ozone changes

Models and measurements have been combined in studying all of these issues, but we concentrate here on examples involving **chlorine activation/deactivation**, **PSC formation**, and **denitrification**.

Polar measurements are made using ground-based, balloon, aircraft, and satellite instruments. Each approach has its own strengths and weaknesses, and data from all of them have been used in conjunction with modelling in polar process studies. We focus here on **satellite** and **aircraft** measurements.
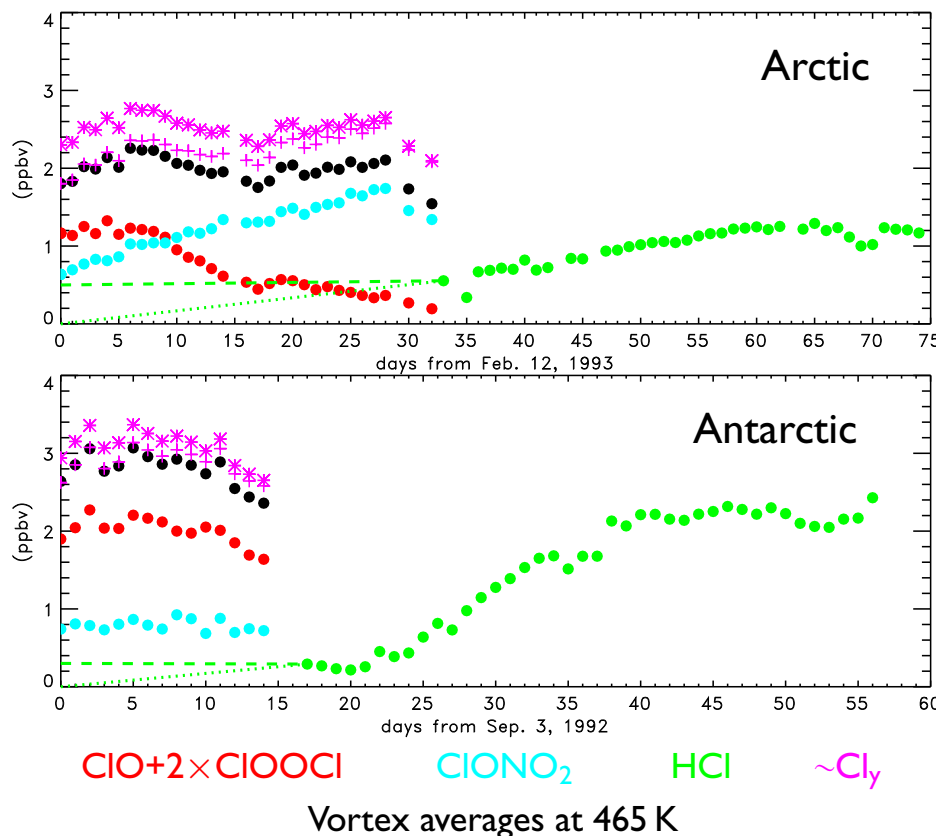
- ✦ Satellite Data:
  - ✧ Global (or nearly so) coverage obtained with a single instrument
    - → facilitates observation of remote places
    - → allows constituent observations to be put into a broader geophysical context and related to the large-scale meteorology of the region in which they were made
    - → allows interhemispheric differences in polar processes to be investigated
  - ✧ Daily (or nearly so) sampling over multiple years
    - → allows intraseasonal and interannual variability to be tracked
    - → facilitates trend investigations
    - → increases the possibility of observing how the global atmosphere responds to perturbations (e.g., large volcanic eruptions, solar variability, meteorological forcing)
  - ✧ Simultaneous, colocated, commonly-calibrated measurements of multiple atmospheric constituents
    - → allows several independent constraints on models, reducing possibility of fortuitous agreement
  - ✧ Spatial resolution often comparable to that of models

- ✦ Aircraft Data:
  - ✧ Much better vertical and horizontal resolution
    - → allows small-scale structure to be resolved
  - ✧ Much better accuracy and precision

- ✦ CFCs released at the ground slowly migrate into the stratosphere, where they are destroyed primarily through UV photolysis, releasing chlorine atoms.
- ✦ Total inorganic chlorine $Cl_y = HCl + ClONO_2 + HOCl + Cl + ClO + 2 \times ClOOCl$.
- ✦ Partitioning of stratospheric chlorine among the reactive species (primarily ClO) that participate in catalytic cycles of ozone destruction and the 'benign' reservoir species (primarily HCl and $ClONO_2$) is crucial for regulating the abundance of stratospheric ozone.
- ✦ Despite its importance, lack of measurements prevented the theoretical understanding of stratospheric chlorine partitioning from being rigorously tested until the launch of the Upper Atmosphere Research Satellite (UARS) in September 1991.
- ✦ UARS instruments measured the main components of Cl_y, as well as other species affecting chlorine partitioning (note that the list of species for each satellite is not complete):
  - ✧ Microwave Limb Sounder (MLS): ClO, O_3, HNO_3
  - ✧ Cryogenic Limb Array Etalon Spectrometer (CLAES): $ClONO_2$, HNO_3
  - ✧ Halogen Occultation Experiment (HALOE): HCl, O_3, NO, NO_2
- ✦ UARS data allowed the $ClONO_2$-ClO and HCl-$ClONO_2$ partitioning to be examined and generally showed good agreement with results from 2D model simulations, suggesting that the major mechanisms governing stratospheric chlorine partitioning are well understood [e.g., Dessler et al., Rev. Geophys. 36, 183, 1998].
- ✦ Normally, ~95% of Cl_y in the daytime lower stratosphere is in the form of HCl and $ClONO_2$.
- ✦ Inside the winter polar vortex, however, heterogeneous reactions on the surfaces of PSC particles convert chlorine from reservoir to reactive forms (**chlorine activation**).
- ✦ UARS MLS ClO observations showed that in both hemispheres the winter polar vortex in the lower stratosphere becomes almost completely filled with enhanced ClO, with abundances large enough to indicate virtually complete conversion of stratospheric chlorine to reactive forms [Waters et al., Nature 362, 597, 1993].
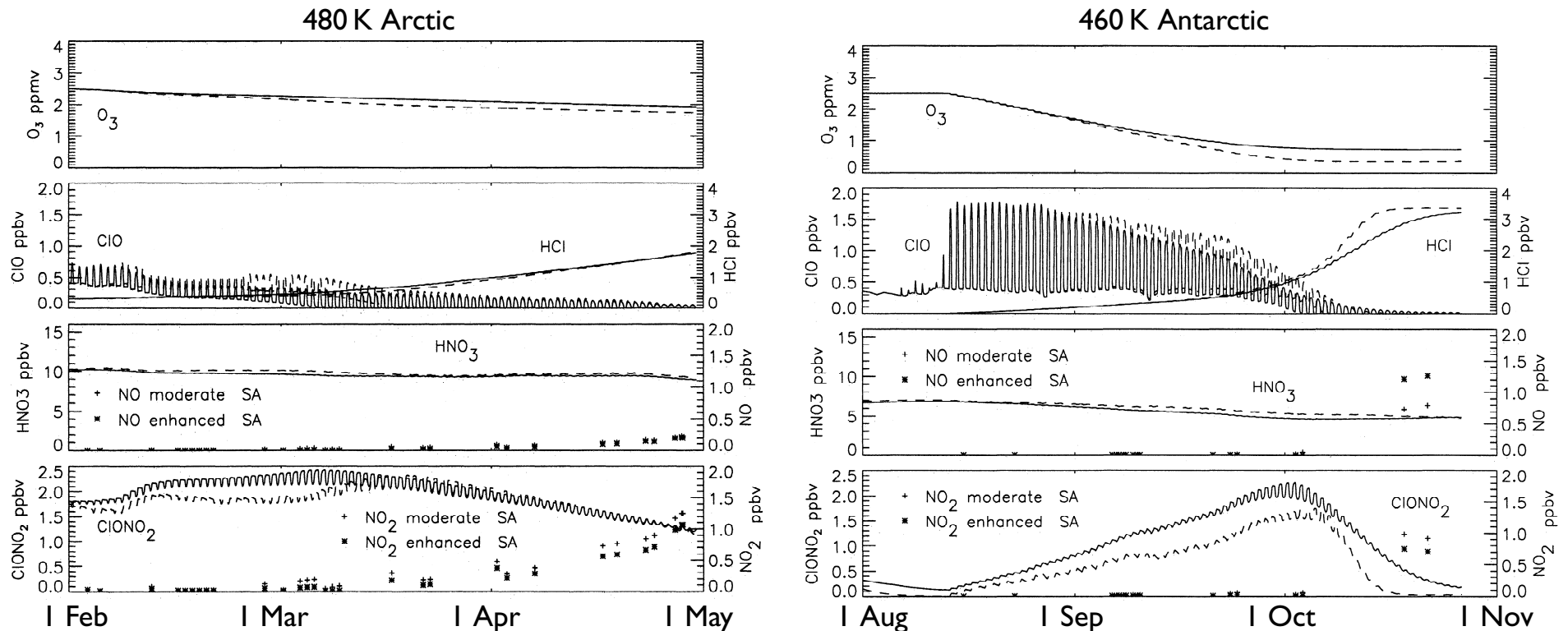
✦ **Deactivation** is the process by which reactive chlorine is converted back into Cl_y reservoir species, curtailing chemical ozone loss.

✦ The two main chlorine deactivation pathways are: $ClO+NO_2+M \rightarrow ClONO_2 + M$, and $Cl+CH_4 \rightarrow HCl+CH_3$.

✦ UARS data revealed an interhemispheric difference in the reformation of chlorine reservoir species in the late winter/early spring.

Arctic

(ppbv)

days from Feb. 12, 1993

Antarctic

(ppbv)

days from Sep. 3, 1992

$ClO+2 \times ClOOCl$ $ClONO_2$ HCl $\sim Cl_y$

Vortex averages at 465 K

Santee et al. [JGR 101, 18835, 1996]

Chipperfield et al. [JGR 101, 18861, 1996]

✦ In the Arctic, the decrease in $ClO+2 \times ClOOCl$ during late winter is balanced by an increase in $ClONO_2$, and HCl increases only gradually.

✦ HNO_3 abundances are sufficient to supply NO_2 for the rapid formation of $ClONO_2$.

✦ In the Antarctic, ongoing PSC activity slows the ClO decline until early spring, $ClONO_2$ remains relatively constant, and HCl increases rapidly.

✦ Denitrification inhibits reformation of $ClONO_2$ by removing gas-phase HNO_3, preventing NO_2 release.

✦ SLIMCAT (3D chemical transport model) results suggest that the primary recovery in the south is not into $ClONO_2$ but HCl, and that the reaction $OH+ClO \rightarrow HCl+O_2$ (previously suggested to be important in the upper stratosphere and subsequently confirmed to occur through laboratory studies) suppresses $ClONO_2$ formation and promotes HCl production.

480 K Arctic

460 K Antarctic

Figure 2. Model calculations are shown for the same time period as the northern hemisphere observations. The solid line is for the model run with the baseline is for a model run that surface area.

Figure 4. Model calculations are shown for the same time period as the southern hemisphere observations. The solid line is for the baseline model; the dashed line is for 4 times the surface area.

◆ Douglass et al. [JGR 100, 13967, 1995] use UARS measurements and a box model to elucidate chlorine deactivation inside the Arctic vortex and the Antarctic "collar" region, where only mild denitrification but extremely low ozone conditions occur.

◆ Following the model predictions of Prather & Jaffe [JGR 95, 3473, 1990] Douglass et al. find that very low O_3 concentrations lead to a highly nonlinear chemical system in which the Cl/ClO ratio is up to 10 times higher, accelerating the formation of HCl through the reaction of Cl with CH_4:

◇ $O_3 > 0.5$ ppmv — rapid $ClONO_2$ formation, slow growth of HCl
◇ $O_3 \lesssim 0.5$ ppmv — rapid HCl formation

◆ Without the ability to explore different scenarios through modelling analysis, the apparent hemispheric inconsistencies in the measurements could not have been explained

$$\frac{[Cl]}{[ClO]} = \left\{ k_{ClO,NO}[NO] + 2 \times J_{Cl_2O_2}\frac{[Cl_2O_2]}{[ClO]} + J_{ClONO_2}\frac{[ClONO_2]}{[ClO]} + J_{HOCl}\frac{[HOCl]}{[ClO]} \right\} \times \{k_{Cl,O_3}[O_3]\}^{-1} \qquad (2)$$

✦ Both box models and 3D chemical transport models substantially underestimate observed ozone loss during recent cold Arctic winters.

✦ One method of quantifying chemical ozone loss is the "Match" technique, in which trajectory calculations are used to identify air masses sampled multiple times over the course of a few days.

✦ Rex et al. [GRL 30, 1008, doi:10.1029/2002GL016008] found that unrealistically high abundances of reactive chlorine are required to explain ozone loss rates derived from Match during several cold Arctic Januaries using standard reaction kinetics, even assuming complete activation of chlorine.

✦ In contrast, Rex et al. showed that ozone loss rates measured in late February/early March 1996 are in excellent agreement with those computed based on UARS MLS ClO observations.

✦ Match trajectories spent a considerably smaller fraction of their overall sunlit time at high solar zenith angles in February/March than they did in January.

✦ These analyses suggest that mid-winter ozone loss occurs by an unknown process involving a photolytic step at high solar zenith angles.

✦ UARS MLS data are too sparse for comparisons in any January or in February/March in any year other than 1996.
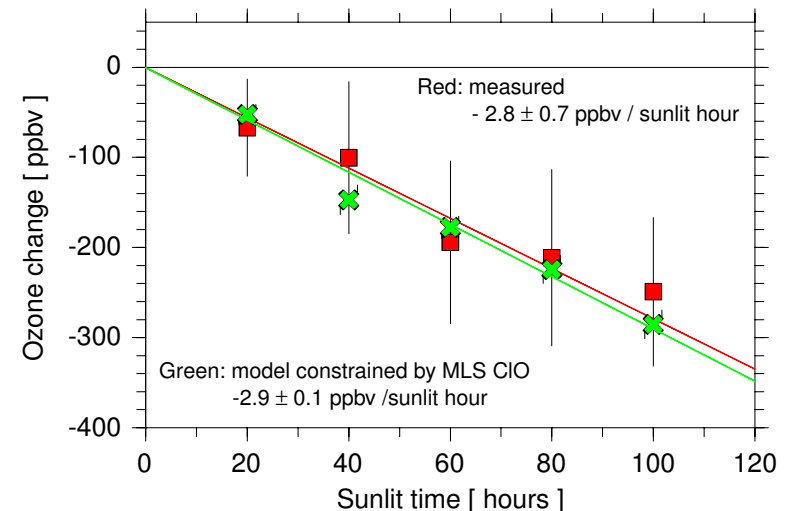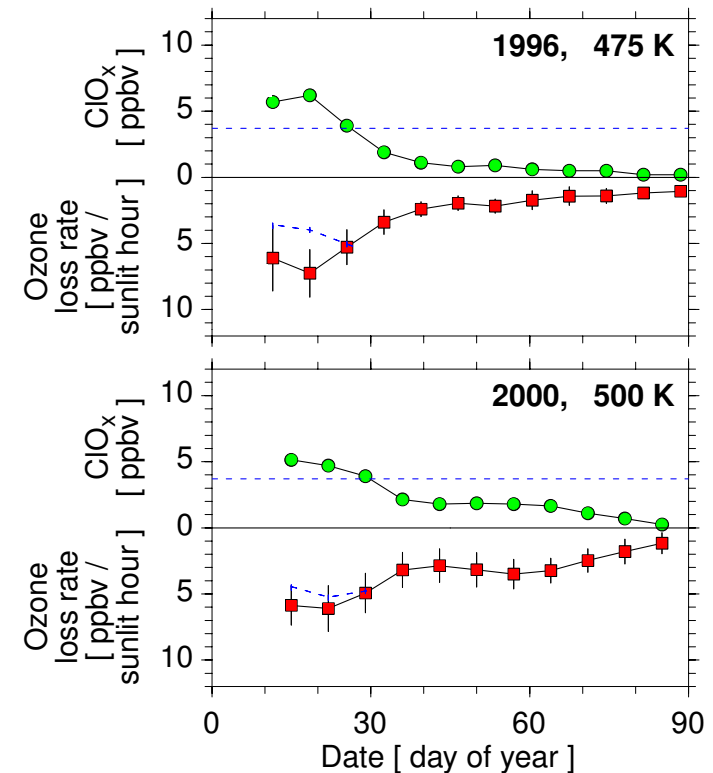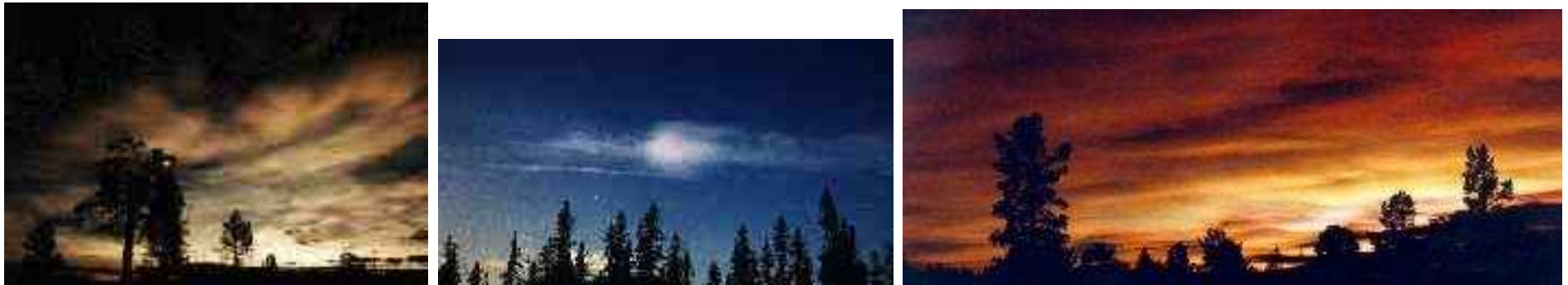
Type I PSCs:

✦ form at $\sim$195 K for typical stratospheric conditions, several degrees above the water ice frost point

✦ provide surfaces on which the heterogeneous reactions activating chlorine can occur, and sequester condensed-phase HNO_3 as a major constituent

✦ are divided into several subclasses, the major ones being:

✧ Type Ia: larger (mode radii of $\sim$1–10 μm), non-spherical, solid particles

→ NAT: nitric acid trihydrate ($HNO_3 \cdot 3H_2O$)

→ NAD: nitric acid dihydrate ($HNO_3 \cdot 2H_2O$)

✧ Type Ib: smaller (mode radii of $< \sim$0.5 μm), spherical, liquid particles

→ STS: supercooled ternary solutions ($H_2SO_4/HNO_3/H_2O$)

✦ specific Type I PSC formation mechanisms remain uncertain

✦ many clouds are likely to have a mixed, or non-equilibrium, composition

✦ sedimentation leads to **denitrification** (irreversible removal of reactive nitrogen from stratosphere)
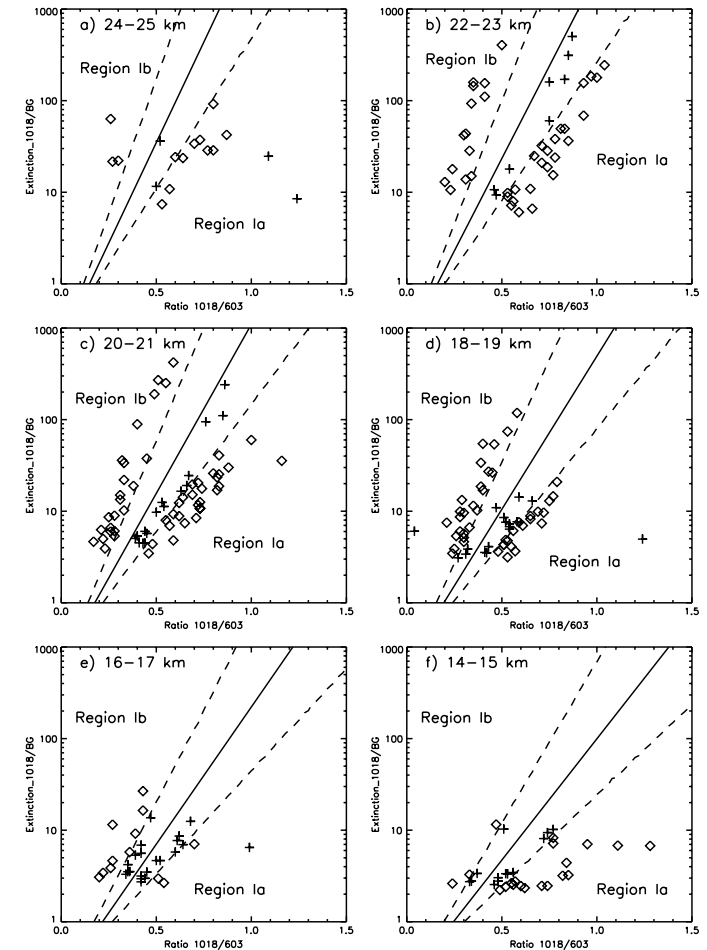
Type II PSCs:

✦ form at temperatures a few degrees below the water ice frost point ($\sim$188 K)

✦ are composed of large (mode radii of $\sim$5–50 μm) water ice crystals, and can also incorporate HNO_3

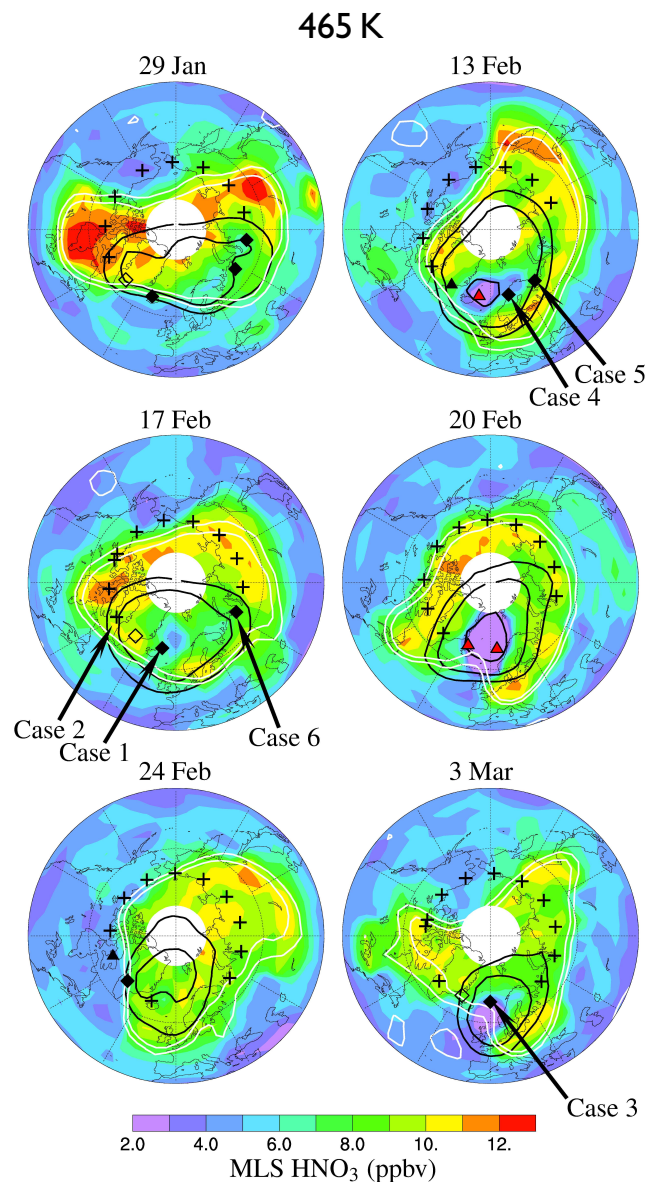✦ sedimentation leads to dehydration (and also denitrification)

✦ Recently, Strawa et al. [JGR 107, 8291, doi:10.1029/2001JD000458, 2002] developed a technique for discriminating Type 1a and 1b PSCs using aerosol extinction measurements from the Polar Ozone and Aerosol Measurement (POAM) instrument.

✦ Idealized simulations using the IMPACT (Integrated MicroPhysics and Aerosol Chemistry on Trajectories) model [e.g., Drdla et al., 2002] show that at equilibrium STS exhibits high extinction and strong wavelength dependence (as expected for a large number of small particles), whereas NAT exhibits low extinction and weak wavelength dependence (as expected for a small number of larger particles).

✦ Plotting the ratio of simulated PSC extinction to background extinction at 1 μm against the wavelength dependence (represented by the ratio of 1.018 μm to 0.603 μm extinctions) reveals that the STS and NAT curves bifurcate as the particles grow and extinction increases.

✦ POAM aerosol extinction observations obtained during the 1999/2000 Arctic winter exhibit a similar pattern.

✦ Scatter in the data may arise from cloud inhomogeneity within the POAM sample volume; intermediate values may indicate mixtures of STS and NAT particles.
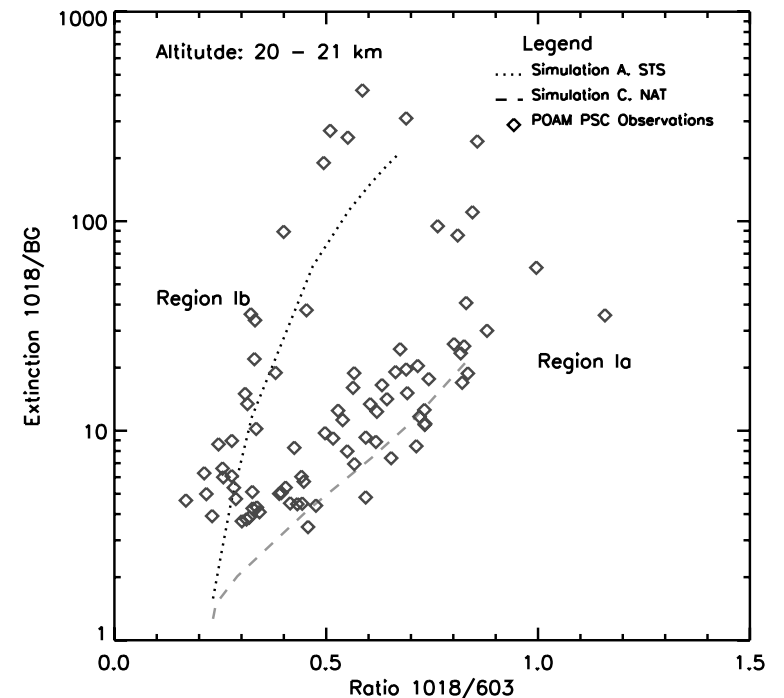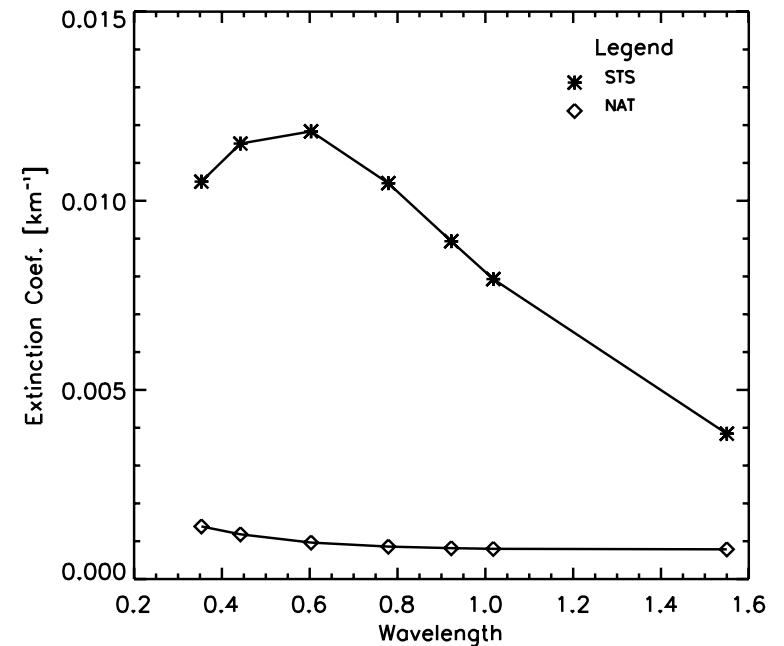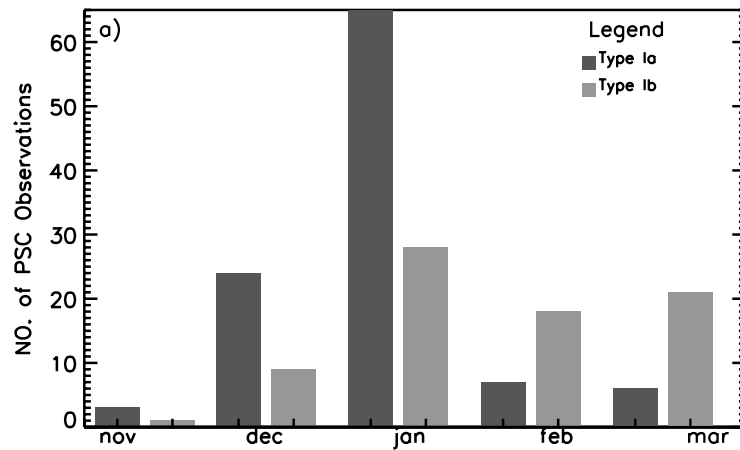
- Guided by the simulations, an altitude-dependent set of lines for discriminating Type Ia and Ib PSCs in POAM data was developed.

- The validity of the classification method was confirmed through comparisons with coincident lidar observations.

- Over the course of the 1999/2000 winter, 32% of PSCs were classified as Type Ia, 23% as Ib, and 45% as undetermined.

- The number of observed PSCs peaked in January, and the percentage of Type Ia PSCs was greatest in December and January.

- In February and March, the percentage of Type Ib PSCs was higher.

- Both are observed at about the same average altitude in December/early January, but the altitude of Type Ia PSCs is 2–3 km lower than that of Type Ib in February/March

- These changes are consistent with the occurrence of denitrification.

- This PSC discrimination method has now also been applied to SAGE III aerosol extinction data [Poole et al., GRL 30, 2216, doi:10.1029/2003GL018496, 2003].

465 K

29 Jan

13 Feb

Case 5
Case 4

17 Feb

20 Feb

Case 2
Case 1   Case 6

24 Feb

3 Mar

Case 3

2.0   4.0   6.0   8.0   10.   12.
MLS HNO$_3$ (ppbv)

POAM Measurements:

+ No PSC   ▲ high Zmin
◇ PSC "2", 16–22 km   ◆ PSC "3", 16–22 km
△ PSC "2", >22 km   ▲ PSC "3", >22 km

✦ In the final example using satellite measurements, Santee et al. [JGR 107, 10.1029/2000JD000227, 2002] employed a Lagrangian approach to track PSC evolution from formation through dissipation by correlating UARS MLS observations of gas-phase HNO$_3$ with POAM observations of aerosol extinction at 1 μm from the Arctic late winter of 1995/1996.

✦ 5-day diabatic air parcel trajectories were calculated both backward and forward from POAM occultation events that were both inside the vortex and at a temperature below 200 K (using meteorological analyses from the UK Met Office).

✦ MLS and POAM data were interpolated to the parcel positions along the trajectory and compared to results from STS and NAT equilibrium composition models before, during, and after a PSC episode.

✦ Overall, a strong correlation was found between low-temperature, low-HNO$_3$, and high-extinction points.

✦ In most cases the observed behavior along the trajectories was within the range predicted by the PSC models.

✦ Thus these satellite HNO$_3$ and aerosol extinction data can be used to track the broad patterns of PSC development.

✦ However, a large degree of overlap in the model/measurement comparisons made it very difficult to determine the specific composition or phase of the PSCs that formed.

✦ One of the most straightforward and least ambiguous examples (out of the 75 cases examined) is shown here.

✦ The MLS HNO_3 data are matched best by the STS model throughout most of the PSC episode, but agreement is not perfect and the data exhibit large scatter.

✦ The POAM extinction data also agree well with the STS model in this case.

✦ In most cases, however, conclusive determination of PSC composition was precluded.

✦ No single set of model assumptions allowed a majority of cases to be explained, and for many cases a variety of different formation scenarios could be invoked.

✦ Factors complicating data/model interpretation include:

  ✧ large uncertainties in both measurements (including the meteorological analyses) and model calculations

  ✧ lack of simultaneous, colocated HNO_3, aerosol extinction, H_2O, and temperature measurements

  ✧ poor horizontal and (especially for MLS) vertical resolution of the data

  ✧ cloud inhomogeneity, either within or between the footprints of the two instruments

  ✧ use of equilibrium PSC composition models

✦ In applying the same technique to ILAS data, Rivière et al. [JGR 108, 4718, doi:10.1029/2003JD003718, 2003] encountered similar difficulties.

(a) Case 1: 17 Feb at 488 K (~20 km), 67.4N, 348.1E
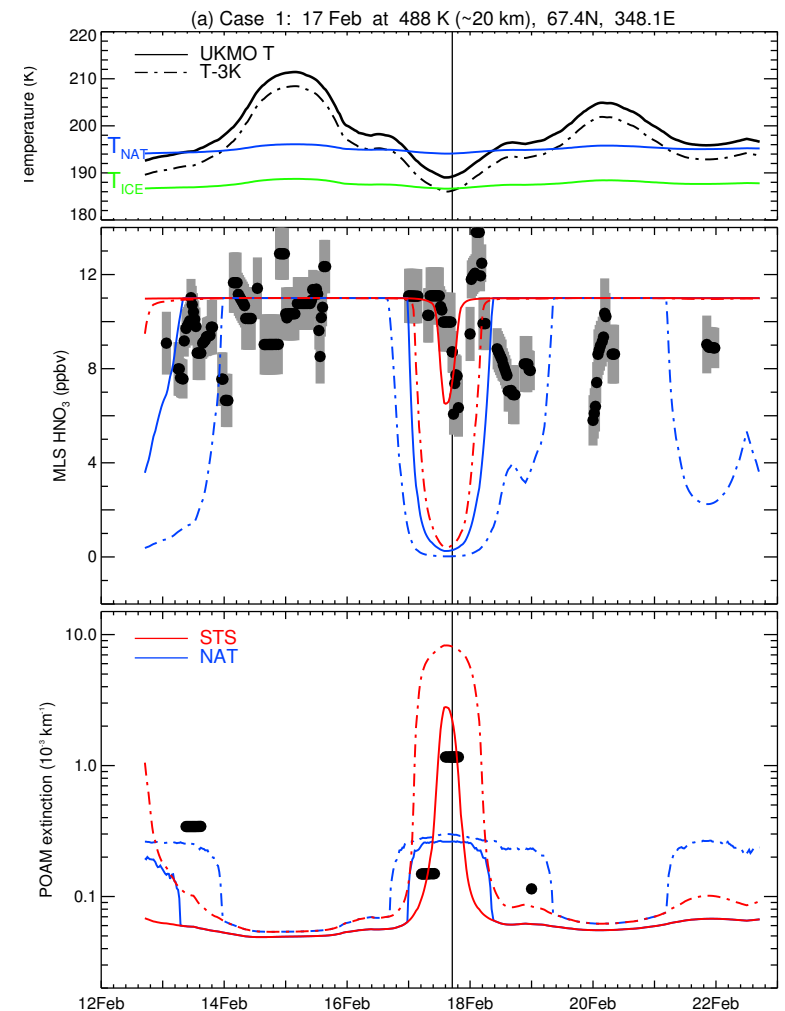
✦ During the 1999/2000 winter, the Arctic experienced exceptionally cold conditions, with temperatures below 195 K persisting over a larger area in the lower stratosphere for a longer period of time than during any of the previous 20 years [e.g., Manney and Sabutis, GRL 27, 2589, 2000].

✦ Thus the 1999/2000 Arctic winter was a particularly fertile period for PSC studies.

✦ Coincidentally, a major NASA/EU field campaign was conducted during this winter: the SAGE III Ozone Loss and Validation Experiment (SOLVE).

✦ One of the most exciting discoveries made during the SOLVE campaign was the detection of small $(10^{-5}-10^{-3}\,cm^{-3})$ concentrations of very large (5–20 μm diameter) HNO_3-containing particles over a broad range of altitudes (15–21 km) and latitudes (60–85°N) [Fahey et al., Science 291, 1026, 2001; Northway et al., JGR 107, 8298, doi:10.1029/2001JD001079, 2002].

✦ These have become known as "Fahey's rocks".

✦ 3D microphysical model simulations show that Fahey's rocks are consistent with NAT PSCs, and that synoptic-scale ice clouds probably do not play a major role in their nucleation and growth [Carslaw et al., JGR 107, 8300, doi:10.1029/2001JD000467, 2002].

✦ Such low number concentrations and large particle sizes imply that a highly selective nucleation mechanism is at work, but its details remain uncertain.

✦ Calculations of the instantaneous downward flux of HNO_3 associated with these large particles indicate that they can cause substantial denitrification (without dehydration) [Fahey et al., 2001].

✦ UARS MLS detected moderate denitrification over a sizeable fraction of the Arctic vortex in 2000, providing the strongest evidence yet for the occurrence of denitrification in the Arctic over spatial scales large enough to be detected in satellite measurements [Santee et al., GRL 27, 3213, 2000].

✦ Aircraft measurements also indicated severe and extensive denitrification during this winter [e.g., Popp et al., GRL 28, 2875, 2001] and showed it to be highly spatially inhomogeneous [Gao et al., GRL 28, 2879, 2001].

✦ Drdla et al. [JGR 107, 8318, doi:10.1029/2001JD001127, 2002] compared in situ Multiangle Aerosol Spectrometer Probe (MASP) measurements of total particle concentration collected during SOLVE to equilibrium model calculations of binary H_2SO_4/H_2O solutions, STS, NAT, NAD, and SAT (frozen sulfuric acid tetrahydrate).

✦ Growth of the largest 5–10% of particles is not detected by the MASP total concentration measurement.

✦ Observed and predicted particle concentrations are plotted as a function of temperature relative to the NAT condensation point (T–T_{NAT}) for various N_2O bins (sulfate aerosol characteristics are strongly correlated with N_2O; $N_2O < 300$ ppbv indicates stratospheric air).

✦ Similar behavior was observed on most flights, with a dramatic increase in particle concentration for T–$T_{NAT} < -4$ K.

✦ The majority of measurement points lie along a smooth curve, implying that particles grew continuously across the range of sampled temperatures.

✦ The smooth growth and the temperature range over which it occurs are inconsistent with the modelled behavior of solid particles (NAT, NAD, or SAT).

✦ The limited scatter in the data suggests that the particles are very near equilibrium.

✦ A liquid ternary solution (STS) model is able to reproduce the observed behavior.
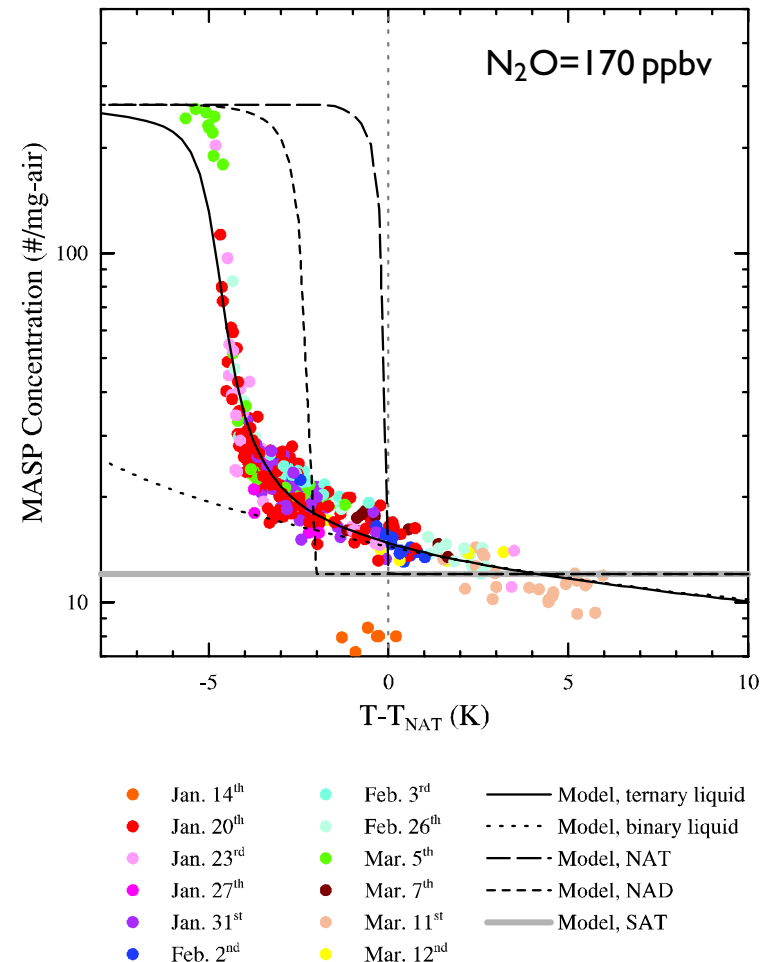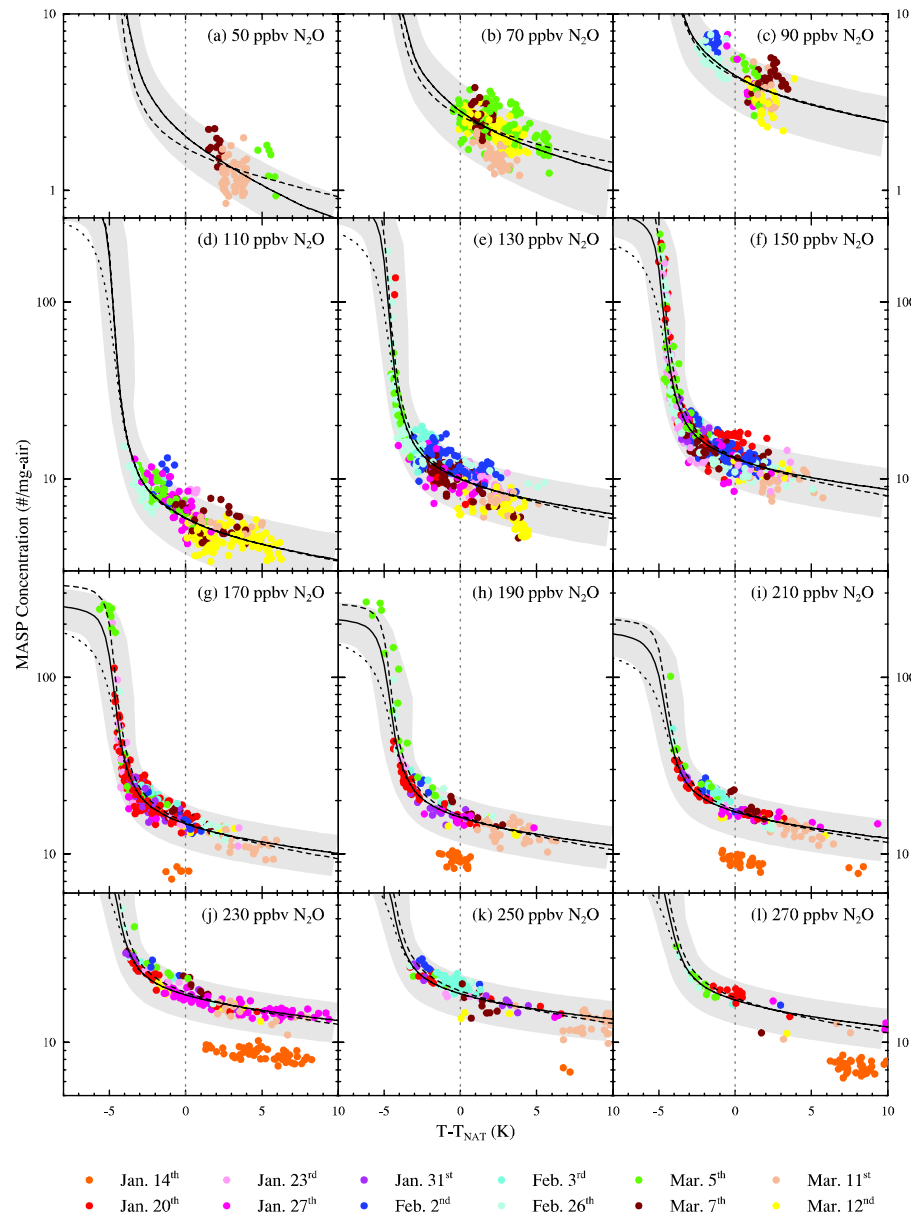
- ✦ At nearly every N_2O level, a wide range of temperature was sampled, allowing changes in particle concentration to be examined.

- ✦ The observed aerosol behavior is consistent with the expected growth of liquid particles, from temperatures 5 K above the NAT condensation point to temperatures 6 K below it.

- ✦ At temperatures more than 3.5 K below the NAT threshold, rapid growth consistent with STS is seen; the observed growth cannot be explained by solid particles.

- ✦ Extrapolating the percentage of MASP particles that are liquid to account for all of the smaller, unmeasured particles that are even less likely to freeze, Drdla et al. infer that more than 90% of the aerosol is liquid.

- ✦ Although there is clear evidence for the presence of very large solid particles during SOLVE, the smaller particles remained liquid, suggesting that the freezing process producing the large particles is selective.

- ✦ The widespread presence of liquid aerosol during SOLVE further implies that even Type Ia PSCs probably contain a mix of particle types.

✦ Drdla et al. [JGR 107, 8312, doi:10.1029/2001JD000782, 2002] used the IMPACT coupled microphysical/photochemical model to investigate the freezing mechanisms that may have led to the formation of solid PSCs during the SOLVE winter.

✦ A number of scenarios were tested through simulations along a large set of winter-long (November 1999 – April 2000) trajectories.

✦ Potential freezing processes examined in the model scenarios include:

   ✧ no freezing ("Liquid")

   ✧ homogeneous freezing to form ice ("IceFrz")

   ✧ homogeneous freezing to form NAT ("HomFrz")

   ✧ heterogeneous freezing to form NAT ("HetFrz")

   ✧ freezing to form NAD ("NADHomFrz" and "NADHetFrz")

✦ Detailed model/measurement comparisons were not performed, but three measurement criteria were used in a qualitative sense to 'disqualify' some of the model scenarios:

   ✧ DC-8 lidar measurements showed that solid-phase PSCs were present by at least 5 December

   ✧ ER-2 measurements of total reactive nitrogen (NO_y, principally HNO_3 during polar winter) showed that most of the sampled air was denitrified, typically by 70%

   ✧ ER-2 water vapor measurements showed that dehydration was very limited

✦ The "Liquid" scenario is clearly incompatible with SOLVE measurements: no solid PSCs form, and denitrification (though nonnegligible, since in the absence of solid particles liquid ones can grow large enough to sediment) is much less severe and extensive than observed.

✦ The "IceFrz" scenario can also be ruled out: PSC formation is too limited in both space and time, and denitrification is too weak and is invariably associated with dehydration.

✦ The NAD scenarios compare less favorably with the SOLVE criteria than the corresponding NAT ones, but they cannot be excluded.
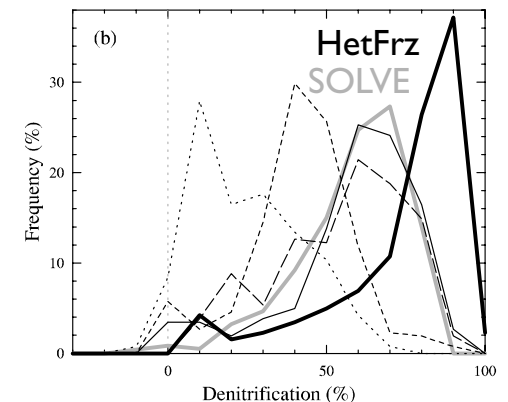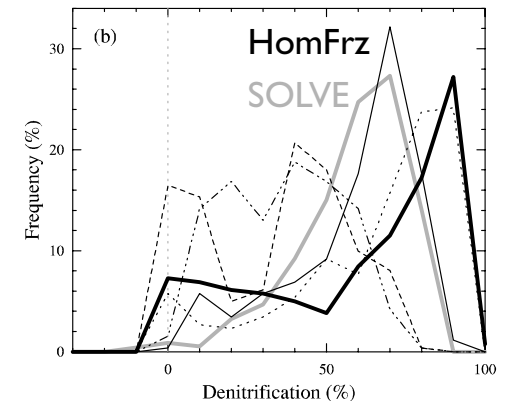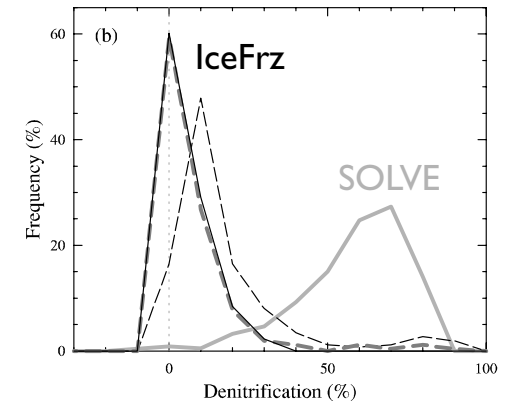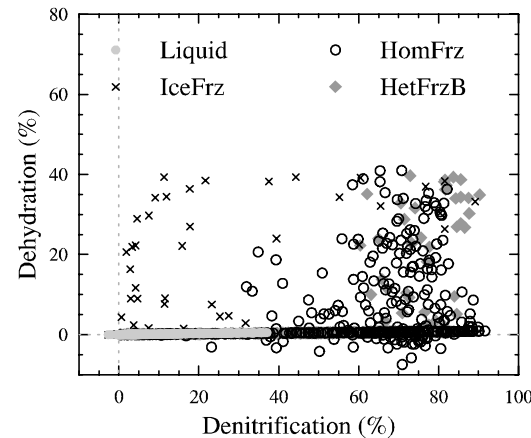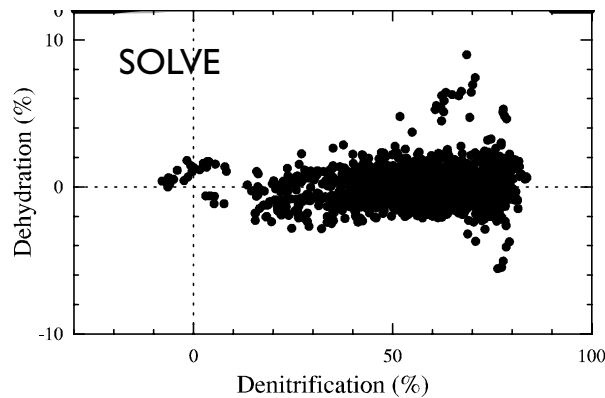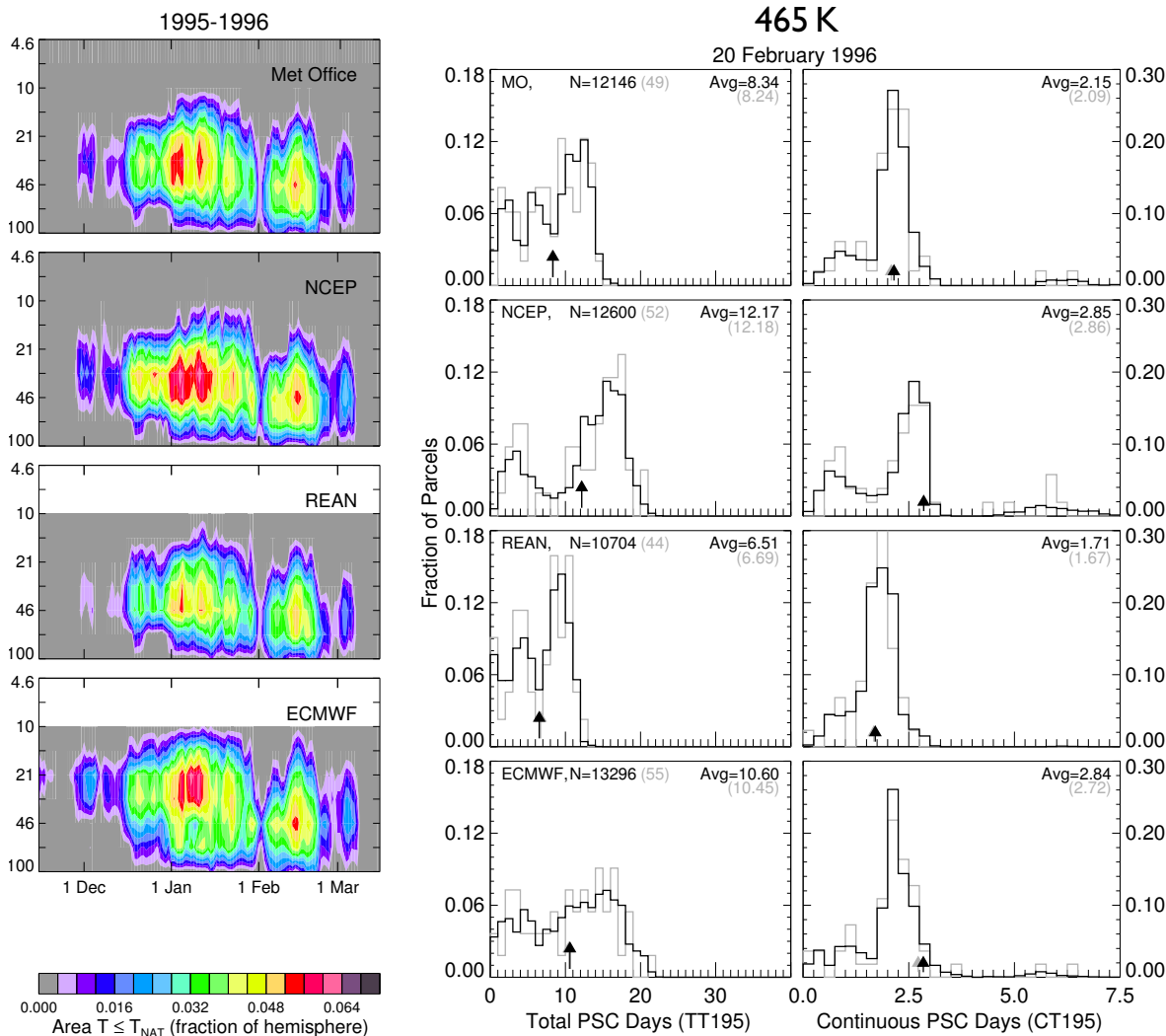
- ✦ Idealized homogeneous freezing ("HomFrz") produces early PSC formation and severe denitrification, but liquid PSCs are completely suppressed and denitrification is strongly coupled to dehydration.

- ✦ Heterogeneous freezing ("HetFrz") provides the best overall match to the SOLVE observations, leading to early and extensive PSC formation and strong denitrification that is largely decoupled from dehydration.

- ✦ But "HetFrz" still overpredicts dehydration: compared to the observations, more denitrified air parcels are also dehydrated (by a factor of 10), and dehydration levels are higher (up to 40% vs. under 10%).

- ✦ Reproducing key characteristics of PSCs observed during SOLVE requires a freezing mechanism that is active above the ice frost point.

- ✦ A heterogeneous freezing process has interesting implications:

  - ✧ Sedimentation of particles containing heterogeneous nuclei results in a steady decline in solid PSC concentrations over the winter.

  - ✧ Year-to-year variability in the number of nuclei is possible.

  - ✧ Long-term trends in nuclei concentrations could affect the extent and degree of denitrification.

- Davies et al. [JGR 107, 8322, doi:10.1029/ 2001JD000445, 2002] used the SLIMCAT model to quantify the effect of widespread denitrification on Arctic ozone loss during the SOLVE winter.

- Two different parameterizations of denitrification were employed: (1) sedimentation of ice particles containing co-condensed HNO_3 (the previous SLIMCAT default approach) and (2) sedimentation of large NAT particles (based on NAT rocks).

- When forced with UK Met Office analyses, the ice scheme produces denitrification, but its magnitude is underestimated and the NO_y profile is displaced upwards by 20 K compared to the observations.

- The NAT-based model simulations reproduce the observed vertical distribution of denitrification (and renitrification at lower altitudes), resulting in good agreement between measured and modelled NO_y.

- Model comparisons with ER-2 chlorine species are inconclusive, and agreement between different model runs and ozone is inconsistent with the NO_y and Cl_y comparisons.

- Widespread and severe denitrification during SOLVE is shown to contribute 20–30% of observed ozone loss.

- Modelled ozone loss is sensitive to the meteorological analyses used.

ER-2 NO_y
UKMO-ice   ECMWF-ice
UKMO-NAT   ECMWF-NAT
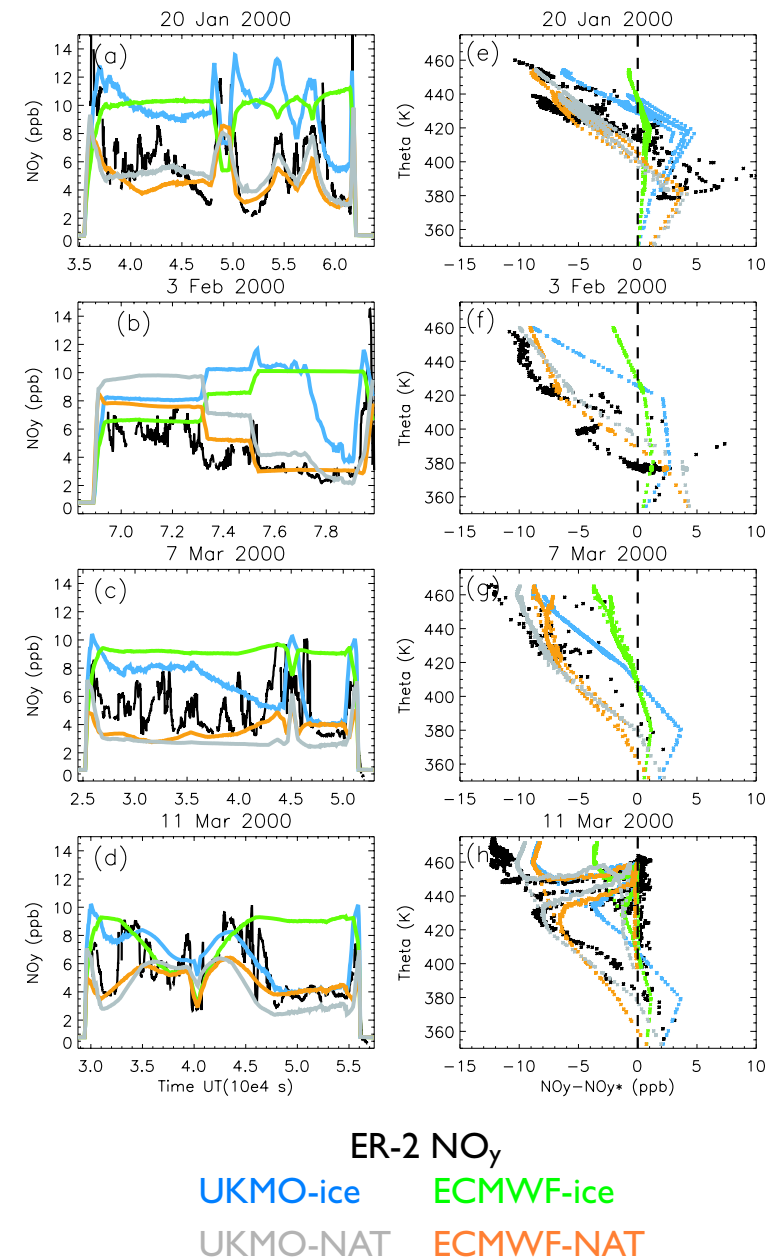
✦ Quantitative comparisons of commonly-used meteorological analyses reveal that the results of polar process studies can be strongly influenced by the choice of analyses used to obtain temperatures and temperature histories [Manney et al., JGR 108, 8328, doi:10.1029/2001JD001149, 2003]

✦ Manney et al. [2003] examined two Arctic winters characterized by temperatures that were comparably low but meteorological situations that were otherwise very different: 1995/1996 and 1999/2000.

1995-1996

465 K

20 February 1996

Met Office

NCEP

REAN

ECMWF

MO,   N=12146 (49)   Avg=8.34 (8.24)   Avg=2.15 (2.09)

NCEP,   N=12600 (52)   Avg=12.17 (12.18)   Avg=2.85 (2.86)

REAN,   N=10704 (44)   Avg=6.51 (6.69)   Avg=1.71 (1.67)

ECMWF, N=13296 (55)   Avg=10.60 (10.45)   Avg=2.84 (2.72)

Fraction of Parcels

Total PSC Days (TT195)

Continuous PSC Days (CT195)

Area T ≤ T_{NAT} (fraction of hemisphere)

✦ Minimum lower stratospheric temperatures typically differ by up to 5 K between analyses.

✦ Areas of low temperature vary up to ~25% between analyses during cold periods, with variations occasionally >50%.

✦ Temperature histories along trajectories exhibit substantial qualitative and quantitative differences in total time spent below 195 K (related to chlorine activation) and time continuously below 195 K (potential PSC lifetime, related to denitrification).

✦ Differences were generally larger in 1995/1996, when the cold region was often near the vortex edge, than in 1999/2000, when the cold region was near the vortex center.

✦ Manney et al. [JAS, submitted, 2004] performed a similar diagnostic comparison of commonly-used meteorological analyses for the 2002 Antarctic winter.

✦ Overall, wind and temperature fields from the various analyses were found to provide a more consistent picture of the large-scale dynamics of this winter than they did in the Arctic.

465 K, 26 May 2002
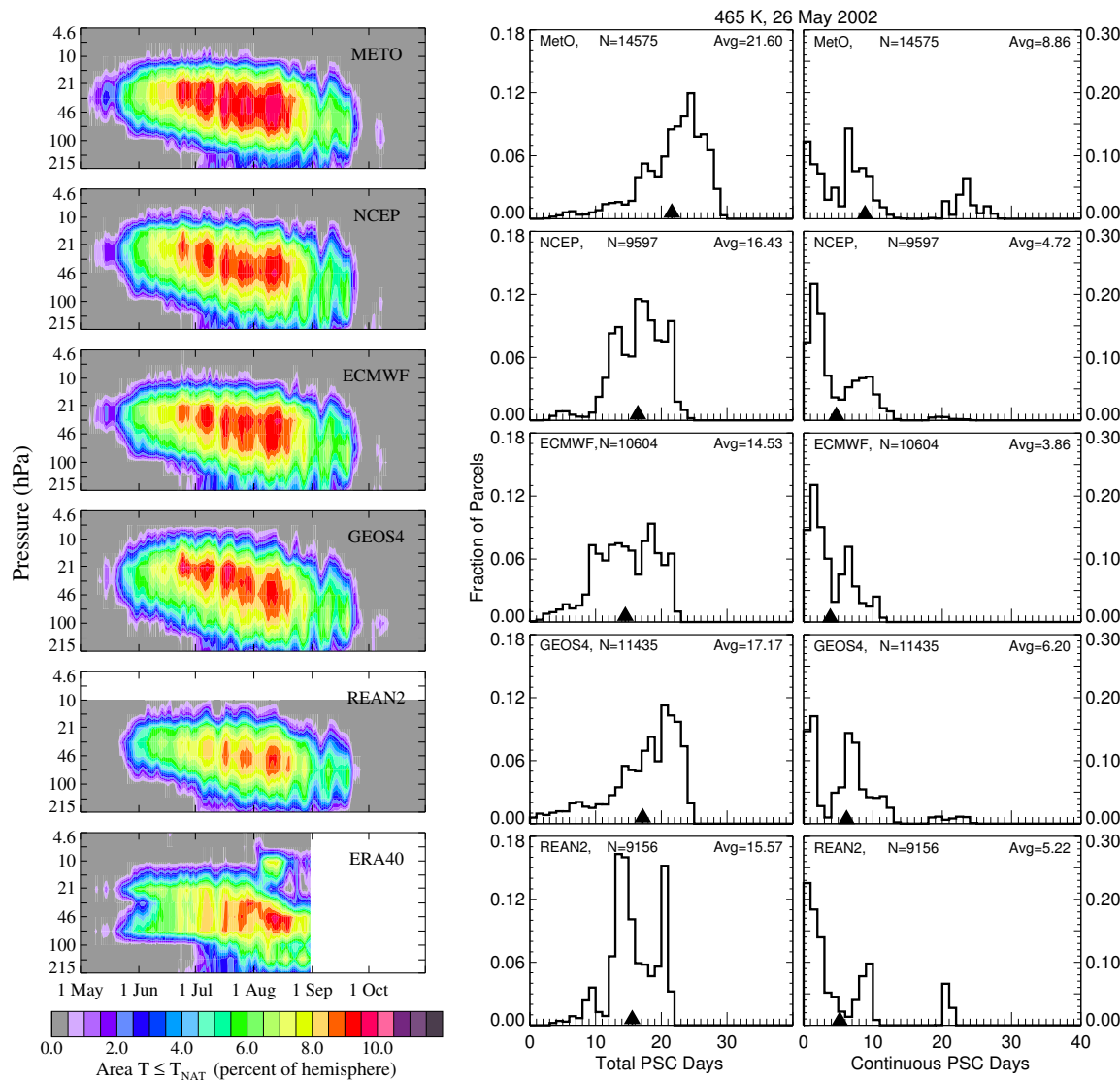
✦ Nevertheless, temperature differences between analyses can still have a significant effect on polar process studies in the Antarctic, particularly in the fall or early spring, when conditions are marginal for PSC formation.

✦ For example, the onset and duration of temperatures below PSC existence thresholds show significant variations in May.

✦ Modelling studies are beginning to explore this issue; SLIMCAT results from simulations forced by different analyses have been shown to display concomitant variations in chemical processing and ozone loss in both the Arctic [Davies et al., JGR 107, 8322, doi:10.1029/2001JD000445, 2002] and the Antarctic [Feng et al., JAS, in press, 2004].

A few critical questions related to polar processes that require further measurements and modelling to resolve are (not an exhaustive list!):

✦ Polar stratospheric clouds:
  ✧ What are the phase and composition of PSC particles, and what mechanisms for selective freezing or nucleation are at work in forming large, denitrifying particles?
  ✧ What are the dominant mechanisms governing PSC formation, and are they the same in both hemispheres?
  ✧ Will the spatial extent, duration, or frequency of occurrence of PSCs increase in the future in response to climate change and/or natural variability?

✦ Denitrification and dehydration:
  ✧ What are the main mechanisms for denitrification in the polar vortices, and are they the same in both hemispheres?
  ✧ Are denitrification and dehydration more typically coupled or independent processes, and does severe denitrification require temperatures near the ice frost point?
  ✧ Is widespread severe denitrification necessary for massive ozone loss?

✦ Chlorine Partitioning and chemical ozone loss:
  ✧ Is our present understanding of chlorine activation and recovery in the winter polar lower stratosphere quantitatively correct for different aerosol loadings?
  ✧ What loss mechanism is responsible for the rapid ozone depletion observed in mid-winter at high solar zenith angles?
  ✧ Does the chlorine budget in the stratosphere balance, or must additional but currently unknown chlorine species be accounted for?

A challenge for future research is to effectively marry the complementary strengths of satellite and aircraft measurements together with modelling in addressing these issues.

✦ NASA's EOS Aura satellite:

    ✧ is scheduled for launch on 19 June 2004

    ✧ will be in a near-polar, sun-synchronous, 705-km orbit with a 1:45 pm ascending equator-crossing time

    ✧ will be flying in formation in the same plane as (and only minutes apart from) PARASOL, CALIPSO, CloudSat, Aqua, and OCO (the "A-train")

    ✧ has a 6-year design lifetime

✦ The four Aura instruments, which will observe the same air mass within ~13 minutes, are :

    ✧ HIRDLS (High Resolution Dynamics Limb Sounder) — infrared limb-scanning radiometer measuring multiple azimuths behind Aura — T, O_3, H_2O, HNO_3, $ClONO_2$, CH_4, N_2O, NO_2, N_2O_5, CFC-11, CFC-12, and aerosol/cirrus heights, amounts, and particle characteristics

    ✧ MLS (Microwave Limb Sounder) — microwave limb-scanning spectrometers measuring ahead of Aura — see next page for measurements

    ✧ OMI (Ozone Monitoring Instrument) — nadir-viewing UV/VIS imaging spectrometer — O_3 profiles, column abundances of O_3, BrO, OClO, NO_2, HCHO, SO_2, and aerosol optical thickness

    ✧ TES (Tropospheric Emission Spectrometer) — infrared FTS measuring both the nadir and the limb behind Aura — T, O_3, H_2O, HNO_3, CH_4, CO, and NO_2, plus several 'special' products

✦ Overall science objectives are to address the following questions:

    ✧ Is the stratospheric ozone layer recovering?

    ✧ What are the sources and distributions of tropospheric pollutants?

    ✧ What are the roles of upper tropospheric water vapor, aerosols, and ozone in climate change?

✦ EOS MLS is an advanced-technology follow-on to UARS MLS, with greatly enhanced capability.

✦ Radiometers in 5 broad spectral bands centered near 118, 190, 240, 640, and 2500 GHz will measure key species in the upper troposphere, stratosphere, and mesosphere.