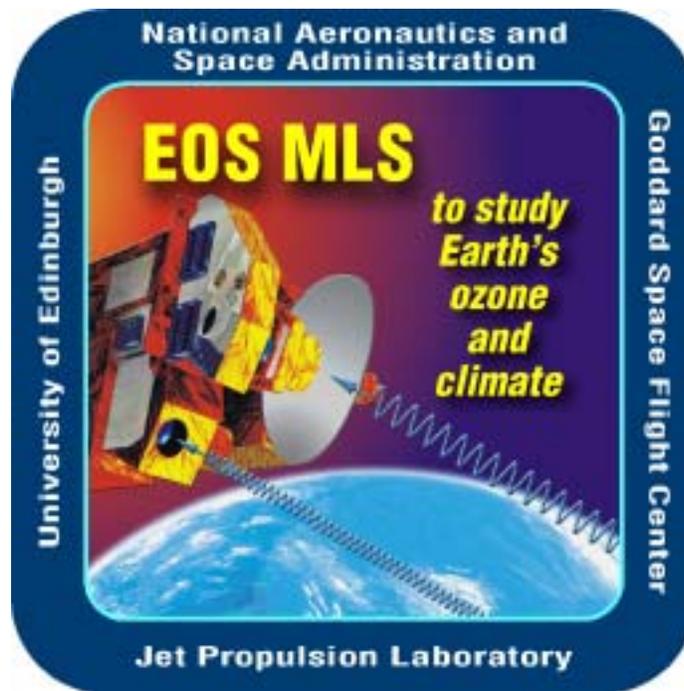
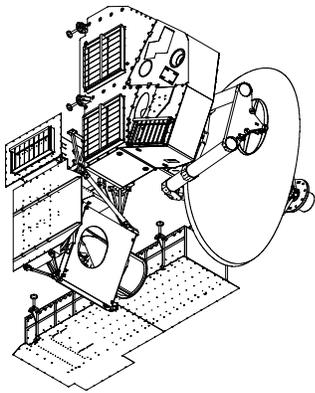


Earth Observing System (EOS)

Microwave Limb Sounder (MLS)

EOS MLS Science Objectives



Revision: Draft 0.2

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edited by J.W. Waters



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EOS MLS Science Objectives

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EOS MLS SCIENCE OBJECTIVES

release record

Note: the draft (blue font) entries will be deleted before release of Version 1.0.

Version	date released	comments
0.1 draft	8 Nov 2002	First complete draft circulated for review by co-authors.
0.2 draft	20 Dec 2002	Update of previous version to include comments from co-authors. Changes from Version 0.1 are in blue font. Pink font indicates some reference information that is still needed.
1.0	sometime later	Initial version

EOS MLS SCIENCE OBJECTIVES

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1 Introduction

This document describes the science objectives of the Earth Observing System Microwave Limb Sounder (EOS MLS) on EOS Aura to be launched in 2004. See Waters [1999] for an overview of the EOS MLS experiment. Companion instruments to MLS on Aura are the High Resolution Dynamics Limb Sounder (HIRDLS), the Ozone Monitoring Instrument (OMI), and the Tropospheric Emission Spectrometer (TES). See <http://eos-aura.gsfc.nasa.gov> for information on the Aura mission.

EOS MLS measures atmospheric composition, temperature, cloud ice, and pressure from observations of millimeter and submillimeter-wavelength thermal emission as the instrument field-of-view is scanned down through the atmospheric limb. The EOS MLS geophysical parameter measurements, with their expected range of useful altitudes are shown in Figure 1-1. The measurements cover 82° S to 82° N on each orbit, and all are made simultaneously and continuously. Precision estimates for all the measurements as a function on altitude are given by Filipiak [1999]. The standard vertical grid for output data products is 6 points per decade change in atmospheric pressure (~3 km vertical resolution), with plans to produce a few products (e.g., H₂O in the upper troposphere at a finer vertical resolution of 12 points per decade pressure (~1.5 km vertical resolution)).

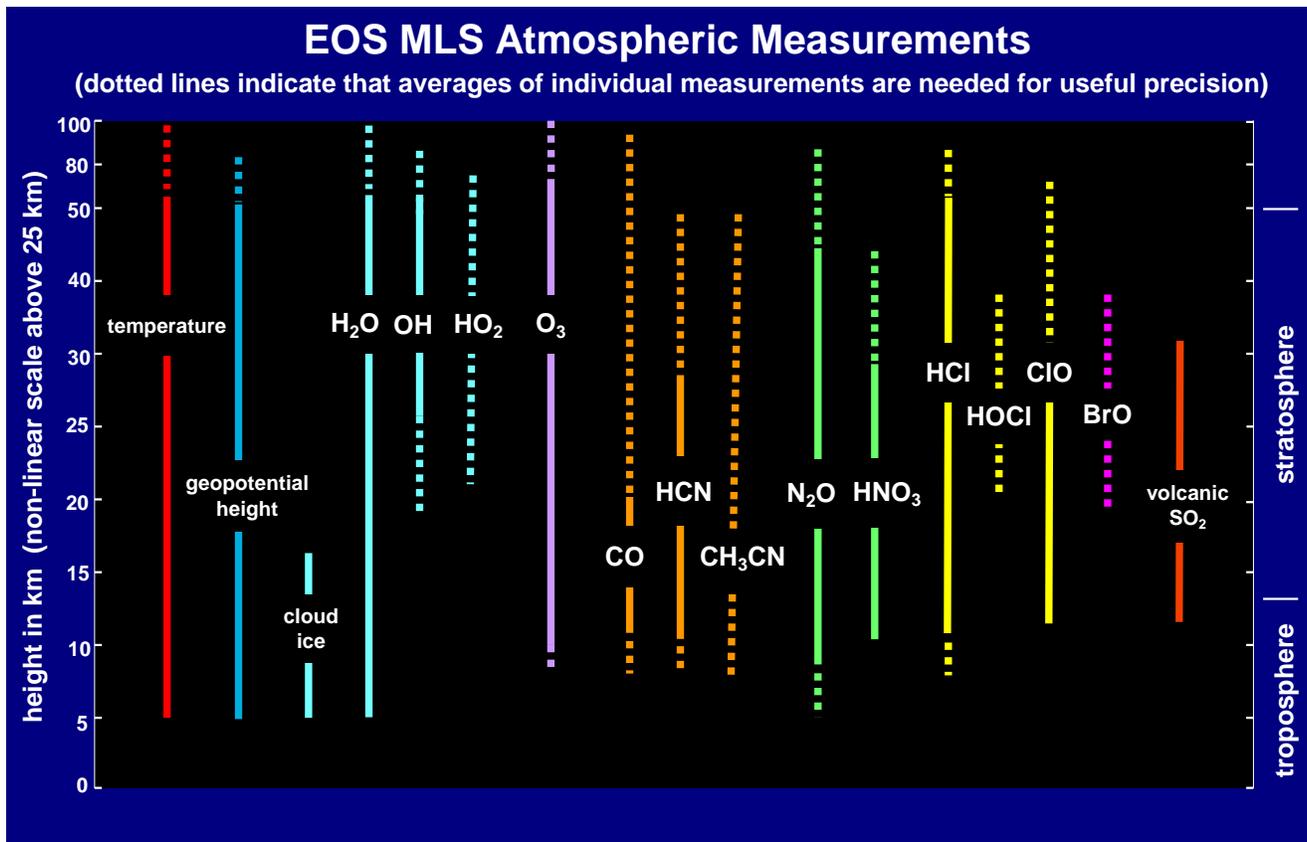


Figure 1-1. EOS MLS measurements. *[Need to do some minor updates on this figure.]*

MLS ‘looks forward’ from Aura, in the direction of orbital motion, and measurements are obtained along the suborbital path. The Aura orbit is sun-synchronous with ~705 km altitude, 98° inclination and ~1:45 pm ascending equator crossing time. Figure 1-2 shows the MLS measurement coverage for a 24 hour period, and Figure 1-3 shows the measurement geometry and the limb scan tangent point locus for the nominal operational scan. A complete limb scan including radiometric calibration, is performed 240 times per orbit under normal operations: every 1.5°, ~25 s, ~165 km along the orbit path. A vertical profile for each data product is produced every 1.5° along the orbit path, near the nominal location of the tangent point locus of each scan. The operational limb scan favors the upper troposphere and lower stratosphere - regions of emphasis for Aura - by spending more time at these lower altitudes, but also goes up to ~95 km on each scan. Figure 1-4 shows measurement local time as a function of latitude, and the variation over an annual cycle where the measurements (on both sides of the orbit) are in day and in night.

The EOS MLS instrument contains passive radiometers covering broad bands in five spectral regions centered at:

- 118 GHz (2.5 mm wavelength) for temperature and pressure;
- 190 GHz (1.6 mm wavelength) primarily for H₂O and HNO₃;
- 240 GHz (1.3 mm wavelength) primarily for O₃ and CO;
- 640 GHz (0.47 mm wavelength) primarily for HCl, ClO, N₂O, BrO, HO₂, HOCl;
- 2.5 THz (0.12 mm wavelength) primarily for OH.

The field-of-view vertical width decreases from 6.5 km at 118 GHz to 1.5 km at 640 GHz, and is 2.5 km at 2.5 THz; the horizontal width decreases from 13 km at 118 GHz to 3 km at 640 GHz, and is 2.5 km at 2.5 THz.

EOS MLS is a ‘second-generation’ MLS experiment, following the ‘first-generation’ instrument flown on NASA’s Upper Atmosphere Research Satellite (UARS) [e.g., Waters et al. 1999]. The major improvements in EOS MLS measurement capability over those of UARS MLS include (1) many more stratospheric species, (2) measurements to lower altitudes, (3) better global coverage with ±82° latitude coverage on each orbit, and (4) generally better precision and spatial resolution. The improvement in ozone measurement precision at 100 hPa, for example, is ~20×. See Waters [1999], Jarnot [1999], and Livesey and Wu [1999] for details on the EOS MLS instrument and data processing. An updated list of MLS scientific publications is maintained on the MLS web site (<http://mls.jpl.nasa.gov>).

The research areas that will benefit from the EOS MLS data can conveniently be grouped into the three categories

- Stratospheric Ozone,
- Tropospheric Ozone and Pollution,
- Climate Variability.

The following chapters describe specific science objectives for the use of MLS data in each of these categories. Key questions, as identified by NASA Earth Science Enterprise [2000], for each of these areas are stated at the beginning of each chapter and provide an overall focus for the objectives. We include both the objectives that will be addressed by the MLS team and its direct collaborators, as well as those that we expect will be addressed by the broader scientific community using MLS and other data.

Figure 1-2. EOS MLS measurement locations for a 24 hour period. Each cross gives the location of the tangent point for an individual limb scan. The continuous line is the suborbital track, which is slightly displaced from the tangent points because of Earth's rotation during the time in which the satellite moves forward to the tangent point latitude. The ascending portions of the orbit are those with the southeast-northwest tilt. Daily coverage at high latitudes in the Southern Hemisphere is analogous to that of the Northern Hemisphere shown here.

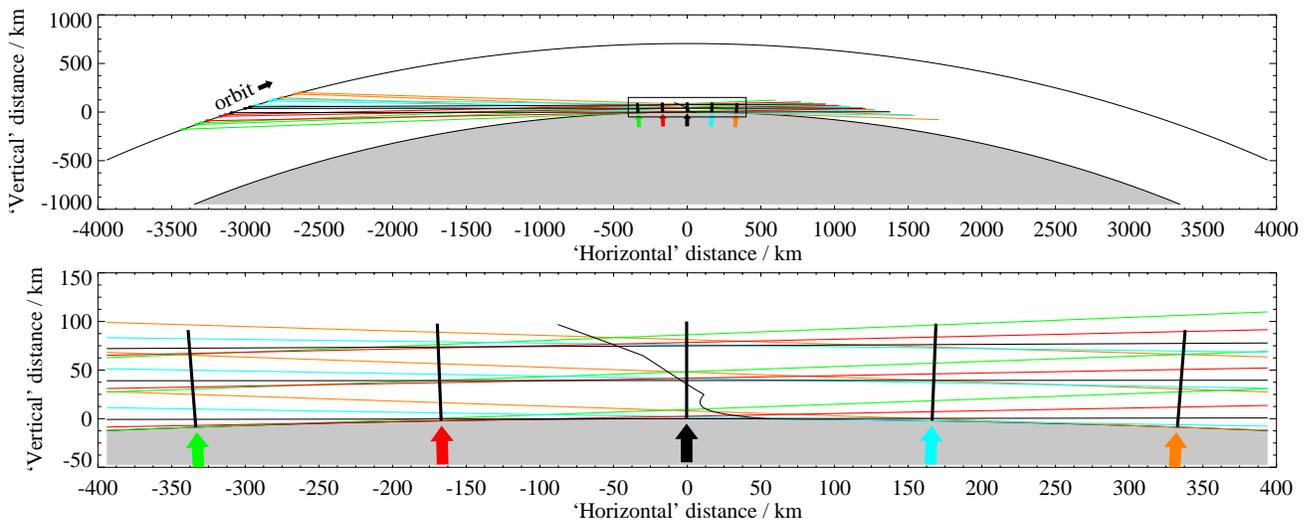
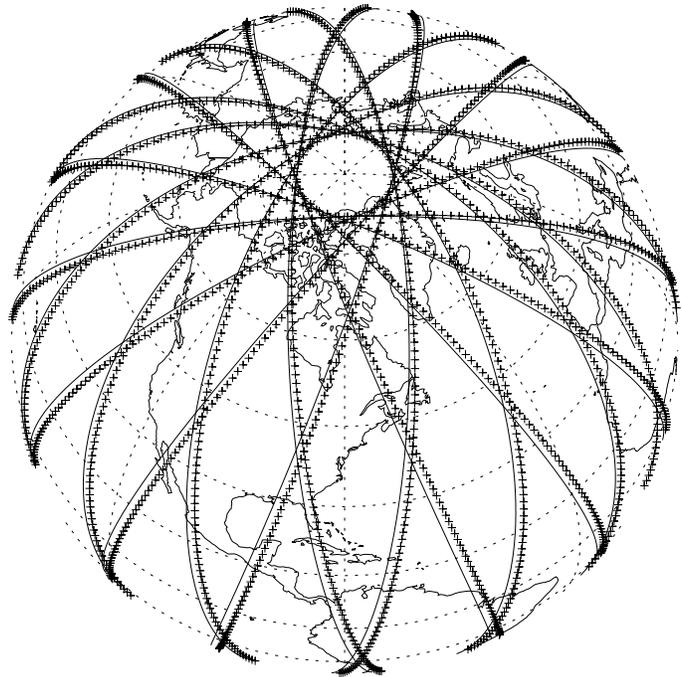


Figure 1-3. MLS viewing geometry and limb scan tangent point locus. The top panel shows the viewing geometry of EOS MLS, which looks forward from the Aura platform. The bottom panel is an expansion of the boxed region in the upper panel. The nearly-vertical lines, above the upward-pointing arrows, show the locations for five successive retrieved vertical profiles. The nearly horizontal lines show the limb ray paths for a few scan positions. The somewhat irregular line going upward and to the left between about -100 and 50 km "horizontal" distance is the locus of the tangent point (including refractive effects) for the MLS nominal operational limb scan. Changes in its slope are due to changes in the antenna scan rate; more time is spent observing the higher-priority tropospheric and lower stratospheric regions. The limb scan is performed in an upward direction, and the 'backward' slope of the tangent point locus is due to tangent points at higher altitudes being nearer the satellite and the faster scan rate that compensates for the 'forward' orbital motion.

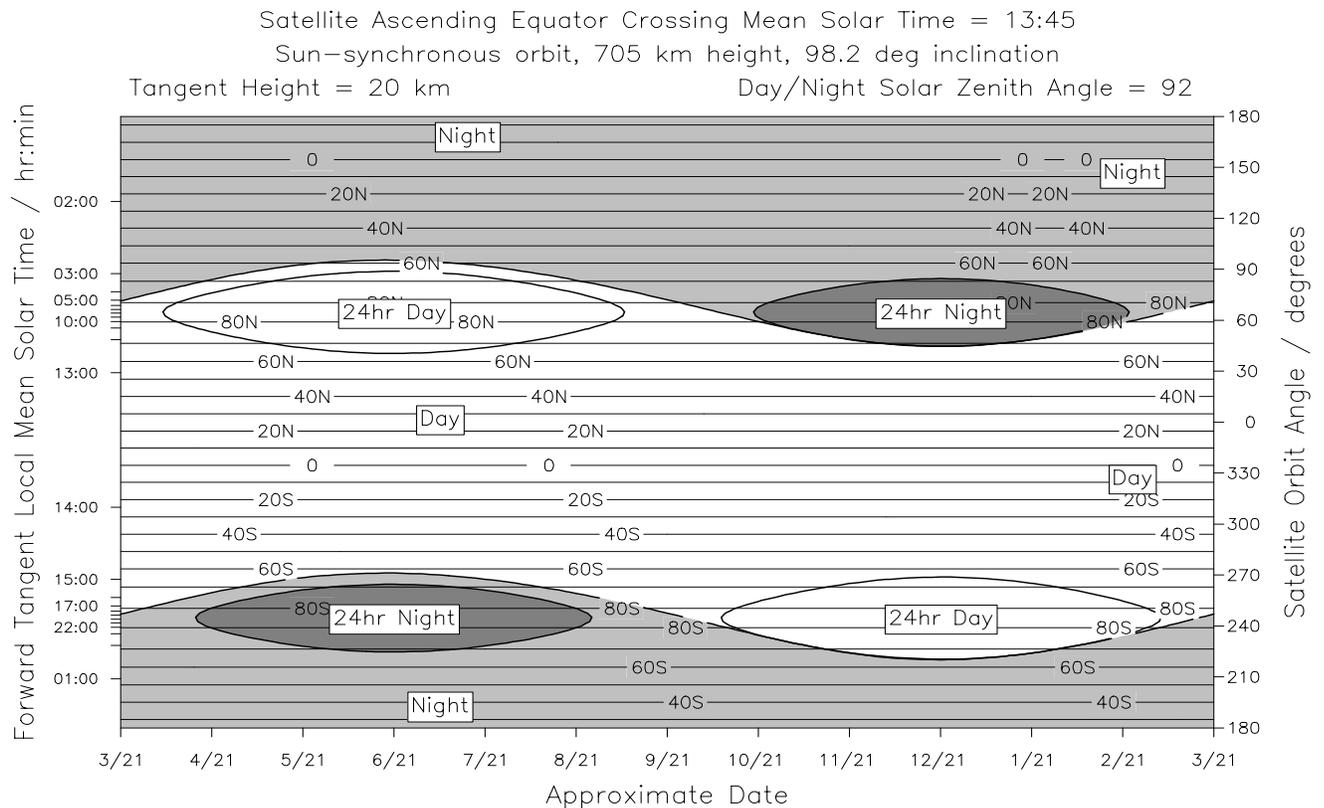


Figure 1-4. Local time of MLS measurements as a function of latitude and orbit angle, and variation - over an annual cycle - of the latitude range where MLS measurements are in day and in night. A way to understand this figure is through the following steps. (1) Start with the “Satellite Orbit Angle” along the right vertical axis. This is just the angle which labels the satellite location along the orbit, with zero set arbitrarily at the ascending (north-going) crossing of the equator. (2) Now consider the horizontal lines. These show the latitude (in 10 degree increments) of an MLS measurement for a given satellite location. The MLS measurement latitude is offset from the satellite latitude because MLS looks forward from Aura and makes measurements ~25 degrees of orbit circle ahead of the satellite. (3) Next consider the “Forward Tangent Local Mean Solar Time” along the left vertical axis. This gives the local times (for both the ascending and descending sides of the orbit) at which the MLS measurement for a given latitude (indicated by the horizontal lines) occurs. The ticks are at hourly intervals of local time. Local time for measurements at a given latitude does *not* change as a function of day-of-year because the Aura orbit is sun-synchronous. (4) Now consider the regions indicated as day (no shading) and night (gray-shaded). The boundary between these corresponds to a local solar zenith angle (sza) of 92°, the approximate sza for sunrise and sunset at an altitude near the tropopause. The local time for the day/night boundary is a function of day-of-year (e.g., the sun sets/rises earlier/later in winter than in summer). (5) Finally, the regions labeled “24 hr Day” and “24 hr Night” encompass latitudes/times where the atmosphere experiences a 24 hour “day” or “night” - irrespective of the MLS measurements.

2 Stratospheric Ozone

The following key questions [NASA, 2000] are directly related to stratospheric ozone.

- *How is stratospheric ozone changing, as the abundance of ozone-destroying chemicals decreases and new substitutes increase?*
- *How do stratospheric trace constituents respond to change in climate and atmospheric composition?*
- *How well can future atmospheric chemical impacts on ozone and climate be predicted?*

We divide discussion of the MLS science objectives related to stratospheric ozone, and the above key questions, into the following subsections:

- 2.1 Global stratospheric ozone:** objectives related, for the most part, to issues having to do with stratospheric ozone on a global scale;
- 2.2 Polar winter processes:** objectives related to issues having to do with polar winter processes and their local effect on stratospheric ozone;
- 2.3 Stratospheric H₂O:** objectives related to understanding aspects of H₂O that can affect stratospheric ozone on a global scale;
- 2.4 Volcanic effects on stratospheric ozone:** objectives to be pursued in the event of a volcanic eruption during the Aura mission that could affect stratospheric ozone.

2.1 Global Stratospheric Ozone

We further divide discussion of objectives related to global stratospheric ozone into the following subsections:

- 2.1.1 Stratospheric ozone:** objectives related to stratospheric ozone itself;
- 2.1.2 Chlorine chemistry:** objectives related to stratospheric chlorine chemistry and its effects on ozone;
- 2.1.3 Hydrogen chemistry:** objectives related to stratospheric hydrogen chemistry and its effects on ozone;
- 2.1.4 Bromine chemistry:** objectives related to stratospheric bromine chemistry and its effects on ozone;
- 2.1.5 Dynamics and transport:** objectives related to stratospheric dynamics and transport and their effects on ozone;
- 2.1.6 Mesospheric topics:** objectives related to improving our understanding of the mesosphere that may benefit our understanding of the stratosphere and, thus, of stratospheric ozone.

2.1.1 Stratospheric Ozone

An overarching question regarding global stratospheric ozone is whether its abundance will recover as expected in the next few decades following international regulations now in effect

on ozone depleting substances. Stratospheric total chlorine will be at or just over its peak during the Aura mission, and stratospheric total bromine is expected to still be increasing - but more slowly than previously [World Meteorological Organization, 2002]. We thus might expect some abatement of stratospheric ozone depletion during Aura's lifetime, and an eventual global recovery which may not be definitively detectable until later [Reinsel et al., 2002; World Meteorological Organization, 2002]. Climate change could possibly delay ozone recovery - both through stratospheric cooling that exacerbates heterogeneous processes leading to ozone destruction, and possibly by causing changes in transport through the tropical tropopause layer that could affect the amount of H₂O (and perhaps other substances) in the stratosphere. Projected increases in stratospheric H₂O may delay the recovery of stratospheric ozone by 10-30 years [Dvortsov and Solomon, 2001; Shindell, 2001]; [one may be able to separate the upper stratospheric halogen versus greenhouse gas effects on ozone recovery by analyzing the latitude dependence of ozone trends \[Shindel and Grewe, 2002\]](#).

Following are examples of some specific objectives and investigations in this area that are expected to be performed with the MLS and related data.

- **Producing an accurate record of global three-dimensional ozone changes during the Aura mission.** The MLS ozone data will be compared and combined with that from HIRDLS, OMI, and TES Aura instruments and other datasets (e.g., UARS, SAGE, ENVISAT, and ground-based data) to produce an accurate record of stratospheric ozone changes during the Aura mission. Validation and science objectives are intimately tied in such analyses, but providing continuity with observations from previous datasets is clearly needed for an accurate assessment of changes. Time series analyses that include various temporal modes (e.g., annual, semi-annual, QBO) will be used to extract the underlying trends, accounting for the added complication that a recovery (or pre-recovery) period is not characterized by a simple linear trend. With significant improvements in precision over that of UARS MLS, the EOS MLS measurements of ozone profiles down into the upper troposphere (with negligible or small interference from aerosols and thin ice clouds) are expected to be especially valuable for quantifying changes in the lower stratosphere.
- **Understanding global changes in upper stratospheric ozone.** The upper stratosphere may experience an abatement and reversal in ozone declines before the lower stratosphere, partly because of the [smaller effects of dynamics at these altitudes where chemical timescales are much shorter](#) [World Meteorological Organization, 2002]. Climate change, with associated temperature decreases in the upper stratosphere, may hasten ozone recovery in this region. MLS profiles of O₃ and ClO (the main radical responsible for upper stratospheric O₃ depletion), will be used to critically evaluate this issue. The MLS O₃ and ClO data will also be used to further investigate interhemispheric asymmetries in the ozone trend at 1 hPa seen in SBUV data that could not be explained by UARS MLS ClO data [Considine et al., 1998].
- **Understanding global changes in lower stratospheric ozone.** MLS data on H₂O, HO_x, BrO, HOCl, and temperature (with other Aura measurements of NO_x) will be used to further constrain and test models of ozone change in this region. Tracers such as N₂O will be used to facilitate the separation of dynamical and chemical effects.
- **Understanding variations in the source gases H₂O, N₂O and CH₄ that can affect stratospheric ozone.** MLS [and HIRDLS](#) measurements of H₂O and N₂O, and HIRDLS measurements of CH₄, will be used to constrain models that predict how variations in these long-lived source gases might affect stratospheric ozone. While the expected changes for

N₂O are not very large over a five year period (about 1%), much larger and unexpected changes in stratospheric source gas abundances occurred in the 1990s [WMO, 1998] for H₂O and CH₄. Continued monitoring and analyses of the H₂O variations (from MLS and other Aura instruments) will be pursued, in conjunction with observed changes in stratospheric CH₄, a stratospheric source for H₂O.

- **Confirming that the upper stratospheric “model ozone deficit problem” has been resolved.** While it would seem that the long-standing stratospheric “model ozone deficit problem” has been largely resolved [WMO, 1998], partly from changes in the branching ratio for converting ClO to HCl (via reaction with OH), and partly from improvements in modeled radiation fields, more stringent and extensive tests of the upper stratospheric ozone budget can be performed with the Aura data than has previously been possible. The suite of essentially co-located measurements from MLS and the other Aura instruments will be used to provide unique global constraints for such investigations (from box models constrained by long-lived tracers to 3-D chemical-transport models). In addition to ozone, the key Aura measurements are zonal mean radical abundances of OH, HO₂, ClO, and BrO from MLS and NO_x from TES and HIRDLS.

2.1.2 Chlorine Chemistry

Understanding and monitoring stratospheric chlorine chemistry is very important during the Aura time period when total stratospheric chlorine will be 6 to 7 times the ‘natural’ abundance of chlorine in the stratosphere [WMO, 1998, 2002]. Some specific objectives and investigations related to global stratospheric chlorine chemistry are given below. (Objectives and investigations concerned with polar winter processes are covered later in section 2.2. Issues in chlorine chemistry that could arise from volcanic effects are also covered later in section 2.4.)

- **Understanding total stratospheric chlorine variations and trends.** The chlorine loading of the stratosphere is now at or near the peak, with HCl and ClONO₂ column abundances having plateaued in recent years [World Meteorological Organization, 2002]. In the absence of other factors, the decline in stratospheric chlorine is expected to lead to an increase in global ozone [e.g., WMO, 2002]. Previous measurements and analyses have shown that the upper stratospheric HCl abundance yields the total chlorine loading of the stratosphere, tracking the measured total tropospheric chlorine loading with a delay of about 6 years [e.g., Anderson et al., 2000]; declines in tropospheric chlorine abundances since about 1994 have already been demonstrated [e.g., Montzka et al., 1996]. It is possible, however, that our understanding of stratospheric chlorine loading is incomplete: Waugh et al. [2001] found an inconsistency in the timing and magnitude of the peak in total chlorine abundance at 55 km deduced from UARS Halogen Occultation Experiment (HALOE) HCl observations and the amount inferred from surface observations, perhaps indicating that stratospheric chlorine is not decreasing as predicted. A thorough understanding of the changes in stratospheric chlorine concentrations is fundamental to determining whether ozone is recovering ‘as expected’. Spatial and temporal variations in HCl from MLS will be compared to the few other estimates that may exist during at least part of the Aura timeframe (e.g., satellite data from ACE and HALOE; campaign data from balloon flights at somewhat lower altitudes). In particular, high priority will be given to comparing the MLS and HALOE measurements of upper stratospheric HCl, so that the MLS record can be connected to the existing long-term HALOE record. The measured changes in total chlorine will be used for possibly determining/confirming a causal

relationship between expected decreases in chlorine and changes in upper stratospheric ozone.

- **Understanding stratospheric reactive chlorine variations and trends.** ClO is the dominant form of reactive chlorine in the stratosphere, and provides a measure of the rate at which chlorine destroys stratospheric ozone. Unexpected increases in the amount of ClO can provide early warning of unexpected threats to stratospheric ozone. The EOS MLS measurements of ClO will be analyzed for determining and monitoring the global variation in stratospheric reactive chlorine during the Aura time period. As found by Froidevaux et al. [2000], based on UARS MLS ClO data, variations in the total amount of global reactive chlorine can be dominated by factors other than the amount of total chlorine in the stratosphere. Unusual changes in CH₄, for example, have been shown to have a significant impact on middle and upper stratospheric ClO abundances [Siskind et al., 1998; Froidevaux et al., 2000]. HIRDLS CH₄ will thus be extremely valuable for helping understand observed variations in upper stratospheric ClO. As a major improvement over UARS MLS, EOS will measure both ClO and HCl. Monitoring the ClO/HCl ratio - which can be measured very accurately since the MLS ClO and HCl measurements are commonly-calibrated, co-located, and made simultaneously - will possibly provide a better 'warning' of unexpected changes in chlorine chemistry than could be obtained from either ClO or HCl alone. Referencing such changes with respect to tracers N₂O (measured by both MLS and HIRDLS) and CH₄ (measured by HIRDLS) will be used to help separate chemical and dynamical effects.
- **Testing our understanding of chlorine partitioning in the middle and upper stratosphere.** The upper stratospheric chlorine budget and partitioning seem to be quite well understood, based on the most recent comparisons between models and observations [World Meteorological Organization, 1998]. However, Aura gives a suite of global measurements that will be used for a more stringent and extensive test of this partitioning than has previously been possible. The key measurements unique to MLS include ClO, HCl, OH, and HO₂; those unique to HIRDLS include ClONO₂ and CH₄; and both MLS and HIRDLS measure the key parameters temperature, O₃, N₂O and H₂O. There are only some minor missing components (e.g., H₂CO and H₂) from this suite [e.g., Siskind et al., 1998] that will require model values. These measurements will be used for continued testing of our understanding of the linkage between ClO and HCl, and potential changes in partitioning between free chlorine and its reservoirs.
- **Testing our understanding of chlorine partitioning in the lower stratosphere.** Chlorine species in the lower stratosphere have been measured by balloons, aircraft, and UARS MLS and used for tests of the chlorine partitioning. Further global testing of the chlorine partitioning will be done using MLS measurements of ClO and HCl, along with ClONO₂ from HIRDLS, and NO_x from HIRDLS and TES; such testing will be helped by observed tracer (N₂O, CH₄) distributions and (constrained) model comparisons. Further discussion related to objectives for testing our understanding of lower stratospheric chlorine partitioning are given in section 2.2 in relation to polar winter processes and in section 2.4 in relation to volcanic effects.
- **Bounding the importance of cirrus cloud effects on stratospheric chlorine chemistry and ozone loss.** Heterogeneous chemistry effects of cirrus in the near-tropopause region on chlorine and ozone have been a topic of recent interest [Borrmann et al., 1997; Solomon et al., 1997]. Models of this process fall short of explaining the pronounced

minima in upper tropospheric O₃ that have been detected in the presence of ice clouds by ground-based midlatitude lidar [Reichardt et al., 1996]. Based on ER-2 measurements near the tropopause, showing a consistent absence of enhanced (larger than about 1 pptv) ClO in the midlatitude lowermost stratosphere, Smith et al. [2001] conclude that the cirrus-driven mechanism for chlorine activation and in situ ozone loss is unimportant at midlatitudes (Lelieveld et al. [1999], however, conclude that it may be important in the middle and high-latitude lowermost stratosphere). The issue of heterogeneous chemistry on cirrus will become more important if a global increase in cirrus occurs as part of climate change. Improvements over UARS MLS in ClO sensitivity, altitude coverage and spatial resolution will make investigation of chlorine activation by cirrus more feasible than was possible with UARS MLS ClO - but precision and resolution may still limit the results that can be obtained (since averaging will be required). Nevertheless, analysis of lower stratospheric ClO from EOS MLS will be performed with the objective of confirming that this mechanism is indeed unimportant on a global and continuous basis, and setting bounds on its potential effect.

2.1.3 Hydrogen Chemistry

EOS MLS will provide the the first global and continuous measurements of OH and HO₂ over the full region of 18-95 km, and for a multiyear period. These measurements will substantially improve our understanding of stratospheric hydrogen chemistry. (See Brasseur and Solomon [1986], Pickett and Petersen [1996] and Brasseur et al. [1999] for general information on stratospheric hydrogen chemistry.) This chemistry dominates ozone destruction in the altitude regions of ~20-25 km and above ~45 km [e.g., Osterman et al., 1997]. Its understanding is thus essential for adequate understanding of how stratospheric ozone loss might change in response to climate and composition changes. Our current understanding of hydrogen chemistry in the upper stratosphere is in question due to the Middle Atmosphere High Resolution Spectrograph Investigation (MAHRSI) OH observations that do not appear to be consistent with current theory [Conway et al., 2000].

The principal source gases and reservoirs for stratospheric hydrogen are H₂O and CH₄, and the oxidation of CH₄ is a major source of H₂O in the upper stratosphere. H₂O will be measured by MLS and HIRDLS, and CH₄ by HIRDLS, so Aura will measure the suite of primary source gases for stratospheric hydrogen that will provide valuable boundary conditions for chemical models.

Following are some objectives and investigations related to stratospheric hydrogen chemistry that will be addressed with MLS data.

- **Testing our understanding of hydrogen chemistry in the mesosphere and upper stratosphere.**

The major source of stratospheric HO_x is reaction of H₂O with electronically-excited oxygen atoms O(¹D) generated by O₃ photolysis at wavelengths shorter than 320 nm:



O(¹D) also produces OH to a lesser extent by reaction with CH₄:



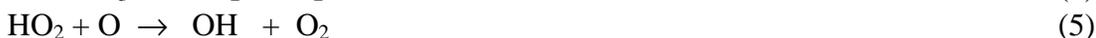
CH₃ can be oxidized to give HO₂ and CH₂O (formaldehyde), which is further oxidized to CO₂ with the formation of more HO_x radicals. Because formaldehyde reactions are relatively slow, its oxidation is not thought to be an important source of HO_x.

In the mesosphere, OH and H are also directly produced by photolysis of H₂O at wavelengths shorter than 180 nm.

The major termination reaction for HO_x is



There are also a number of reactions that convert between the HO_x radicals, for example the ozone catalytic destruction reactions:



H also converts to HO₂ by the 3-body reaction:



that, surprisingly (even in the mesosphere), is the significant way that H is converted to HO₂. Throughout most of the upper atmosphere, over 85% of the H reacts using reaction (8) with the balance coming mostly from reaction (7). In the daytime above 35 km, OH and HO₂ partitioning is dominated by reactions (5) and (6) and is independent of O atom concentration. Reactions (6)-(8) constitute a way to obtain H concentration once O atom concentration is inferred from photolysis of O₃ and O₂. Nonetheless, the H concentration is less than 1% of total HO_x below 50 km, rising to 10% at 60 km and 50% at 70 km.

In the mesosphere and upper stratosphere, simultaneous observations of OH and HO₂ along with water and O₃ provide strong tests of HO_x chemistry and ozone destruction processes. Simultaneous MLS observations of both OH and HO₂ will allow us to look at this region of the atmosphere with much more discrimination than has previously been possible. Comparisons of the MLS measurements with predicted OH and HO₂ are best done on a monthly basis since HO₂ will only be available as monthly zonal means. On the other hand, predicted HO_x concentrations are non-linear functions of water and ozone concentration. An appropriate way to perform the comparison is to calculate OH and HO₂ on a daily basis using maps of the source species. Then a monthly zonal average prediction can be compared with the monthly zonal average observations. In most parts of the atmosphere, photochemical equilibrium calculations can be used because the HO_x reactions are very fast. In the mesosphere, where the reactions are slower, a model will be used that integrates the time-dependent rate equations.

Comparisons of OH and HO₂ concentrations versus altitude will help to understand the mesospheric HO_x dilemma seen in the MAHRSI measurements showing that there is less OH above 50 km than predicted by model calculations, but more OH below 50 km than predicted. Changes in the rates of reactions (3), (5), and (6) cannot resolve the differences. Increasing the rate of reaction (3) will make total HO_x decrease with corresponding decrease in both OH and HO₂. On the other hand, modification of reactions (5) and (6) will make complementary changes in OH and HO₂. The MLS simultaneous measurements of OH and HO₂, made over all latitudes and seasons, should help resolve this dilemma and

provide insights as to whether modifications are needed in the standard chemistry given above.

- **Testing our understanding of hydrogen-nitrogen chemistry couplings in the lower stratosphere.**

In the lower stratosphere, the reactions mentioned under the previous bullet are still significant, but at these altitudes HNO₃ (nitric acid) also participates in reactions that alter the concentrations of HO_x, for example:



Reactions (9)–(11) can be combined under conditions of photochemical equilibrium to give

$$[\text{OH}] = J_9 [\text{HNO}_3] / \{k_{10} [\text{NO}_2][\text{M}] - k_{11} [\text{HNO}_3]\} \quad (12)$$

It is clear from expression (12) that increasing HNO₃ will increase OH by reaction (12). On the other hand, reaction (11) has the net result that HNO₃ is a sink for HO_x rather than a source. Expression (12) has been used [Pyle et al., 1983] to infer OH from satellite measurements of HNO₃ and NO₂. However, the time constant for OH loss via reaction (10) is 1 hour above 35 km so the assumption of photochemical equilibrium may not be appropriate except at lower altitudes.

With the improved understanding of HO_x and oxygen chemistry at higher latitudes, nitrogen chemistry can be studied with new completeness. The simultaneous MLS observations of OH, HO₂ and HNO₃ and HIRDLS observations of NO₂ (and HNO₃) will allow tests of expression (12) or related calculations that explicitly integrate the rate equations (and thereby do not require reactions to be in photochemical equilibrium). As with the studies at higher altitudes, comparison will be done on a monthly-averaged basis by appropriately averaging predictions obtained from daily fields of water, ozone, nitric acid, methane, and NO₂.

- **Testing our understanding of hydrogen-halogen chemistry couplings in the lower stratosphere.**

Knowledge of HO_x concentrations will also help testing and understanding of stratospheric halogen chemistry. OH is the primary way that the reservoir molecule HCl is converted into active chlorine. OH also reacts with ClO: the ~6% branching of OH + ClO to form HCl + O₂ [Lipson et al., 1997] significantly reduces model predictions of upper stratospheric ClO abundances, bringing them in close agreement with UARS MLS measurements, and reduces the long-standing model upper stratospheric 'O₃ deficit' problem [e.g., Khosravi et al., 1998]. HO₂ reacts with Cl to produce two products. One pathway forms OH and ClO, therefore influencing the active chlorine partitioning. The other pathway forms HCl and O₂ to return active chlorine to the reservoir.

OH is also the primary way that the reservoir molecule HBr is converted into active bromine. HO₂ also reacts with Br to produce HBr and O₂. In all these cases, the MLS measurements of OH and HO₂ will add constraints to broader studies of halogen chemistry budgets.

- **Testing our understanding of the stratospheric hydrogen budget**

Analyses of HALOE measurements of $\text{H}_2\text{O} + 2 \text{CH}_4$ concentration have provided insight into the stratospheric hydrogen budget [e.g., Harries et al. 1996a,b]. This is because CH_4 is oxidized to H_2O through reaction with $\text{O}(^1\text{D})$ and OH , eventually producing 2 H_2O molecules for each CH_4 molecule. Above around 60 km nearly all of the CH_4 is oxidized [e.g., Park et al. 1996]. In addition to H_2O and CH_4 , the significant hydrogen reservoir species is H_2 . Models [e.g., Garcia and Solomon, 1994] predict the H_2 concentration to be ~0.5 ppmv throughout most of the middle and upper stratosphere (but with abundances up to ~1.5 ppmv in the polar night), increasing to ~3-6 ppmv (latitude and seasonal dependent) near the mesopause, and generally in agreement with the values inferred by Harries et al. [1996b]. Limited direct observational evidence [quoted by Harries et al., 1996b, with references therein] also indicate the stratospheric H_2 mixing ratio is about 0.5 ppmv. By comparison, the $\text{H}_2\text{O} + 2 \text{CH}_4$ mixing ratio is typically 5-7 ppmv below ~60 km, decreasing to less than 1-2 ppmv (depending upon latitude and season) at ~80 km [Harries et al., 1996b]. Deficits of $[\text{H}_2\text{O}]$ above 60 km relative to $[\text{H}_2\text{O} + 2 \text{CH}_4]$ measured at lower altitudes are indicators of H_2 . The MLS and HIRDLS H_2O measurements, and the HIRDLS CH_4 measurements, will provide a data set extending that of HALOE to include measurements at all time of day and night (including in the polar night) for improving our understanding of the stratospheric and mesospheric hydrogen budget. Since H_2 also reacts with $\text{O}(^1\text{D})$ and OH , the inferred H_2 adds additional constraints on mesospheric HO_x chemistry.

2.1.4 Bromine Chemistry

Stratospheric bromine chemistry and its role in stratospheric ozone loss is a topic of increasing scientific interest. The abundance of bromine in the stratosphere is still increasing (albeit more slowly than a few years ago) [WMO, 2002], and bromine is 40-100 times more effective (depending upon location) in destroying ozone than chlorine [Brasseur et al., 1999]. However, the total amount of bromine in the atmosphere is much less than that of chlorine (~20 pptv compared to ~4 ppbv), so currently it is a less significant contributor to ozone depletion than chlorine, accounting for between 30-50% of the observed loss (depending on latitude, altitude and season) [e.g., World Meteorological Organization, 1999]. The processes involved in gas phase depletion of ozone by bromine are similar to those of chlorine. A major difference between gas-phase bromine and chlorine chemistry, however, is that the relative amount of bromine in the reactive BrO form (about 50% at midlatitudes between altitudes of ~15-35 km) is much larger than the relative amount chlorine in ClO form [e.g., Lary, 1996]. The larger relative abundance of BrO is due to the fact that the reactions of Br with CH_4 and H_2 to form HBr are endothermic, unlike the corresponding reactions of Cl to form HCl . HBr is also much more reactive with OH , lowering its atmospheric lifetime to a couple of days compared to about one month for HCl [Brasseur et al., 1999]. Bromine's contributions to ozone depletion associated with heterogeneous chemistry are less direct than those of chlorine, as bromine is not directly activated by PSCs or other aerosols. However, bromine does figure in some of the chlorine catalytic cycles which deplete ozone (for example through the role of BrCl) [e.g., Solomon, 1999].

EOS MLS will provide the first global measurements of the stratospheric BrO profile, and its best BrO measurements will be in the altitude range of ~25-45 km [Filipiak, 1999] where ~50% of total bromine is expected to be in the form of BrO . Because of the small BrO

abundances, however, averages of large amounts of EOS MLS data will be required to obtain useful BrO measurements - as was the case for upper stratospheric ClO measurements from UARS MLS.

Some specific objectives and investigations planned with the MLS data are itemized below.

- **Determining the global distribution, and seasonal variations, of stratospheric BrO.** MLS monthly zonal means for BrO will be used to produce the first global climatology of the distribution of stratospheric BrO and its variations on seasonal and interannual time scales. More complex averages, over specific regions and date ranges, will also be examined for their possible usefulness in studying more localized phenomena. Possible examples include polar winter phenomena, with investigations to compare the amount of BrO in the Arctic and Antarctic winter vortices and to study the year-to-year variability in vortex BrO compared with that of ClO. See section 2.2 for more discussion of objectives related to polar winter processes.
- **Testing models of stratospheric bromine chemistry.** The MLS observations of the BrO global distribution, seasonal variations, and diurnal variations near the orbit extremes will be compared with predictions from models of stratospheric bromine chemistry to test whether the dominant features seen in these first global measurements of BrO are captured by current models.
- **Determining and monitoring the total amount of stratospheric bromine.** Model results (from the UGAMP model [e.g., [Chipperfield et al., 1999](#)] whose output is being used for MLS retrieval simulation tests) show that the vast majority of bromine above the stratopause is in the form of BrO in the absence of sunlight. The suitably-averaged MLS observations of nighttime BrO above the stratosphere will be investigated to determine if they form a useful measure of total bromine. These data, with a suitably long mission, could possibly give useful trend information.
- **Possibly, in combination with OMI, providing information on tropospheric BrO.** We will investigate the possibility of using MLS BrO data, in conjunction with BrO column data obtained from OMI, to deduce tropospheric BrO loading. This may be useful in characterizing dramatic enhancements in column BrO observed in the polar spring by the Global Ozone Monitoring Experiment (GOME) [[Wagner and Platt, 1998](#); [Richter et al., 1998](#)]. Any such study would have to take into account the relatively poor temporal and spatial resolution of the MLS observations compared to the column information.

2.1.5 Dynamics and Transport

Adequate understanding of stratospheric ozone variations requires understanding the stratospheric dynamics that affect ozone. An objective of MLS, in collaboration with HIRDLS which provides crucial ‘cross-track’ measurements not available from MLS, is to improve our understanding of stratospheric dynamics. Important contributions from MLS include (1) measurements in the presence of ice clouds and dense volcanic aerosol that can degrade the HIRDLS (and other infrared and visible) measurements and (2) routine measurements every 1.5° along the orbit track (compared to the ~4° spacing of the HIRDLS routine operational measurements).

2.1.5.1 Dynamical effects on ozone trends

Downward trends have been seen in mid-latitude ozone [e.g., SPARC, 1998, WMO 1999, Staehelin et al., 2002, and references therein]. The trends arise from a combination of multiple chemical and dynamical processes. Variability and trends in wave activity, tropopause characteristics and other upper tropospheric dynamical processes have a direct effect on midlatitude column ozone. Wave activity propagating into the stratosphere and Rossby wave-breaking also affect stratospheric ozone distributions.

Hood et al. [1999] related trends in midlatitude column ozone to those in Rossby wave breaking and lower stratospheric zonal winds, and found that as much as 40% (25%) of the midlatitude ozone decline in February (March) could be attributed to trends in wave-breaking. Fusco and Salby [1999] also concluded that trends in planetary wave activity could account for much of the wintertime midlatitude ozone trends in the northern hemisphere. Hoppel et al. [1999] showed traveling wave features in the 20-30 km altitude range in ozone in the summertime; the analysis of Wagner and Bowman [2000] indicated that these features arose from breaking of westward-moving waves that propagated up into the middle stratosphere. Other studies have suggested “frozen in” variations from the vortex breakup as an explanation for summertime trace gas variability [see, e.g., Orsolini, 2001]. Several other studies (see references in Hood et al. [1999]) have shown relationships between total ozone trends and trends in dynamical variables that directly affect total ozone, including tropopause height. In addition, numerous studies show a strong relationship of both day-to-day timescale [see Hood et al., 2001, and references therein] and interannual [e.g., Appenzeller et al., 2000; Orsolini et al., 1998; Orsolini and Doblas-Reyes, 2002; and references therein] variability in column ozone to dynamical processes. These dynamical and transport processes are also intimately involved in extra-tropical stratosphere-troposphere exchange, as a substantial portion of that exchange takes place as a result of quasi-isentropic transport on isentropes that encompass both stratospheric and tropospheric regions [e.g., Holton et al., 1995; Appenzeller et al., 1996; and references therein]. In order to adequately separate changes in ozone related directly to dynamical processes from those more directly related to chemical processes, we need a more detailed understanding, based on verifying model processes with data analyses, of the exact mechanisms and transport processes leading to these dynamically induced ozone changes. Global 3D ozone fields can be used in conjunction with transport models to diagnose the contributions of various processes to observed changes. Allen and Nakamura [2002], for example, used "reconstructed" hemispheric ozone fields and assimilated meteorological analyses to separate and examine the roles of specific dynamical processes in ozone "minihole" formation. Their analysis was limited, however, by having to use a 3D ozone field reconstructed from sparse solar occultation data.

The MLS and HIRDLS global daily 3D ozone (and other) fields will make analyses of the type described above much more powerful, and the following studies are expected to be performed:

- **Quantifying and understanding the roles of various dynamical processes in day-to-day and interannual changes in ozone.** Daily 3D ozone fields from MLS and HIRDLS will be used in analyses similar to that of Allen and Nakamura [2002] to quantify the roles of various dynamical processes in day-to-day, seasonal and interannual ozone changes. Relationships between lower stratospheric temperatures, tropopause heights, column ozone, 3D ozone structure and wave fluxes (all of which can be derived from MLS and/or HIRDLS data) will be studied to help understand day-to-day variations in column ozone and interannual variability therein [e.g., Sabutis et al., 1997; Figure 2.1.5-1].

Figure 2.1.5-1, now at end after references will be inserted somewhere near here.

- **Elucidating and understanding the relationships between possible trends in stratospheric ozone, tropopause height, and Upper Troposphere / Lower Stratosphere (UT/LS) dynamical variations.** MLS and HIRDLS temperature and geopotential height observations in the tropopause region provide direct measurement of tropopause height and UT/LS dynamical variations (independent of the analyzed operational meteorological datasets) and will be correlated with column ozone derived from MLS/HIRDLS to elucidate the relationships between changes in the two, and analyzed with ozone profiles to understand the mechanisms behind these relationships. In particular, they will be used in studies similar to that of Steinbrecht et al. [1998] to examine relationships between ozone trends and trends in tropopause height and temperature. Relationships between trends in wave fluxes and other dynamical variations [e.g., Hood et al., 1999] can also be explored.
- **Improving understanding of summertime ozone variability.** Dynamical variables (temperature/geopotential height, waves, EP fluxes, measures of mixing) will be analyzed with MLS 3D ozone fields to obtain a more complete understanding of summertime ozone variability in the lower to middle stratosphere.
- **Improving understanding of extratropical stratosphere-troposphere exchange.** MLS and HIRDLS O₃ and H₂O in the UT/LS region will be used to study the exchange of air between stratosphere and troposphere via transport along isentropic surfaces that cross the tropopause. The 3D global daily fields from Aura are suited to examining this directly, by identifying H₂O and O₃ values on isentropic surfaces characteristic of the troposphere and stratosphere, or via methods such as correlations between H₂O and ozone, which have a distinctly different character in the troposphere and stratosphere. To use the latter method, averaging techniques will be developed to obtain robust tracer correlations from the noisy Aura data. Trends and variability in extratropical stratosphere-troposphere exchange thus identified will also be related to the other dynamical processes discussed here.

2.1.5.2 Effects of polar processes on midlatitude ozone

Polar vortex processes may affect midlatitude ozone through either the export of PSC-activated air to midlatitudes with subsequent in-situ ozone loss, or the dilution of mid-latitude ozone in spring when the vortex breaks up and ozone-depleted air is transported to midlatitudes.

Norton and Chipperfield [1995] examined the transport of chemically activated air out of the Arctic vortex during winter in a 3D chemical transport model (CTM) study; they found large interannual variability in the amount of activated air exported from the vortex, ranging from 5% to 25% of the mass at midlatitudes (10 to 50% of vortex mass) near 475K; larger amounts were seen near and below 400K. They concluded that in situ chemical loss in midlatitudes on polar stratospheric cloud (PSC) activated air exported from the vortex could contribute significantly to midlatitude ozone loss, but could not account for much of the large midlatitude ozone decrease seen in the 1992-1993 Arctic winter.

Knudsen et al. [1998], Knudsen and Grooss [2000], and Knudsen and Andersen [2001] used transport calculations to examine the effects of dilution on midlatitude ozone in the

Arctic, that is, midlatitude changes resulting from export of ozone depleted air from the polar vortex. Hauchecorne et al. [2002] used a high resolution advection model to examine transport of ozone-depleted air from the Arctic vortex. Atkinson and Plumb [1997], Öllers et al. [2002], and Li et al. [2002] used trajectory models and a CTM (Li et al.) to examine transport of ozone-depleted air out of the Antarctic vortex. Each of these studies found significant roles for dilution in ozone changes at midlatitudes. Millard et al. [2002] and Lee et al. [2002] used CTM studies to examine the roles of dilution in the Arctic and Antarctic, respectively. Millard et al. [2002] found large interannual variability in the impact of polar processes on midlatitude ozone loss, with large differences between comparably cold winters depending on the symmetry and evolution of the vortex. Lee et al. [2002] noted the importance of in-situ ozone loss in the “subvortex” in the Antarctic, where (because of weaker confinement) ozone-depleted air was more readily transported to midlatitudes. Li et al. [2002] found that transport out of the vortex could explain about 10% of the observed midlatitude ozone decline in the SH in the period *before* the vortex breakup (September, October).

These studies rely heavily on model results, with few data available for verification. Knudsen et al. [1998], Knudsen and Grooss [2000], Knudsen and Andersen [2001], and Atkinson and Plumb [1997] rely on vortex-averaged estimates of ozone loss or ozone reconstructed from sparse solar occultation observations to initialize their models of transport out of the vortex. Norton and Chipperfield [1995], Millard et al. [2002], and Lee et al. [2002] are primarily CTM studies, with little data - and no global daily 3D data - to compare with their results. The Aura data will provide the means for much more detailed study of these phenomena, and some objectives are itemized below.

- **Improved understanding and quantification of export of vortex-processed air to mid-latitudes.** Observed evolution of 3D ozone and other trace gas fields (HNO_3 , H_2O , N_2O) will be examined to directly observe export of ozone depleted (denitrified, dehydrated) air from the vortex during breakup in spring or during stratospheric sudden warmings. Morrey and Harwood [1998] and Santee et al. [1999], for example, used UARS MLS H_2O and HNO_3 , respectively, to obtain information about export of dehydrated or denitrified air from the Antarctic vortex.
- **Applying tracer correlation methods to Aura data to identify and characterize vortex air that has been transported to mid-latitudes.** Tracer correlations based on aircraft, balloon and satellite data have been shown to be a powerful tool for distinguishing air masses with different histories and thus for understanding many atmospheric processes [e.g., Michelsen et al., 1999; Manney et al., 2000; and references therein]. Careful methods for data selection, sorting and averaging will be developed for applying correlation techniques to the Aura data, which are noisier than the aircraft, balloon and satellite data used in previous tracer correlation studies. Methods will be devised to use “anomalies” in tracer correlations to identify vortex air that has been transported to midlatitudes. Tracer correlations between vortex and extravortex differ as early as early November when the polar vortex is developing [e.g., Michelsen, et al., 1998; Manney et al., 2000]; Figure 2.1.5-2 shows how differences between ozone/ N_2O correlations in early November 1994 could be used to identify filaments originating in different air masses. Vortex and extravortex correlations differ even more later in winter, because of both effects of descent and mixing and any chemical loss (ozone loss, denitrification) that may have taken place; thus anomalies in correlations in midlatitude observations should be easier to identify.

Figure 2.1.5-2, now located at end after references, will be inserted somewhere near here.

- **Using air parcel histories and Aura data to study and analyze observations of vortex air at midlatitudes.** Air parcel history calculations will be used to verify the above observations of air escaping from the vortex (Manney et al. [2000], for example, used high resolution trajectory profile calculations and changes in tracer correlations to identify laminae arising from filaments of air drawn off the vortex) and to detail the origins of that air and compare their calculated histories with long-lived trace gas observations. The MLS 1.5 degree along-track spacing makes its data particularly useful for identifying filaments drawn off the vortex in isentropic fields; and the approximately 1 km vertical resolution of HIRDLS makes its data particularly suited to detecting laminae in profiles that arise from such filaments. In addition the Aura long-lived trace gas observations are well-suited to use with high-resolution balloon observations and air parcel histories to help understand the origins of air sampled by balloons.
- **Improving and testing models of transport of air out of the polar vortex.** 3D trace gas fields (ozone and others) will be used to initialize CTMs for detailed modeling studies of transport of air out of the polar vortex, such as those of Millard et al. [2002] and Lee et al. [2002], and others (see above). The observed 3D fields will also be used to verify the model results. Ozone loss estimates from EOS MLS data (Section 2.2) will also be used to initialize studies analogous to those of Knudsen et al. [1998], Knudsen and Grooss [2000], Knudsen and Andersen [2001], and Atkinson and Plumb [1997] modeling the transport of ozone depleted air out of the vortex, and EOS data used to verify these model results.
- **Improving our understanding of the effects of vortex-exported air on in situ processing at midlatitudes.** Where export of PSC-processed air from the vortex is identified, MLS and HIRDLS observations will be used to identify its composition for modeling in situ loss in midlatitudes (models of chemistry along trajectories would be suited to these studies).

2.1.5.3 Tropical transport and its effect on stratospheric ozone

The tropical (rising) branch of the stratospheric residual circulation brings tropospheric air into the stratosphere. There is substantial evidence [see, e.g., Holton et al., 1995] that this dynamically-driven circulation extends **downward** into the upper few kilometers of the tropical troposphere (14-16.5 km), giving it a primary role in shaping the conditions in the tropical tropopause. In particular, the seasonality [Reid and Gage, 1996] and interannual variability [Randel et al., 2000] in the temperature (and hence water vapor mixing ratio) of the tropical tropopause are consistent with control by the stratospheric residual circulation, which in turn is driven by extratropical wave activity. Air in the tropical lowermost stratosphere is, therefore, very low in ozone but rich in trace constituents with surface sources and stratospheric sinks, like methane and the chlorofluorocarbons.

While the previous generation satellite observations have revealed the large-scale features and have permitted inferences about the dynamics of the lower stratosphere, the tropical tropopause in particular is still insufficiently understood. The roles of the stratospheric residual circulation on the one hand and convectively driven disturbances (e.g., convection itself, convectively generated Kelvin waves, radiative cooling above convective anvils) on the other hand are still hotly debated [e.g., Sherwood and Dessler, 2001]. Understanding the tropical tropopause is crucially important for predicting future trends in stratospheric water

vapor and hence the recovery of the ozone hole, yet our understanding is so poor that we cannot even explain the sign of the recent trend in water vapor (minus methane oxidation). See section 2.3.1 for more discussion of objectives related to improving our understanding of dehydration of the stratosphere and related tropical tropopause investigations.

Some specific objectives to be addressed with MLS, and other Aura, data include those itemized below.

- **Improve our understanding of the stratospheric “tape recorder”, and obtain better estimates of vertical velocity and other parameters as in Mote et al. [1998]**

Rising air in the tropics remains remarkably little changed by other stratospheric air or by vertical mixing. Avallone and Prather [1996] attempted to quantify the rate of dilution on the basis of ozone profiles, while Hall and Waugh [1997] and Mote et al. [1998] used UARS measurements of methane and water vapor to simultaneously constrain the horizontal dilution and vertical diffusion rates. These rates are surprisingly low, as attested by the fact that the seasonal cycle of water vapor, imprinted at the tropical tropopause, is preserved for a year and a half as the air rises. The new multi-year dataset of water vapor and methane from Aura will permit us to revisit this issue, comparing the new values with those of the early-to-mid 1990s to look for evidence of interdecadal variability or possible changes in stratospheric dynamics resulting from anthropogenic changes in atmospheric composition.

- **Improve our understanding of the seasonal cycle and quasi-biennial oscillation (QBO), and how it affects stratospheric ozone and other trace constituents**

The QBO influences the distribution of stratospheric trace constituents like ozone in a number of important ways [Baldwin et al., 2001]. First, it induces a secondary residual circulation, with tropical ascent either reduced or enhanced depending on the phase of the QBO; this circulation is closed via a subtropical branch, which tends to be stronger in the northern hemisphere. One consequence of this residual circulation, important for ozone and water vapor, is the fluctuations in temperature that result, which probably affect the entry value of water vapor [Giorgetta and Bengtsson, 1999] and certainly affect the temperature-dependent photochemistry of ozone. Second, the QBO influences extratropical wave propagation into the tropics; such waves shape the distribution of trace constituents by inducing isentropic mixing zones, at the edge of which steep tracer gradients are observed [Gray, 2000]. As Dunkerton [2001] pointed out, the interaction of the QBO and annual cycle slowly populates in phase space as QBO transitions occur at different times in the calendar year. The new [Aura](#) measurements will permit us to check his predictions and extend our understanding of how the QBO and annual cycle interact to influence tracer distributions.

2.1.5.4 Gravity waves and their effect on circulation and ozone

Gravity waves (GWs) play important roles in determining the atmospheric circulation and thermal structure [e.g., Lindzen, 1981; Holton, 1982]. Without such small-scale wave forcing, the modeled vortices would be too strong and upper stratospheric temperatures would be too low in the polar stratosphere [Hamilton et al., 1994; Pawson et al., 2000]. Since the scales of most GWs cannot be resolved [by most comprehensive atmospheric models such as GCMs, parameterization is needed to account for wave momentum and energy deposition](#). Current

GW parameterizations are still very crude [Lindzen, 1981; Fritts and Lu, 1993; Medvedev and Klaassen, 1995; Hine, 1997], allowing only ‘reasonable’ circulation patterns and thermal structure to be obtained in the model.

Ozone loss calculations and predictions require realistic temperature and wind fields from models or assimilation analyses where uncertainties of these variables remain unacceptable [Randel and Wu, 1999; Marshall, 2002]. Studies have shown that model results at this level of accuracy are very sensitive to their GW drag parameterization. Improvements are needed in observing, understanding and modeling of global GW properties and variability.

GWs also affect ozone depletion through their role in forming polar stratospheric clouds (PSCs) [e.g., Cariolle et al., 1989; Carslaw et al., 1998]. PSCs near the vortex edge, **due to the earlier exposure they have to sunlight, can have a larger effect on ozone loss than PSCs near vortex center**. Lee et al. [2001] argued that more PSCs on the vortex edge in the future could lead to delay in the ozone recovery. One of the uncertainties in predicting future ozone loss is the **uncertainty in predicting** PSC formation and coverage. Both GWs [Carslaw et al., 1998] and synoptic-scale tropospheric forcing [Teitelbaum et al., 2001] can contribute to PSC formation; quantitative understanding of these processes is needed. See section 2.2.2 for more discussion on this topic.

Aura observations and collaborative theoretical investigations can make substantial contributions towards better understanding and modeling global GWs and their effects. Some objectives and expected investigations are itemized below.

- **Understanding global gravity wave distribution and propagation.** MLS and HIRDLS will provide unique measurements of GWs in the stratosphere and mesosphere. MLS will measure waves of long ($> \sim 5$ km) vertical and short ($< \sim 100$ km) horizontal wavelengths, significantly improved over UARS MLS in detecting short vertical-scale GWs [e.g., Wu and Waters, 1996; McLandress et al., 2000]. HIRDLS observations will feature GWs of relatively short ($> \sim 1$ km) vertical wavelengths but long ($> \sim 300$ km) horizontal wavelengths from optically-thin radiances, similar to CRISTA [Preusse et al., 2002]. MLS complements HIRDLS by using observations from optically-thick radiances that provide shorter horizontal scales and wave propagation direction [Wu and Waters, 1997].
- **Characterization and modeling of gravity wave sources.** One of the key uncertainties in GW studies is the nature, location, and variation of their sources. Gravity waves are known to be generated from weather-related processes, such as: flow over mountains [e.g., Ralph et al., 1993], fronts [e.g., Eckermann and Vincent, 1995], squall lines [e.g., Alexander et al., 1995], deep convection [e.g., Alexander and Holton, 1997], cyclogenesis [e.g., Powers, 1997], typhoons [e.g., Sato, 1993], and jet streams [e.g., Kaplan et al., 1997]. Global/regional studies with satellite observations and GW model simulations have shown great strength in identifying and understanding GW source and propagation properties [Alexander, 1998; Eckermann and Preusse, 1999; Jiang et al., 2002]. The new observations from Aura will be used to test theories and model parameterization schemes for gravity wave drag, and also - **very importantly** - to constrain tunable parameters in the models.
- **Quantification of gravity wave effects on PSC formation.** GW-related PSC formation will be investigated with the HIRDLS, TES and OMI PSC measurements and the MLS GW measurements that can be made in the presence of PSCs. The near-simultaneity and coincidence of these Aura measurements will be especially valuable for the studies of this sort since PSCs and GWs **vary rapidly in time and space**.

Complexities of GWs (e.g., wave spectra and sources) require observations from many aspects, including vertical amplitude growth and wave spectra. For example, observations from radiosonde networks [Allen and Vincent, 1995] and Global Positioning System (GPS) satellite occultation measurements [Tsuda et al., 2000] will broaden the information that can be obtained on GW properties. Joint studies among Aura, GPS and ground-based observations will offer unprecedented studies of the complexities associated with GW processes.

2.1.6 Mesospheric topics

Processes in the mesosphere can affect stratospheric ozone chemistry and circulation, and understanding the mesosphere is thus valuable for understanding stratospheric ozone. Also, because some of the chemical reactions that affect ozone in the stratosphere also occur in an overall simpler scheme in the mesosphere, the mesosphere can sometimes be used to test our understanding of aspects of stratospheric chemistry that could be more difficult to test by direct stratospheric observations.

Although MLS is targeted at the stratosphere and upper troposphere, it will make some measurements up through the mesosphere. These include daily global maps of temperature, O₃, H₂O, CO and OH, daily zonal means of HCl, and monthly zonal means of HO₂ and possibly HCN. Its daily global daytime and nighttime measurements will give an unprecedentedly clear view of the seasonal cycle of phenomena associated with these measurements. This view complements the more detailed view of the diurnal cycles which may be obtained from the precessing orbits of UARS and TIMED.

Ozone balance in the mesosphere is controlled largely by the HO_x catalytic cycle, and MLS will measure more HO_x species than has previously been possible, as well as H₂O, which provides the source of HO_x. This should lead to a more detailed understanding of mesospheric ozone chemistry, and mesospheric HO_x chemistry that is also operative in the stratosphere as discussed in section 2.1.3. MLS data will be used in studies to test the mesospheric predictions of models such as WACCM and Time-GCM.

All the MLS mesospheric species measurements will contribute to improving our understanding of mesospheric dynamics. UARS MLS radiances from H₂O, for example, clearly showed the strong summer-to-winter meridional flow in the mesosphere [Pumphrey and Harwood, 1997]. A strong solar cycle dependence in upper mesosphere water has been observed with HALOE [e.g., Chandra et al., 1997]. The EOS MLS daily global measurements will provide greater detail about how the solar cycle affects the seasonal cycle of H₂O. CO, having a source in the thermosphere due to CO₂ photodissociation, is a good tracer of vertical transport within the mesosphere and between the mesosphere and stratosphere. MLS measurement of it will show the descent of mesospheric air into the winter polar stratosphere [see section 2.2.1]. Much of the mesospheric dynamics is driven by gravity waves, which will be studied by MLS as described in section 2.1.5.4.

Polar mesospheric clouds (PMCs, also called noctilucent clouds when they are visible from the ground) occur at the high latitude summer mesopause. Quoting from DeLand et al. [2002]:

“High latitude summer mesopause temperatures are the lowest found on Earth, typically reaching 130 K [Lübkin, 1999]. Such low temperatures imply that the particles comprising PMCs are some form of condensate. Hervig et al. [2001] recently provided strong evidence that PMC consist of water ice. Stevens et al. [2001] have shown from MAHRSI and CRISTA satellite measurements that supersaturation is not always required for PMC

existence, nor is it sufficient. Thus, we anticipate that PMC observations will be dependent on water vapor concentration and mesospheric temperatures. Both of these quantities are expected to be sensitive to solar activity variations and long-term climate change. Thomas et al. [1991] used observations from the Nimbus-7 SBUV instrument to demonstrate a solar activity dependence in PMC frequency. Shettle et al. [2002a] used three satellite data sets to show that “decadal change” occurred between the 1980s and the 1990s. ... Data from the SAGE II instrument have recently been analyzed to produce a 16-year data set covering 1.5 solar cycles [Shettle et al., 2002b].”

DeLand et al. [2002] present PMC results from five separate SBUV instruments over a time period of 23 years, and find that the distribution of PMC brightness by season appears to be changing over time. EOS MLS will be able to observe the temperature and H₂O mixing ratio at the altitude of the PMCs, with sufficient precision to correlate seasonal and interannual changes in temperature and H₂O with the occurrence of PMCs, and thus will provide more information on the processes that form these clouds and how they respond to, or may be very sensitive indicators of, climate change.

Additional mesospheric topics that might be investigated with MLS data include the following.

- (1) the possibility of mesospheric sources of HCN [Kopp et al., 1990] and N₂O [Zipf and Prasad, 1982],
- (2) possible improvement in the modeling of meteoric metal layers which require estimates of H₂O, O₃, HCl and possibly CO that can react with the easily detectable (by other means than MLS) Na, Ca, and Fe atoms to give the undetectable species in which the metals are presumably transported to the lower regions of the atmosphere.
- (3) using daily MLS mesospheric H₂O measurements to provide information on whether “small comets” might be a significant contributor to water in the mesosphere as has been suggested [e.g., Frank and Sigwarth, 1993; 1997] but is controversial [e.g., Dessler, 1991; World Meteorological Organization, 1999 (chapter 6, 6.3.1)].

2.2 Polar Winter Processes

A primary objective for Aura is to answer the question: “Is the ozone layer changing as expected?” This seemingly straightforward question encompasses a myriad of intertwined issues. In this and the following subsections, we will highlight several of the critical unresolved questions related to polar processes and ozone loss, briefly review the progress made in tackling these questions up to now to provide context, and then illustrate how the large complement of high-quality data from Aura will be applied to studying these issues in the future. Although these investigations will focus on specific details, the overarching goal in addressing these questions is to aid in the development of a reliable and accurate predictive capability so that the changes in ozone in response to anticipated reductions in stratospheric chlorine loading and possible climate change in the coming years can be assessed.

In recent years the average area of the Antarctic ozone hole has continued to increase, although its growth rate is less rapid now than it was during the 1980s, and it has frequently persisted into early summer [Uchino et al., 1999]. Because the size, depth, and persistence of the ozone hole vary substantially from year to year (the 2000 ozone hole was the largest on

record, whereas the 2002 ozone hole was one of the smallest in the past 10 years [Van Velthoven et al., 2002], identifying the maximum in the severity of ozone depletion may be difficult. Nevertheless, Hofmann et al. [1997] have argued that by carefully monitoring certain indicators over a limited altitude range in the lower stratosphere, recovery in the Antarctic ozone profile may be conclusively detected as early as 2008, which is within the Aura mission operational lifetime. Simulations with coupled chemistry-climate models have also suggested that the Antarctic ozone hole is currently at or near maximum, with recovery predicted to start in years ranging from 2001 to 2008, depending on the model [World Meteorological Organization, 2002]. Thus EOS MLS will be in the unique position of being able to measure both the total abundance of ozone-destroying chemicals and the abundance of ozone itself during the period of expected peak ozone depletion and the early stages of ozone hole recovery.

Several factors may impede the recovery of stratospheric ozone, however, despite the reduction in total chlorine abundances. Although increasing abundances of the greenhouse gas CO₂ are projected to accelerate the return of globally averaged column ozone to pre-1980 amounts [Rosenfield et al., 2002], they will delay recovery in the Arctic polar lower stratosphere [e.g., Shindell et al., 1998; Austin et al., 2000; Rosenfield et al., 2002; Pitari et al., 2002]. Changes in stratospheric temperature and water vapor are coupled to changes in ozone through radiative, chemical, and dynamical processes. A statistically-significant cooling of the polar lower stratosphere has occurred over the past three decades [Ramaswamy et al., 2001]; this cooling has been linked in large measure to springtime ozone depletion, with smaller contributions from the increases in well-mixed greenhouse gases and stratospheric water vapor [WMO, 2002]. Climate models generally predict that increases in well-mixed greenhouse gases will continue to cause decreases in the temperature of the lower stratosphere, with ozone changes enhancing the stratospheric cooling over the next 20 years [World Meteorological Organization, 2002]. A strong correlation has been noted between lower polar temperatures and the persistence of the vortex in spring over the last 40 years [Waugh et al., 1999], and some (but not all) models predict an increase in the strength and stability of the polar vortices as greenhouse gas concentrations rise [WMO, 2002]. In addition to changes in temperature, a substantial increase in stratospheric water vapor (~1%/year over ~50 years), indicative of long-term climate change, has been documented [Rosenlof et al., 2001]. Increasing water vapor not only accentuates radiative cooling in the lower stratosphere [e.g., Forster and Shine, 1999; Dvortsov and Solomon, 2001], it also enhances the effectiveness of the heterogeneous reactions that control chlorine partitioning [e.g., Chipperfield and Pyle, 1998]. Both lower temperatures and larger H₂O abundances promote the formation of polar stratospheric clouds (PSCs) and the occurrence of denitrification; in fact, Tabazadeh et al. [2000] have shown that the effect on Arctic PSC formation and denitrification of increasing the H₂O mixing ratio by 1 ppmv is comparable to that of lowering the temperature by 1 K. Although these effects are expected to have a negligible impact in the Antarctic lower stratosphere, where chlorine activation and ozone depletion are already nearly complete [e.g., Rosenfield et al., 2002], they may lead to a substantial intensification of ozone loss within a future colder, more humid, and more persistent vortex in the Arctic [e.g., Chipperfield and Pyle, 1998; Waibel et al. 1999; Tabazadeh et al. 2000].

Thus the Arctic lower stratosphere may currently stand at a threshold, with a realistic possibility for much more severe ozone loss in the near future (though the occurrence of an Arctic ozone "hole" is unlikely [World Meteorological Organization, 2002]). The importance of Aura's capability for making measurements directly relevant to polar processes and ozone loss during this pivotal time cannot be overstated. Indeed, at the Arctic Ozone Loss Workshop

held in Potsdam, Germany, in March 2002, a general strategy for future observations and process studies was outlined. Quoting from the Report on the Workshop [Rex et al., 2002b]: “... It was agreed that continuous observations of O₃, ClO, H₂O, HNO₃, temperatures and polar stratospheric clouds, preferably with large spatial coverage, will be critical for characterising the changes expected as earth's climate changes and as chlorine loading decreases throughout the century. Process studies addressing nucleation of PSC particles and subsequent chlorine activation and denitrification will be crucial to constrain model parameterisations of particle microphysics, necessary for developing a credible predictive capability. New studies of early- and mid-winter ozone losses and the possible role of unknown halogen reactions will be important for reducing the probability of a ‘surprise’. ... In particular, research programmes should focus on the response of Arctic ozone to changes in wave driving, source gas abundances (e.g., chlorine, bromine, methane, and H₂O), radiative forcing, and denitrification.” The Aura mission in general, and EOS MLS in particular, was designed for these kinds of studies, and, as will be seen in detail in the following subsections, MLS will make important contributions to many of these issues.

Although after nearly two decades of study the processes involved in polar ozone loss are felt to be well understood, we should be careful to guard against complacency. The unprecedented bifurcation of the 2002 Antarctic ozone hole [van Velthoven et al., 2002] serves as a reminder that the stratosphere does not always behave ‘as expected’.

2.2.1 Polar Winter Vortex Development and Evolution

Understanding vortex evolution and variability during the fall/early winter, and their relation to vortex conditions later in winter when polar processing occurs, is crucial to improving our knowledge of how transport, mixing, and dynamical conditions affect this processing. Descent and mixing in and around the vortex, and the interannual variability in these processes, must be better quantified in order to estimate their effects on tracer correlations for denitrification and/or ozone loss studies. It is also important to validate transport model simulations of descent and mixing, which are essential to many polar process studies.

Key questions related to the development and evolution of the stratospheric winter polar vortices that will be addressed with Aura data are itemized below.

- **What are the effects of the development of low temperatures, a confined vortex region, and variations in early winter vortex strength and morphology on the onset and timing of polar processing, and on processing that takes place later in winter? What are the detailed distributions of trace gases in and around the developing polar vortex in fall?** The development of the stratospheric polar vortex in fall and early winter, interannual variability therein, and associated transport processes, strongly affect vortex conditions later in winter when most polar processing takes place [e.g., Manney et al., 1999a, 2001; Manney and Sabutis, 2000]. Dynamics studies show considerable interannual variability in the fall/early winter polar vortex [e.g., Manney and Sabutis, 2000; Manney et al., 2001, 2002b], especially in the Arctic. Calculations of trace-gas transport in the model simulations discussed by Manney et al. [2002b] indicate that meteorological variability would also be expected to lead to significant variability in transport. Analyses of POAM and UARS MLS data [C. E. Randall, G. L. Manney, unpublished results] do, indeed, suggest considerable interannual variability in early winter transport. Studies of the synoptic evolution of the fall/early winter polar vortex show that a common, but not

ubiquitous, feature is a double-peaked jet in the upper stratosphere [e.g., Manney and Sabutis, 2000; Manney et al., 2001, 2002b]. Transport in/through the upper stratosphere at this time has not been much studied, because few long-lived trace gas data - especially hemispheric, temporally-extended data - exist for detailed studies; this transport is important to polar-process studies since air in this region in early winter eventually descends to the lower stratosphere in late winter or spring [e.g., Plumb et al., 2002, and references therein]. Manney et al. [1999b, 2000] used ATMOS data from November 1994 and trajectory modeling to study transport in and around the Arctic vortex in fall; they found that considerable vortex descent had already taken place in early November, and had been accompanied by sufficient weak or sporadic mixing that tracer correlations within the developing vortex (the "protovortex") had been altered from those outside (this was also apparent in the $\text{NO}_y/\text{N}_2\text{O}$ correlations shown by Michelsen et al. [1998]); this is consistent with results seen by Ray et al. [2002] for the fall 1999 vortex. Manney et al. [2000] also found extensive filamentation in early winter, arising from both air drawn off the protovortex and air being drawn up from low latitudes around the vortex or into the developing anticyclone. Although the above studies have shown some aspects of transport in and around the early winter vortex, there are currently few data available for more thorough transport studies during this period. The most useful data for these studies are hemispheric or global daily fields of long-lived trace gases (including, for these purposes, H_2O), and there are currently no multiannual datasets of such fields. We will use global daily fields of N_2O and H_2O from MLS and trace gases from HIRDLS to study many aspects of transport in early winter: First, we will analyze the evolution of these fields with respect to the polar vortex, and use them (with the aid of trajectory calculations) to trace the origins of the air that ends up in the polar vortex as it develops. This will include studies of interannual variability in the magnitude and patterns of descent, and mixing processes and filamentation (MLS will have sufficient vertical resolution to observe some laminae in trace gas profiles, and its approximately 1.5 degree along-track horizontal coverage will aid in detecting filaments that result in lamination; the approximately 1 km vertical resolution of HIRDLS will be adequate to observe many laminae). We will derive methods for examining trace gas correlations from MLS and HIRDLS observations; in early winter, both correlations between long-lived tracers, and correlations of ozone with long-lived tracers, can be used to detect changes in tracer correlations between midlatitudes and the developing vortex arising from descent and mixing [e.g., Michelsen et al., 1998; Manney et al., 2000], and to identify vortex air that has been drawn out into midlatitudes [e.g., Manney et al., 2000; Figure 2.1.5-2]. MLS and HIRDLS temperature measurements will be used in comparison with temperatures from standard meteorological analyses to give us more precise knowledge of the evolution of temperatures and their uncertainties as they drop toward levels where polar processing can occur. In addition, continuous 3D ozone fields will allow extensions of studies such as Kawa et al. [2002] examining the contributions of chemical and dynamical effects to the distribution of ozone in the early winter polar vortex.

- **How much descent occurs in the vortices in fall and early winter, and where does the air that in late winter/spring is in the lower stratospheric vortex originate in fall?** It is important to study the entire depth of the stratosphere, since a substantial fraction of air from the mesosphere and upper stratosphere descends to the lower stratosphere by late winter [e.g., Fisher et al., 1993; Manney et al., 1994; Ray et al., 2002; Plumb et al., 2002; and references therein]. Extensive meteorological data have heretofore been almost entirely limited to the stratosphere; current data with which to study descent from the

mesosphere are very sparse. We will use MLS geopotential height and temperature measurements extending into the mesosphere to calculate winds and descent rates at higher altitudes, extending our knowledge of the dynamics of the polar vortex above the stratopause and modeling descent from the mesosphere, which is essential to many polar processing studies. Analyses of H₂O transport using Aura data will provide further information. In addition, we will devise appropriate ways of averaging the "noisy" MLS products CO and HCN to extract information on descent from the mesosphere (see section 2.1.6).

- **What are the effects of varying amounts of mixing, both within the vortex, and between the vortex and its exterior? What are the effects of PSC-processed, denitrified and/or dehydrated, and ozone-depleted air that escapes the vortex (either during vortex breakup in spring or sporadically during winter) on global ozone?** Ray et al. [2002] showed that balloon observations of many long-lived tracers (including SF₆, a good tracer of mesospheric air) were best matched by a model that included differential descent within the vortex, including a substantial fraction of mesospheric air, and mixing within the vortex increasing after early winter. Analyses of global daily N₂O and H₂O fields (as well as O₃ and HNO₃ under conditions where polar processing is not ongoing, such as during the vortex breakup) from Aura will allow direct observation of the presence, or lack thereof, of air with vortex characteristics outside the vortex and, conversely, intrusions of midlatitude air into the vortex. 3D fields of trace gases within the vortex (including long-lived species, ClO and ozone), along with trajectory calculations to detail their histories, will be used to analyze mixing within the vortex. Such mixing is important not only for understanding the changes in trace gas correlations that result, but it also has important implications for, for example, the spread of PSC-processed air throughout the Arctic vortex [e.g., Manney et al., 2002c] and the uniformity, or lack thereof, of ozone loss within the vortex. The effects of processed vortex air on midlatitudes are discussed in detail in section 2.1.5.2.

2.2.2 Polar Stratospheric Clouds

Polar stratospheric clouds (PSCs) play several pivotal roles in controlling the cumulative amount of polar ozone loss [e.g., Solomon, 1999]. Despite a considerable number of observational, laboratory, and modeling studies over the last 15 years, however, specific PSC formation mechanisms remain uncertain [for more complete surveys of PSC-related studies, see Tolbert, 1994; Carslaw et al., 1997; Peter, 1997; WMO, 1999, 2002; Toon et al., 2000].

Some key questions related to polar stratospheric cloud formation that will be addressed with Aura data are given below.

- **What are the phase and composition of PSC particles? In particular, are large, denitrifying PSC particles composed of nitric acid trihydrate (NAT), and what mechanisms for selective nucleation are at work in their formation?** Diagnosing PSC composition using existing satellite measurements is challenging. Hervig et al. [1997] discuss the inherent limitations in studying this issue with satellite limb observations, in particular from UARS HALOE, arising from cloud inhomogeneity over the sample volume. Similarly, individual UARS MLS HNO₃ measurements represent averages over a ~400km x 400km x 6km volume of air. Although both crystalline and liquid PSCs routinely extend continuously over spatial scales comparable to or larger than the MLS footprint [World Meteorological Organization, 1999; Toon et al., 2000], it is possible that

multiple cloud types could exist within the volume of air sensed by MLS. Recently, Santee et al. [2002] assessed the viability of inferring PSC composition by correlating UARS MLS observations of gas-phase HNO_3 with Polar Ozone and Aerosol Measurement II (POAM) observations of aerosol extinction. A Lagrangian approach was used whereby HNO_3 and aerosol extinction data from the Arctic late winter of 1995/1996 were compared to results from equilibrium PSC composition models along air parcel trajectories. This approach was very successful in capturing the broad patterns of PSC development and evolution. A strong correlation was found between low-temperature, low- HNO_3 , and high-extinction points. Extracting detailed quantitative information about the composition of specific PSCs proved very difficult, however. Large uncertainties and a large degree of overlap in the comparisons between models and data precluded conclusive identification of specific PSC types in most cases. Many deficiencies encountered in the Santee et al. [2002] study will be ameliorated with the Aura mission. Better precision and horizontal (and possibly vertical) resolution of the EOS MLS HNO_3 data will reduce ambiguity in data/model comparisons. Perhaps more importantly, PSC composition models are extremely sensitive to both temperature and water vapor concentration [e.g., Hanson and Mauersberger, 1988; Worsnop et al., 1993; Tabazadeh et al., 1994]; lack of data forced Santee et al. [2002] to rely on meteorological analyses for temperature and on an assumed, constant H_2O mixing ratio. PSC composition studies will be greatly aided by having simultaneous, colocated HNO_3 , H_2O , and temperature measurements with good precision (provided by both MLS and HIRDLS). The availability of high-resolution aerosol measurements from HIRDLS and OMI will substantially increase the number of coincidences and will generally afford vastly better data coverage along trajectories than did the POAM occultation data used by Santee et al. [2002]. Because high-latitude data coverage will be continuous (no yaws as on UARS), complete PSC lifecycles, from formation through dissipation, will be observed. In addition, uninterrupted coverage will allow intraseasonal changes in cloud development to be tracked. That is, since the timing, altitude, and geographic location of PSC incidence depend critically on temperature, on the concentrations of H_2O and HNO_3 , and possibly also on the availability of heterogeneous freezing nuclei [e.g., Drdla et al., 2002], details of the PSC formation process, and the dominant particle composition, may change over the course of a PSC season. EOS MLS and HIRDLS measurements will also enable interhemispheric and interannual comparisons of PSC formation and evolution based on common datasets. Thus the improvements in the EOS MLS measurements over those of UARS MLS, and the availability of the high-resolution HIRDLS measurements, will make studies similar to that of Santee et al. [2002] (but perhaps employing more sophisticated microphysical models) more fruitful. See section 2.2.3 for related discussions.

- **What are the dominant mechanisms governing PSC formation?** Whether or not the composition of individual PSCs can be determined from EOS MLS or HIRDLS measurements, the study of Santee et al. [2002] and previous studies [Santee et al., 1996, 1997, 1998, 1999; Massie et al., 1997; Tabazadeh et al., 2000] have demonstrated that MLS measurements of gas-phase HNO_3 can be used to infer the existence of PSCs. Thus they can be used to investigate the dominant PSC formation mechanisms. Lately much attention has been focused on the role of mesoscale temperature fluctuations induced by mountain waves in promoting cloud formation [Meilinger et al., 1995; Tsias et al., 1997, 1999; Carslaw et al., 1998; Wirth et al., 1999; Rivière et al., 2000; Dörnbrack et al., 2002; Hu et al., 2002]. NAT PSCs have now been detected in the atmosphere [Voigt et al., 2000] (during the 1999/2000 SOLVE/THESEO campaign [Newman et al., 2002]), but the particles were relatively small and were observed in a lee wave cloud. Although these

kinds of particles may contribute substantially to chlorine activation [e.g., Carslaw et al., 1998], their importance for denitrification is still unclear [Tolbert and Toon, 2001; Dhaniyala et al., 2002]. Recently, Teitelbaum et al. [2001] used POAM and TIROS Operational Vertical Sounder (TOVS) data to assert that the primary PSC formation mechanism in the Arctic is synoptic-scale uplift, which is also associated with the occurrence of ozone miniholes. Similarly, Spang et al. [2001] showed that a PSC event evident in Cryogenic Spectrometers and Telescopes for the Atmosphere (CRISTA) HNO_3 data in the Antarctic was caused by a variation in tropopause height associated with an ozone minihole. Saitoh et al. [2002] inferred Arctic PSC composition from Improved Limb Atmospheric Spectrometer (ILAS) aerosol extinction and HNO_3 data and argued that HNO_3 -containing hydrates observed in March had not been influenced by mountain-induced lee waves but rather could be explained from their synoptic-scale temperature histories; similar conclusions had been reached earlier based on aircraft [Tabazadeh et al., 1996] and balloon [Larsen et al., 1997] observations. With the suite of Aura measurements we will be in an excellent position to investigate the influence of synoptic-scale dynamics on PSC formation and compare it with that of mesoscale temperature fluctuations arising from mountain waves. By correlating PSC detections based on both MLS HNO_3 and HIRDLS aerosol extinction measurements with such factors as temperature, tropopause pressure, and column ozone abundances (to help identify ozone miniholes), we will be able to assess the role of synoptic-scale uplift in promoting PSC activity.

- **Will the spatial extent, duration, or frequency of occurrence of PSCs increase in the future in response to climate change and/or natural variability?** Aura observations will afford the opportunity to monitor whether trends in lower stratospheric temperature or water vapor abundance (see section 2.2.1) induce significant changes in the prevalence and/or character of PSCs. This is especially important in the Arctic, where at the present time the lower stratosphere is often only marginally cold enough to support PSCs. Statistics of PSC incidence based on Aura data can be compared with the long-term PSC climatology derived from Stratospheric Aerosol Measurement (SAM) II, Stratospheric Aerosol and Gas (SAGE) II, and POAM II and III instruments [Fromm et al., 2002].

Despite the many advances offered by the Aura instruments, the inherent limitations of satellite measurements will leave some critical questions regarding PSC formation out of reach. For example, with Aura data alone we may not be able to discriminate between homogeneous and heterogeneous freezing mechanisms, let alone identify specific heterogeneous nuclei if they do play a role. Nor will the presence of a few large solid particles in a mostly liquid cloud be discernible. Combining aircraft and balloon data (which have high resolution and precision but which are limited in temporal and spatial scope) with Aura data (which have poorer resolution and precision but which provide spatial context and daily coverage) will be a powerful tool in PSC studies. Together, for example in the context of a polar 'mini-mission', they will allow a broader range of PSC development issues to be addressed than would either one alone.

2.2.3 Denitrification and Dehydration

The different meteorological conditions in the two hemispheres give rise to very dissimilar patterns of denitrification and dehydration. In situ [e.g., Fahey et al., 1990] and satellite [Roche et al., 1994; Santee et al., 1999] observations have revealed that both the temporal and the spatial scale of denitrification are substantially greater in the Antarctic than in the Arctic; the

same can be said of dehydration [e.g., Kelly et al., 1990; Santee et al., 1995]. Modeled ozone loss is sensitive to the degree of both processes [e.g., Portmann et al., 1996; Brasseur et al., 1997; Chipperfield and Pyle, 1998; Drdla and Schoeberl, 2002].

Some key questions related to denitrification and dehydration that will be addressed with Aura data are:

- **What are the mechanisms for denitrification in the polar vortices, and are they the same in both hemispheres? Are denitrification and dehydration coupled or independent processes? Are temperatures near the frost point necessary for denitrification?** Although denitrification clearly requires the existence of large HNO₃-containing solid particles that can sediment appreciably before being subject to evaporation, the exact mechanism responsible is uncertain. In situ observations of low water vapor coincident with low total reactive nitrogen over Antarctica in August/September 1987 were interpreted to imply that the processes of denitrification and dehydration are strongly coupled [Fahey et al., 1989, 1990]. Other studies, however, suggested that denitrification occurs in the absence of substantial dehydration in both hemispheres [e.g., Hamill and Toon, 1990; Hofmann and Deshler, 1991; Rosen et al., 1989; Fahey et al., 1990; Hübler et al., 1990]. Theoretical studies [Salawitch et al., 1989; Toon et al., 1990] suggested that selective nucleation of a small number of large NAT particles can efficiently denitrify the stratosphere without the involvement of dehydrating ice particles. Recently, Carslaw et al. [2002] found that highly selective nucleation of NAT particles over wide regions is necessary to explain observations collected during the 1999/2000 SOLVE/THESEO campaign. Tabazadeh et al. [2000] correlated UARS MLS measurements of HNO₃ and H₂O with Cryogenic Limb Array Etalon Spectrometer (CLAES) measurements of aerosol extinction during the 1992 Antarctic winter and showed that denitrification takes place over an approximately two week period in mid-June, prior to the extensive formation of ice clouds; together the UARS data strongly suggest the occurrence of significant denitrification without significant dehydration. A subsequent study based on UARS MLS H₂O data showed that the dehydration process begins between late June and early July in the Antarctic [Stone et al., 2001]. Kondo et al. [2000] examined ILAS HNO₃ data from the 1996/1997 Arctic winter and concluded that denitrification occurred only in air masses that had experienced temperatures below ice saturation. Irie et al. [2001] extended the analysis of the ILAS data and argued that significant denitrification occurred only in air parcels with temperatures below the NAT saturation point for at least four days after experiencing temperatures below the ice frost point. Irie et al. [2001] did not attribute the observed denitrification to the formation of ice particles, but rather to the homogeneous nucleation of NAT at temperatures near the ice frost point [Salcedo et al., 2001]; Tabazadeh et al. [2001] have suggested that such a homogeneous nucleation process may create a “polar freezing belt” within which denitrification takes place. Finally, Fahey et al. [2001] reported the detection of small ($\sim 2 \times 10^{-4} \text{ cm}^{-3}$) concentrations of very large (10-20 micron diameter) HNO₃-containing particles during SOLVE. Widespread denitrification was observed in this winter [Santee et al., 2000; Popp et al., 2001] with very little dehydration [e.g., Drdla et al., 2002]; still, the role of ice formation (possibly involving lee wave clouds) in nucleating these large particles is unclear [Fahey et al., 2001]. Although the observation of such large HNO₃-containing particles appears to have confirmed the notion that denitrification and dehydration are independent processes, it remains uncertain whether these particles were actually composed of NAT or what specific nucleation process led to their formation. Very large particles were observed primarily on

a single flight deep in the polar night [Fahey et al., 2001] during an exceptionally, and persistently, cold winter; the applicability of this finding to more typical Arctic winters is not clear. Nor is it known whether similar large particles form by similar processes in the Antarctic. Data from Aura will help resolve many of these issues. For example, use of EOS MLS and other Aura data in diagnosing PSC composition is discussed in section 2.2.3. In addition, we note that the study of Tabazadeh et al. [2000] was hampered by data missing either because of UARS yaw maneuvers or because of satellite and/or instrument problems (which interrupted measurements during the critical early-June period); however, because neither the CLAES instrument nor the UARS MLS 183-GHz radiometer used for stratospheric water measurements were operational during any other southern hemisphere winters, the 1992 Antarctic winter is the only period in the entire UARS data record that can be used to unambiguously identify the relationship between denitrification and dehydration. The better spatial and temporal coverage and the improved resolution and precision of the EOS MLS temperature, HNO_3 , and H_2O measurements, along with the high-resolution measurements of temperature, HNO_3 , H_2O , and aerosol extinction from HIRDLS, will allow the onset and extent of denitrification and dehydration in both hemispheres to be tracked over multiple years. For instance, the approach of Tabazadeh et al. [2000] can be applied, whereby a denitrified (dehydrated) region is defined as an area in which gas-phase HNO_3 (H_2O) abundances are significantly depleted without a corresponding enhancement in aerosol extinction, indicating the absence of solid HNO_3 -containing (water ice) PSCs. By relating the signature of solid PSCs in the extinction data to the behavior of the gas-phase species, the timing of these processes can be contrasted and compared between hemispheres and from year to year.

- **Is widespread severe denitrification necessary for massive ozone loss?** The interhemispheric differences in the morphology of wintertime HNO_3 may have profound implications, since the canonical view of stratospheric ozone chemistry has held that extensive denitrification is essential for massive ozone loss to occur [e.g., Solomon, 1999]. The persistence of large HNO_3 abundances throughout a typical Arctic winter has been thought to forestall severe ozone depletion [e.g., Brune et al., 1991; Salawitch et al., 1993]. Denitrification has been found to be a contributing factor in localized areas of ozone depletion during some cold Arctic winters [e.g., Rex et al., 1997; Waibel et al., 1999; Becker et al., 2000]. On the other hand, UARS MLS [e.g., Manney et al., 2002a] and other [e.g., Solomon, 1999] measurements have shown that ozone was substantially depleted throughout the Arctic vortex during the mid-1990s, even though widespread denitrification did not occur [e.g., Santee et al., 1999]. While local pockets of denitrified air too small to be detected in the satellite measurements may have existed, they could not have produced the degree of ozone loss detected over the entire vortex. Some modeling studies have now suggested that denitrification is not a prerequisite for extensive ozone loss in the Antarctic [Portmann et al., 1996; Brasseur et al., 1997]. In particular, Portmann et al. [1996] found that continued heterogeneous processing on cold sulfate aerosol particles during late winter and spring was the dominant mechanism in maintaining enhanced ClO, with denitrification playing a secondary role. Because sulfate aerosols are ubiquitous in the lower stratosphere (especially after volcanic eruptions), chlorine activation is not confined to isolated PSC events, expanding the spatial and temporal extent of potential ozone loss. In modeling three Arctic winters, Guirlet et al. [2000] found that low temperatures and consequent activation on PSCs in late February and March were a necessary condition for substantial ozone loss. Finally, Drdla and Schoeberl [2002] also found that ongoing PSC formation during February and early March in 1999/2000 allowed heterogeneous reactions to

reactivate chlorine. Unlike Portmann et al. [1996], however, Drdla and Schoeberl [2002] concluded that denitrification does significantly increase Arctic ozone depletion, but only at the very end of winter after the last PSC event; they suggested that because ozone loss in the Arctic is less intense than that in the Antarctic, it may be more sensitive to factors such as denitrification. Drdla and Schoeberl [2002] also note that the breakup of the vortex in mid-March may have limited the effect of denitrification in 2000; it may therefore play a bigger role in years in which the vortex remains intact longer into spring (or higher temperatures in late winter inhibit further heterogeneous processing). Aura measurements of temperature, aerosol extinction, HNO₃, ClO, HCl, ClONO₂, and O₃ (from EOS MLS and HIRDLS) will allow us to clarify the relative importance of denitrification and chlorine activation or reactivation on PSCs or sulfate aerosols in maintaining enhanced ClO into the spring, and thus facilitating ozone loss, in both hemispheres and over multiple years. These observations can be correlated, for example along large ensembles of trajectories, and compared to model results in the manner described by Drdla and Schoeberl [2002]. Thus Aura data may allow us to determine whether a critical level of denitrification exists beyond which severe ozone destruction commences.

- **How sensitive are these processes to changes in temperature and/or water vapor that may arise through climate change?** That denitrification apparently occurs in the absence of significant dehydration has important implications for forecasting Arctic ozone loss, since up to this time most large-scale chemical and climate models have assumed that these processes are coupled. In addition, the fact that denitrification takes place in a relatively warm month in the Antarctic raises concerns about the likelihood of its occurrence in the future Arctic lower stratosphere [e.g., Tabazadeh et al., 2000]. With Aura data we will be able to track the impact of any trends in temperature or water vapor (see section 2.2.1) on the onset, areal extent, or degree of denitrification and dehydration.

Some inherent limitations in satellite measurements must be borne in mind in addressing these issues. For example, Aura measurements may lack the vertical resolution needed to detect very narrow denitrified or dehydrated layers (although the ~1-km vertical resolution of the HIRDLS instrument may be adequate). They will also be insensitive to small, horizontally-localized pockets of denitrified or dehydrated air. On the other hand, even though the relationships between PSCs, denitrification, and ozone loss are arguably most critical for the future Arctic stratosphere, it may be more productive to investigate the details of these processes in the contemporary Antarctic, which is the only region of the stratosphere to experience massive denitrification so far. Because economic and logistical factors probably preclude a major field campaign in the Antarctic at this time, satellite measurements are the most feasible tool for studying the onset and areal extent of the denitrification and dehydration processes, and their interhemispheric and interannual variations. Data from aircraft and/or balloon flights in the Arctic, however, will be invaluable for investigating the links between small-scale PSC processes and denitrification on a scale detectable in Aura data.

2.2.4 Chlorine Activation and Deactivation

Because winter polar ozone loss is dominated by reactions involving ClO [e.g., Chipperfield and Pyle, 1998], a complete understanding of the chlorine budget and its seasonal evolution is critical to calculating chemical ozone destruction. A number of studies, however, have been unable to reconcile observed and modeled ozone loss rates, particularly in the Arctic.

Some key questions related to chlorine activation and deactivation that will be addressed with Aura data are:

- **Is our present understanding of chlorine activation and recovery in the winter polar lower stratosphere quantitatively correct?** Models have historically been unable to reproduce the peak ClO abundances measured by UARS MLS in the lower stratosphere. Results of 3D model comparisons by Lefèvre et al. [1994] and Lutman et al. [1997], both of which found the maximum ClO abundances measured by MLS to exceed the predicted values, may be partially explained by the use of an early version of UARS MLS ClO data, which had some significant known biases [Waters et al., 1996]. A similar underestimation of MLS measured values has been seen in subsequent studies using later versions of MLS data, however [e.g., Chipperfield et al., 1996; Ricaud et al., 1998; van den Broek et al., 2000; Danilin et al., 2000]. In these studies, models underpredicted the ClO measured by MLS in both hemispheres and in multiple years; in some cases the discrepancies between models and data were as large as 1.5 ppbv. Since this has important implications for the accurate calculation of ozone depletion, reassessing the level of agreement between modeled ClO peak values and those measured by EOS MLS will be a high priority. Also, although UARS MLS measured the global distribution of stratospheric ClO over annual cycles for nearly a decade (albeit with greatly reduced sampling frequency in later years), it never captured the main part of the chlorine recovery period in the southern hemisphere, and it is also possible that it failed to observe the maximum wintertime ClO abundances in the south in some or all of the years [Santee et al., in preparation]. Better spatial and temporal coverage for EOS MLS will remove these shortcomings. In addition, interpretation of the UARS MLS ClO data was complicated by an interval in the middle of every UARS yaw period during which measurements were made only in darkness. EOS MLS will provide better sampling for mapping ClO distributions: Because the Aura orbit will be sun-synchronous, EOS MLS observations at a given latitude on either the ascending or descending sides of the orbit will have the same local solar time throughout the mission, eliminating the intervals of nighttime-only measurements that arose during each UARS yaw cycle. These improvements will facilitate model comparisons.
- **Are the relative abundances of the main chlorine reservoir species, HCl and ClONO₂, adequately understood for pre- and post-PSC conditions and for different aerosol loadings? How sensitive is lower stratospheric chlorine partitioning to aerosol-mediated reactions at temperatures of 200-210K?** Simultaneous measurements, with uninterrupted coverage throughout the activation and recovery periods, of ClO and HCl (as well as averages of HOCl) from EOS MLS and ClONO₂ from HIRDLS will be indispensable for studies of chlorine partitioning. For example, these measurements will be combined to investigate the relative abundances of the main chlorine reservoir species at the beginning of winter prior to activation. This partitioning will then be compared with that measured during equivalent periods in the years both before and after the eruption of Mt. Pinatubo. [HCl normally exceeds ClONO₂ in air masses that have not undergone heterogeneous processing \[e.g., Zander et al., 1996; Michelsen et al., 1996; WMO, 2002\]. Following the eruption of Mt. Pinatubo, however, HCl concentrations in the lower stratosphere were found to be much smaller than models predicted \[Webster et al., 1994b\]. Significant increases in the fraction of total inorganic chlorine residing in HCl were observed in subsequent years in both in situ \[Webster et al., 1998, 2000\] and UARS HALOE \[Dessler et al., 1997; Webster et al., 2000\] data, and partially attributed to variations in the sulfate aerosol loading over this period \[Webster et al., 1998, 2000\].](#) While

this issue has not been completely resolved, the results of Webster et al. [1998, 2000] suggest that chlorine partitioning can be significantly perturbed under enhanced aerosol conditions when temperatures are only moderately low (below about 205 K). Aura instruments will provide measurements of all of the major species involved in this issue with good resolution and precision and will allow the partitioning of chlorine both before and after wintertime processing to be studied in detail.

- **Does the chlorine budget balance, or must additional, but currently 'unknown', chlorine species be accounted for?** The complement of measurements from Aura will be used to reassess this issue. Observations obtained during the 1999/2000 SOLVE/THESEO campaign, including the first atmospheric measurements of the dimer Cl_2O_2 [Stimpfle et al., in preparation??], initially appeared to indicate a discrepancy between the sum of the concentrations of the major inorganic chlorine species and the inorganic chlorine content estimated from measured organic source compounds (e.g., CFCs) [World Meteorological Organization, 2002]. In contrast, a number of previous studies had found good agreement between the inorganic and organic chlorine budgets in the polar lower stratosphere (although none had included measurements of Cl_2O_2) [WMO, 2002]. Subsequent refinements in data processing have mitigated the discrepancy suggested by the SOLVE observations [Rex et al., 2002b], but the possibility of 'missing' chlorine chemistry has not been eliminated entirely. The MLS measurements of ClO and HCl, together with HIRDLS measurements of ClONO₂, as well as CFC11 and 12, will contribute substantially to resolving this issue.
- **How important is ongoing processing or reprocessing on PSCs or sulfate aerosols in maintaining elevated levels of ClO into the springtime?** This topic is addressed in detail in section 2.2.3.
- **What loss mechanism is responsible for the rapid ozone depletion observed in mid-winter at high solar zenith angles?** The ability to quantitatively reproduce observed ozone losses for specific winters is essential if we are to predict the future course of polar ozone with any certainty. Both box models and 3D chemical transport models have been found to substantially underestimate the observed ozone loss during recent cold Arctic winters [e.g., Deniel et al., 1998; Becker et al., 2000, and references therein]. Some of these studies have suggested that the discrepancy is greater in early winter than in late winter. Similar conclusions were reached by Rex et al. [2002a], who found that unrealistically high active chlorine is required to explain ozone loss rates from March during several recent cold Arctic Januaries using standard reaction kinetics, even assuming complete activation of reactive chlorine. In contrast, Rex et al. [2002a] 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. These analyses suggest that January ozone loss occurs by an unknown process involving a photolytic step at high solar zenith angles [Rex et al., 2002a]. Indeed, one of the main conclusions to emerge from the Arctic Ozone Loss Workshop in Potsdam, Germany, in March 2002 was that “ozone losses in mid-winter (i.e., at high solar zenith angles) are real and cannot be fully explained by standard halogen photochemistry” [Rex et al., 2002b]. Previously, MacKenzie et al. [1996] demonstrated good agreement between observed and inferred loss rates based on UARS MLS O₃ and ClO data during the late winter in both the Arctic and the Antarctic, and Wu and Dessler [2001] also saw no evidence of significant discrepancies during Antarctic late winter using UARS MLS data. Given the apparent time-dependence of the disagreement between modeled and measured Arctic ozone loss and possibly better overall agreement in the Antarctic, it is of great

interest to determine the conditions over which the observed extent and degree of chlorine activation, and thus ozone loss, can be accurately modeled. Aura will be in an excellent position to address this issue. A deficiency in the study of Rex et al. [2002a] was the inability to compare Match estimates of ozone loss to modeled loss based on observed ClO in any of the cold Januaries or in February or March of any year other than 1996; MLS ClO data were either unavailable entirely or were too infrequent to be of use in constraining model runs along the Match trajectories. Better temporal coverage, precision (by a factor of about two), and spatial resolution of the EOS MLS ClO measurements will substantially improve similar studies in the future. In particular, measurements of ClO abundances and ozone loss will be obtained at high solar zenith angles in mid-winter. Comparison of ozone loss rates measured directly by Aura with those calculated from models constrained by Aura measurements, perhaps along large ensembles of trajectories, will improve our understanding of the processes responsible for mid-winter ozone depletion.

- **Will bromine become relatively more important in polar ozone destruction in the future? Does wintertime heterogeneous chemistry produce OH enhancements?** EOS MLS will measure stratospheric abundances of species such as BrO, HOCl, and OH. Their retrieval from the MLS radiances will be challenging, however, and it is expected that their precision will be lower than that of most other MLS data products. As a result, averaging over several days to weeks may be necessary to obtain useful precision for scientific studies. Because bromine abundances are still increasing, and because abundances of chlorine source gases are expected to decrease in coming years at rates faster than those of bromine source gases [World Meteorological Organization, 2002], the role of bromine in ozone depletion may grow in importance in the future (although ozone recovery is predicted to occur as stratospheric chlorine levels subside, no matter what happens to the bromine loading [Chipperfield and Pyle, 1998]). EOS MLS measurements of averaged BrO will be useful in broadly following these trends. In addition, model simulations conducted to test the EOS MLS retrieval algorithms currently in development have indicated the occurrence of localized OH enhancements in the lower stratosphere under Arctic winter conditions. They appear to be caused by heterogeneous chemistry producing HOCl ($\text{ClONO}_2 + \text{H}_2\text{O} \rightarrow \text{HNO}_3 + \text{HOCl}$), which then photolyzes to yield OH. The precision of the MLS OH and HOCl measurements will be adequate to test this model prediction.

A handicap in several of these studies is that Aura provides no measurements of Cl_2O_2 . Because ClO and Cl_2O_2 can contain comparable amounts of chlorine during periods of maximum chlorine activation, knowledge of the dimer is crucial for evaluating the overall chlorine budget and following the evolution of active chlorine over the winter. Therefore, as with many of the polar processing issues, the unresolved questions highlighted in this subsection would benefit from a unified approach combining satellite data with measurements obtained using different observational strategies, in particular in situ measurements.

2.2.5 Chemical Ozone Loss in Polar Winter

Quantifying chemical ozone loss is central to addressing the overall question "is the ozone layer changing as expected?" in the context of the Earth's polar stratosphere. Three key questions that will be addressed using empirical ozone loss studies based on Aura data are:

- **How well can we quantify Arctic ozone loss amounts throughout the winter?**
- **How well can we quantify Antarctic ozone loss, including the timing and morphology of the onset of loss in mid-winter, and the extent and magnitude of ozone loss in the lowest part of the vortex and subvortex region?**
- **What are the relative contributions of dynamical/transport processes and chemical processes to observed long-term ozone changes, both in the vortices and the entire high-latitude winter region?**

Results and discussion at the recent Arctic Ozone Loss Workshop (Potsdam, Germany, 4-6 March 2002) revealed that, while different methods and datasets give qualitatively similar results in Arctic ozone loss studies, including the same character of interannual variability, we are still far from detailed quantitative agreement, especially in the vertical structure of ozone loss amounts, in the amount of loss in column ozone, and in quantifying the relatively small ozone losses in midwinter. Recent studies [Chipperfield and Jones, 1999; Andersen and Knudsen, 2002; Manney et al., submitted] have attempted to determine the relative contributions of dynamical/transport processes and chemical processes using model simulations and data analyses. These studies, however, are all severely hampered by limited temporal, horizontal and/or vertical coverage of available data and very large uncertainties in current estimates of Arctic ozone loss amounts. Empirical ozone loss studies attempt to quantify ozone loss by combining models and observations using several methods [see, e.g., Harris, et al., 2002; WMO, 2002]. These methods fall into four broad categories:

- (1) Match techniques, or variations thereof, wherein observations of the same air mass at different times are matched using trajectory calculations, and the ozone changes are used to estimate chemical loss [e.g., Rex et al., 2002, and references therein; Terao et al., 2002].
- (2) Vortex-averaged methods, wherein vortex averages derived from observations are used in conjunction with calculations of vortex-averaged descent to estimate chemical ozone loss [e.g., Bevilacqua et al., 1997; Knudsen et al., 1998; Rex et al., 1998; Lucic et al., 1999].
- (3) Transport model methods, wherein a 3D transport model is used to simulate the evolution of ozone in the absence of chemical loss, and those results are compared with the observed evolution to estimate chemical loss [e.g., Manney et al., 1995, 2002; Deniel et al., 1998; Goutail et al., 1999; Hoppel et al., 2002; and references therein].
- (4) Approaches using the relation of ozone to long-lived tracers, wherein it is assumed that changes in the ozone/tracer correlation over the winter are related to chemical ozone loss [e.g., Müller, et al., 1996, 1997; Richard et al., 2001; Salawitch et al., 2002; and references therein].

Methods (1)-(3) are broadly similar in involving explicit transport calculations to estimate the behavior of ozone in the absence of chemical loss. The transport model method (3) was

originated by Manney et al. [1995], using a trajectory-based "Lagrangian transport" model and UARS MLS data; Manney et al. [2002] used this method to compare Arctic ozone loss in all the winters observed by UARS MLS and noted several ways in which these studies would be greatly improved by using data from the EOS Aura instruments. Continuous daily coverage throughout the winter and spring will be invaluable for calculations of cumulative loss and of the effects of dilution on midlatitude ozone loss at vortex breakup (see section 2.1.5.2). Also important to improving these studies is the extension of ozone measurements down near or across the tropopause, covering all the levels at which ozone loss is expected; UARS MLS estimates of column ozone loss were limited by availability of ozone down only to 100 hPa, when ozone loss is frequently observed below this level [e.g., Goutail et al., 1999]. Uncertainties in all studies involving transport models are currently very large, but can be substantially reduced when hemispheric or global long-lived tracer data are available to validate the transport calculations. Better horizontal and/or vertical resolution from EOS MLS measurements and better precision will also help to more precisely define observed changes and their morphology. Method (3) will be used (as done by Manney et al. [2002] with UARS data) with EOS MLS ozone data, and validated with EOS MLS N₂O data to produce more precise, complete estimates of cumulative Arctic ozone loss throughout the winters observed. It is anticipated that method (3) will also be used with conventional transport models and EOS MLS data, and that studies will be done using methods (1) and (2) with MLS data. This will allow comparison of results from different methods for the same time periods and datasets, greatly enhancing our ability to quantify the uncertainties in estimates of Arctic ozone loss. Empirical estimates of the vertical distribution of ozone loss from the above studies will be used to derive ozone losses in column, and this will be used (with methods similar to those of Andersen and Knudsen [2002] and Manney et al., submitted) along with dynamical models to study the relative importance of dynamical and chemical processes in Arctic ozone changes, on day-to-day, seasonal, and interannual time scales.

Very serious questions remain about the validity of method (4) in the presence of descent and mixing, which may affect tracer correlations in ways that mimic chemical loss [e.g., Michelsen et al., 1998; Plumb et al., 2000]. Descent and mixing in the Arctic are expected to show large interannual variability related to the strength and evolution of the polar vortex. While some studies [e.g., Richard et al., 2001; Muller et al., 2001; Salawitch et al., 2002] have attempted to examine and/or account for these effects, none of these efforts has been completely satisfactory, largely because of the difficulty of realistically estimating these effects without season-long records of hemispheric data for multiple long-lived tracers. Preliminary attempts to look at the effects of realistic precision estimates on tracer correlations (e.g., Figure 2.2.5-1) show promise that we will be able to obtain useful tracer correlations from EOS MLS and HIRDLS data. In addition to using the suite of long-lived tracers from HIRDLS to examine the effects of descent and mixing on ozone/tracer correlations, and hence to determine with much greater confidence the validity and limitations of the tracer correlation approach for estimating ozone loss, we plan to use correlations of MLS ozone and N₂O to derive estimates of ozone loss and compare these with other methods. Not only will this help to assess the validity of the tracer correlation method, but all of these empirical ozone loss studies promise to greatly reduce the now unacceptably large uncertainties in quantitative estimates of Arctic ozone loss. Only by substantially reducing these uncertainties can we accurately discern the amount of interannual variability and possible trends in Arctic ozone loss, and relate them to changes in vortex conditions (e.g., temperature, H₂O trends).

Figure 2.2.5-1, now located at end after references, to be inserted somewhere here.

Manney et al. [1995] used the Lagrangian transport model method to show that ozone loss in the Antarctic began in June. This was subsequently confirmed by Roscoe et al. [1997] and Lee et al. [2000], who showed that loss began first near the polar vortex edge where most sunlight was received. Lee et al. [2001] examined mixing within the Antarctic polar vortex and its effects on the morphology and amount of Antarctic ozone loss. Lee et al. [2002] modeled ozone loss in the Antarctic vortex and subvortex, and showed that a substantial amount of loss occurs in the subvortex region, below where the vortex is most laterally confined, with important implications for mixing with midlatitudes (section 2.1.5.2); they found their results to be broadly consistent with sparse ozonesonde data. MLS (and HIRDLS) global daily ozone observations throughout the winter, extending through most or all of the subvortex region, will be used - along with the methods for determining chemical loss from observations discussed above - to examine in much more detail the morphology and onset of ozone loss, and the extent of ozone loss in the subvortex region. These data, with other constituents measured by Aura instruments, will also be used for better model initialization for studies similar to that of Lee et al. [2002], and for more detailed model/data comparisons.

2.3 Stratospheric H₂O

2.3.1 Dehydration of the stratosphere

One of the most interesting questions in atmospheric sciences today is how and why the humidity of the stratosphere is changing. Recent measurements have shown that the humidity of the stratosphere has been increasing for the last few decades [Oltmans et al., 2000; Rosenlof et al., 2001], with important implications for stratospheric ozone [Kirk-Davidoff et al., 1999; Dvortsov and Solomon, 2001] as well as radiative forcing of the climate system [Forster and Shine, 1999; Smith et al., 2001].

A plausible hypothesis is that climate change increases the amount of water vapor entering the stratosphere. Since water plays an important role in stratospheric ozone chemistry, increases in stratospheric humidity lead to decreases in stratospheric ozone. Such decreases might then feed back onto the troposphere, causes further change in water entering the stratosphere. An understanding of the mechanisms regulating stratospheric humidity is required in order to evaluate this hypothesis.

There is general agreement that air is transported into the Tropical Tropopause Layer (TTL), a transition zone between the troposphere and stratosphere, directly from the planetary boundary layer (PBL) by convection. While some air detrains from the convective regions above the conventionally-defined tropopause, it appears that most detrainment is below this level. Below the tropopause there is net radiative heating, and air slowly ascends across the tropopause into the stratosphere [e.g., Sherwood and Dessler, 2001]. Air passing through the bottom of the TTL in convective updrafts contains several tens of ppmv of water in vapor and condensate, while air slowly rising through the top of the TTL and into the stratosphere contains (on average) 3.85 ppmv of water [Dessler and Kim, 1999]. Clearly, dehydration mechanisms in the TTL remove the majority of the water that enters the stratosphere.

Over the past two decades, two competing theories for this dehydration mechanism have emerged. The first, which we call the “convective dehydration” theory, posits that air emerges from convection fully dehydrated (on average) to stratospheric values [e.g., Sherwood and

Dessler, 2001]. The second, which we call the “gradual dehydration” theory, posits that air is dehydrated after detrainment from convection by repeated exposures to episodic cold events as it slowly ascends into the stratosphere [e.g., Holton and Gettelman, 2001].

Some key questions related to dehydration in the TTL that will be addressed with MLS Aura data include those itemized below.

- **Does convection hydrate or dehydrate the TTL?** In other words, on a given potential-temperature surface in the TTL, is the air that detrains from convection moister or drier than air that is slowly ascending from below? MLS measurements of the three-dimensional structures of water vapor and other trace gases around the tropical tropopause will help us answer this question. Because low ozone (high CO) indicates air that has been recently transported from the planetary boundary layer via convection, coincidence between regions of low water vapor and low ozone (high CO) will suggest that the convective dehydration is the dominant mechanism. Lack of such a correlation will support the gradual dehydration theory. Of particular interest will be the analysis of measurements at 100 hPa, where UARS MLS was not able to measure water vapor.
- **What role does the Asian monsoon play in regulating the humidity of the stratosphere?** The Asian monsoon system has come under increasing scrutiny as a major player in the water budget near the tropopause. Moistening of the TTL and mid-latitude lowermost stratosphere regions appears clearly in observations [e.g., Randel et al., 2000]. But it is not clear how the monsoon affects air entering the stratospheric overworld (potential temperatures > 380 K). Mote et al. [1996] examined this using two-dimensional back trajectories, but their results were inconclusive. Maps of the three-dimensional fields of water vapor, O₃, and other tracers, in combination with transport studies driven by NASA’s Data Assimilation Office, will be used to trace the motion of water vapor to determine the fate of monsoon water vapor.
- **What is the role of “thin cirrus” clouds in the TTL?** The role of thin cirrus clouds (clouds with visible and near-infrared optical depths much less than unity) in the water budget of the TTL is uncertain. The combination of Aura MLS measurements of water vapor with Aqua MODIS [Dessler and Yang, 2002] and Aura HIRDLS measurements of thin cirrus, along with temperatures from the AIRS/AMSU/HSB suite (as well as from MLS and HIRDLS), should help gain insight into the role of these clouds. In particular, we will be able to examine from a broad viewpoint whether thin cirrus formation is slaved to instantaneous temperature or whether other factors such as the history of the air mass are important in determining whether thin cirrus clouds form. The evolution of the humidity fields will be correlated with the appearance and disappearance of the thin clouds and also with environmental conditions. This should yield critical constraints on their role as a dehydrator. In this way, we hope to infer how much water is returned to the vapor phase versus how much is precipitated in the clouds.

2.3.2 Variations in stratospheric H₂O

[This section is, for now, being kept as a placeholder in case we want to collect together the various issues associated with unexplained (primarily long-term) variations in stratospheric H₂O.]

2.4 Volcanic Effects on Stratospheric Ozone

Explosive, sulfur-rich volcanic eruptions can cause significant stratospheric perturbations. In fact, stratospheric aerosol variability over the past 25 years has been dominated by the effects of episodic volcanic eruptions [World Meteorological Organization, 2002], and 30% of the last 150 years have been characterized by volcanic clouds as optically thick as Arctic PSCs [Tabazadeh et al., 2002]. Such an eruption within the next few decades, when stratospheric chlorine loading is still high, could have profound consequences for the ozone layer. This is particularly true in the Arctic, where changes in temperature and/or water vapor associated with climate change (see section 2.2.1) could make stratospheric ozone even more vulnerable to volcanic perturbations.

In the event of a major volcanic eruption during the Aura mission, objectives will include trying to answer (or help answer) the key questions listed below.

- **Can volcanoes have a *direct* effect on stratospheric chlorine levels?** Major volcanic eruptions, such as El Chichon in April 1982 and Mt. Pinatubo in June 1991, can inject large amounts of H₂O, SO₂, and HCl into the stratosphere. Following the eruption of El Chichon, Mankin and Coffey [1984] reported a 40% increase in HCl total column, which they attributed to direct injection by the volcano. In contrast, however, post-Pinatubo observations showed little or no increase in HCl compared to pre-eruption levels [Mankin et al., 1992]. The latter measurements are consistent with the modeling results of Tabazadeh and Turco [1993] indicating that the majority of emitted HCl is rapidly removed from eruption plumes through dissolution in water droplets followed by rainout. Tabazadeh and Turco [1993] concluded that, since this scavenging process can quickly reduce HCl concentrations by as much as four orders of magnitude, substantial stratospheric injection of chlorine by volcanoes is precluded. On the basis of these results, Tabazadeh and Turco [1993] argued that direct HCl injection by El Chichon could have accounted for only a small fraction of the observed stratospheric chlorine increase. With the HCl measurements from EOS MLS, we will be able to evaluate the magnitude of any direct perturbation to stratospheric chlorine levels after a major eruption.
- **How does the enhanced aerosol loading resulting from volcanic eruptions affect stratospheric reactive nitrogen and chlorine concentrations at mid and high latitudes?** Following volcanic eruptions, the SO₂ injected into the stratosphere is rapidly converted into sulfate aerosol [McKeen et al., 1984], increasing the aerosol surface area density in the lower stratosphere by as much as two orders of magnitude [Thomason et al., 1997]. In the case of tropical volcanoes such as El Chichon and Mt. Pinatubo, poleward dispersal of volcanic aerosol depends strongly on the intensity of planetary wave activity and the phase of the QBO [Trepte et al., 1993]; nevertheless, highly enhanced aerosol surface area density was observed in the lower stratospheric midlatitudes and polar regions in both hemispheres in the years following these volcanoes [Thomason et al., 1997]. Increases in aerosol surface area facilitate heterogeneous reactions, notably the hydrolysis of N₂O₅ and, to a lesser extent, ClONO₂. Because N₂O₅ hydrolysis is not temperature-dependent (although ClONO₂ hydrolysis is), globally-distributed volcanic aerosols can vastly extend the latitudinal, altitudinal, and temporal ranges over which heterogeneous processes can alter the chemical partitioning of the stratosphere [Solomon, 1999, and references therein]. Several modeling studies [e.g., Hofmann and Solomon, 1989; Brasseur and Granier, 1992] have demonstrated the potential for the N₂O₅ reaction to cause substantial increases in HNO₃ abundances after large volcanic eruptions. Profiles of HNO₃ measured at both

northern [Webster et al., 1994a] and southern [Rinsland et al., 1994] midlatitudes after Pinatubo compared much better with models that include this reaction. Other HNO₃ datasets indicated a gradual decline in HNO₃ concentrations at both mid and high latitudes as the stratosphere recovered from Pinatubo-induced HNO₃ enhancement [Koike et al., 1994; David et al., 1994; Slusser et al., 1998; Kumer et al., 1996; Randel et al., 1999]. Consistent with the changes in HNO₃, significant reductions in midlatitude N₂O₅ [Rinsland et al., 1994] and NO₂ [e.g., Johnston et al., 1992; Mills et al., 1993] were also observed. Decreased NO₂ limits conversion of ClO to ClONO₂, and indeed [comparison of ER-2 measurements taken both](#) before and after the eruption of Mt. Pinatubo revealed a significant [post-eruption](#) enhancement in midlatitude ClO [Avallone et al., 1993b]; balloon-borne observations in a volcanic cloud over Greenland (outside the vortex) in March 1992 also indicated suppressed concentrations of NO and enhanced concentrations of ClO [Dessler et al., 1993]. UARS MLS measurements showed a decay in ClO of a factor of 2-4 over the period 1992 to 1997 at low and midlatitudes, consistent with a relaxation to nonperturbed conditions [Froidevaux et al., 2000]. Anomalously-high column abundances of OCIO observed in the [Arctic autumn of 1991 \[Perner et al., 1994\]](#) and the Antarctic autumn of 1992 [Solomon et al., 1993], at times when PSCs were extremely unlikely to have formed, also provided strong evidence of heterogeneous chlorine activation on Pinatubo aerosols. Finally, concentrations of HCl in the midlatitude lower stratosphere were found to be much lower in 1993 than models predicted [Webster et al., 1994b]. Significant increases in the fraction of total inorganic chlorine residing in HCl were observed in subsequent years in both in situ [Webster et al., 1998, 2000] and UARS HALOE [Dessler et al., 1997; Webster et al., 2000] data. Using a photochemical model incorporating recent measurements of heterogeneous reaction rates, Webster et al. [1998, 2000] have shown that this increase in the relative abundance of HCl can be partially attributed to the reduction in the sulfate aerosol loading over this period [Webster et al., 1998, 2000]. Although the model still underestimates the growth rate of the fraction of chlorine in HCl by more than a factor of two, the results of Webster et al. [1998, 2000] suggest that chlorine partitioning can be significantly altered under enhanced aerosol conditions when temperatures are only moderately low (below about 205K). Thus many independent measurements have confirmed that stratospheric nitrogen and chlorine partitioning can become highly perturbed after major volcanic eruptions. With Aura, investigations of heterogeneous chemical processing on sulfate aerosol will be facilitated by having nearly simultaneous measurements of HNO₃ (MLS and HIRDLS), N₂O₅ (HIRDLS), NO₂ (HIRDLS, and column amounts from OMI), ClO (MLS), HCl (MLS), ClONO₂ (HIRDLS), and also column OCIO (OMI), [although the lower stratospheric data from HIRDLS and OMI may be compromised under conditions of heavy volcanic aerosol loading](#).

- **What effect does volcanically-enhanced sulfate aerosol have on PSC formation and denitrification?** Calculations indicate that HNO₃ uptake into ternary solutions occurs at higher temperatures, and is larger at a given temperature, under conditions of enhanced sulfate aerosol [Carslaw et al., 1994; Tabazadeh et al., 1994]. The sulfate-rich composition of ternary droplets under volcanic conditions hampers formation of crystalline PSCs [Tabazadeh et al., 2002]. Observational evidence in the wake of the Mt. Pinatubo eruption supports these calculations. For example, measurements of aerosol concentration from balloonborne particle counters [Deshler et al. 1994] showed that PSCs over McMurdo in late winter 1992 were more frequent, were concentrated at slightly lower altitudes corresponding to the peak in the volcanic layer, and were characterized by smaller

particles, than those in previous nonvolcanic years. Lidar measurements at the South Pole [Cacciani et al., 1997] showed that PSC activity decreased in intensity and shifted downward in altitude in concert with the volcanic aerosol layer between 1992 and 1993 as the stratosphere began to revert to normal conditions. David et al. [1998] compiled a 5-year climatology of lidar measurements from Dumont d'Urville and similarly found that following the Pinatubo eruption most PSCs tended to form at lower altitudes around the peak in the sulfuric acid aerosols, and they also noted increased liquid PSC formation at higher temperatures and reduced solid PSC formation under volcanic conditions. Comparison of UARS MLS gas-phase HNO₃ measurements from several Antarctic winters [Santee et al., 1998] also indicated that HNO₃ uptake into ternary solutions occurred at higher temperatures, and the transition from liquid ternary droplets to solid particles was delayed, under volcanic conditions. Although some evidence for a change in PSC character under volcanic conditions has also been seen in backscatter profiles from Alert [Rosen et al., 1994], in general the impact of enhanced aerosol loading on PSC formation in the Arctic has not been extensively documented. The changes in PSC formation arising from increased sulfate aerosol loading may limit the effectiveness of PSC particles in causing denitrification. Measurements of temperature, H₂O, HNO₃ (from both MLS and HIRDLS) and aerosol extinction (from HIRDLS) will be invaluable for assessing the differences in PSC character and denitrification between volcanically perturbed and quiescent periods, should the opportunity arise.

- **How do volcanoes affect stratospheric ozone?** Local heating associated with the increased aerosol loading from Mt. Pinatubo enhanced upward motions in the tropics, carrying low-ozone air to higher altitudes and leading to a reduction in total column ozone in the tropics for months after the eruption [Kinne et al., 1992; Brasseur and Granier, 1992; Schoeberl et al., 1993]. In addition to decreases in tropical ozone induced by changes in atmospheric circulation, significant chemical perturbations at mid and high latitudes are associated with the enhanced aerosol loading. In 1992, Total Ozone Mapping Spectrometer (TOMS) measurements showed for the first time a simultaneous sustained decrease in column ozone over a wide latitude range in both hemispheres [Gleason et al., 1993]. Significant ozone losses observed at midlatitudes have been attributed to the increase in aerosol loading, and its attendant chemical effects, following major volcanic eruptions [e.g., Hofmann and Solomon, 1989; Solomon et al., 1996; Solomon et al., 1998]. Hofmann and Oltmans [1993] showed that unusual Antarctic ozone depletion observed in 1992 at altitudes below PSC formation levels likely resulted from chemical processing by the Pinatubo aerosol. Model simulations by Portmann et al. [1996] have shown that the rapid deepening of the Antarctic ozone hole in the early 1980s, and the large ozone declines observed in the early 1990s, were probably associated with the increase in aerosol surface area arising from volcanic eruptions, in combination with increasing stratospheric chlorine abundances. Some discrepancies between model results and measurements remain unexplained, however. For example, models that include heterogeneous reactions on volcanically-enhanced aerosol suggest that post-eruption ozone depletion is hemispherically symmetric [WMO, 2002], whereas observations indicate different ozone behavior in the two hemispheres following Mt. Pinatubo. Area-weighted averages in the northern and southern hemispheres showed that most of the 1992-1993 ozone losses observed by TOMS occurred in the northern hemisphere [Herman and Larko, 1994]. With the ozone measurements from EOS MLS, HIRDLS, and OMI, we will be able to quantify volcanically-induced changes in ozone at all latitudes.

- **Will climate change make the stratosphere more vulnerable to ozone depletion after a volcanic eruption?** A major volcanic eruption during the Aura timeframe could have a much greater impact on stratospheric ozone than did the eruptions of previous eras. Tabazadeh et al. [2002] have used a coupled chemistry-microphysics trajectory model to show that severe Arctic ozone loss can occur in a volcanic year in which temperatures are similar to those of the relatively cold 1990s and the vortex remains intact into early spring. The magnitude of column ozone loss calculated for the conditions of the (unusually cold) 1999/2000 Arctic winter in an assumed volcanic state was found to be comparable to that projected for a future colder nonvolcanic stratosphere (when denitrification was calculated to have a substantial impact). Possible cooling of the stratosphere, and increases in H₂O, may exacerbate ozone depletion even under background aerosol conditions (see sections 2.1 and 2.2), and will likely have a still greater effect under enhanced aerosol conditions. Thus the extent to which a volcanic eruption might influence ozone recovery in the next decade or so, when stratospheric chlorine loading will be considerably higher than when Mt. Pinatubo erupted, is a topic of great concern which - in the event of a major eruption - will be addressed using the combined Aura measurement suite.

3 Tropospheric Ozone and Pollution

MLS upper tropospheric measurements will contribute to helping answer the NASA Earth Science Enterprise key question,

What are the effects of regional pollution on the global atmosphere, and the effects of global chemical and climate changes on regional air quality?

Unlike stratospheric ozone, which has beneficial properties for the environment below, tropospheric ozone is a pollutant and contributor to urban smog. Tropospheric ozone has increased since pre-industrial times, but the increase seems to have abated during the last decade [World Meteorological Organization, 1998]; however, tropospheric datasets are sparse and not all consistent. Tropospheric ozone is also important as a source of OH, which is produced by reaction between H₂O and O(¹D) from O₃ photolysis. OH in turn regulates the oxidative capacity of the troposphere, a measure of the capability of the atmosphere to “cleanse itself” of many polluting and greenhouse gases [WMO, 1998]. Tropospheric ozone radiative effects on climate change may be significant and are most important in the upper troposphere [IPCC, 1996].

Tropospheric pollution and the global effects of regional pollution have received increased attention in the past decade, with many insightful results from various aircraft campaigns such as TRACE-A [Journal of Geophysical Research 101, No. D19, 1996]; see also WMO [1998]. The oxidation of CO and other hydrocarbons in the presence of NO_x leads to ozone production in the troposphere; there are anthropogenic and natural sources for these ozone precursors. Aura will provide a major step forward in tropospheric observations with satellite measurements of many tropospheric species by TES. The MLS upper tropospheric measurements will complement the TES, with certain scientific objectives described below.

The upper tropospheric region measured directly by MLS has not been studied as extensively as the middle and lower troposphere. It is characterized by relatively rapid mixing, but exhibits signatures of lower tropospheric chemical emissions, gradients, and

inhomogeneities, for example from rapid convection and long-range transport processes. Measurements in this region can therefore help us understand the impact of regional effects on the global atmosphere. The ozone budget in this region will be affected by transport from below as well as from the stratosphere, in addition to local photochemistry involving ozone precursors transported to this region. A better set of measurements in the upper troposphere can lead to improved constraints on pollution sources and transport processes (as prescribed in chemical/transport models), in addition to improved constraints regarding the radiative importance of ozone in this region. Since satellite tropospheric measurements are more challenging and newer than satellite stratospheric measurements, and since a broad suite of measurements is needed for an understanding of the troposphere and its variability, supplementary and complementary measurements from aircraft missions (along with balloons and ground-based networks) will be very important for achieving proper tropospheric data validation as well as common scientific objectives.

The MLS upper tropospheric measurements, made with varying degrees of precision, include O₃, CO, CH₃CN, HCN, H₂O, and temperature. H₂O, temperature and O₃ will have sufficient precision for individual profiles to be useful in most situations. Biweekly or monthly maps will probably be the most useful product for CO, CH₃CN and HCN under normal situations, but single profile measurements will be valuable when the abundances of these species are substantially enhanced, such as can occur in biomass burning episodes. Lifetime estimates in the upper troposphere are of order 3 months for O₃ [Jacob et al., 1996], 2 months for CO, and 5 months for CH₃CN, and HCN [e.g., Singh et al., 2002], so that monthly maps of these species will be valuable. The primary MLS scientific objectives involving tropospheric H₂O (and cloud ice) data are covered separately in sections 2.3 and 4.1, but these measurements also give indications of convection into the upper troposphere, which is relevant to some of the objectives discussed here. A feature of MLS for studying the upper troposphere is that its measurements can be made in the presence of - and through - cirrus - and therefore, MLS measurements of O₃, H₂O, and CO will supplement those of TES and HIRDLS in regions of cirrus. MLS is also the only instrument to provide measurements of CH₃CN and HCN, which are good tracers of biomass burning [e.g., Andreae et al., 2001].

The following are some tropospheric pollution-related scientific objectives to be addressed with MLS measurements in the troposphere.

- **Describing and quantifying the relation between biomass burning and global upper tropospheric pollution.** Biomass burning, particularly in Africa and South America during the dry season, has been shown to be a significant contributor to "wave 1" tropospheric column ozone enhancements over the South Atlantic [e.g., Fishman et al., 1990]. However, biomass burning may be a fairly small (< 15%) contributor to O₃ (and NO_x) in the overall global upper troposphere [Galanter et al., 2000]. CO enhancements in the Pacific upper troposphere have been observed from commercial aircraft and shown to relate to biomass burning seasonal patterns [Matsueda et al., 1999]; in addition, correlations between these CO enhancements (and their interannual variability) and El Nino-related dryness and convection events were demonstrated. Direct linkage between biomass burning and upper tropospheric enhancements in related pollutants, including CO and CH₃CN, via transport by deep convection has been obtained [e.g., Andreae et al., 2001]. A huge (~30x) enhancement in lower stratospheric CH₃CN was detected by UARS MLS in August 1992 and traced to forest fire pollution injected into the stratosphere by an intense thunderstorm as the most likely source [Livesey et al., in preparation, 2003]. HCN has also been observed in unusually high amounts in the upper troposphere [Rinsland et al., 1998;

Zhao et al., 2000, Rinsland et al., 2001]. An objective of EOS MLS, with its improved capability, is to quantify the extent and evolution of such events in the upper troposphere (as well as the lower stratosphere) and their effect on the global atmosphere. Other dynamical mechanisms (e.g., frontal uplift) have also been shown to bring pollution into the upper troposphere [Yienger et al., 2000, Bey et al., 2001]; increased information about these mechanisms will also be part of the envisioned studies.

- **Improving knowledge and understanding of global tropospheric column ozone.** Stratospheric column ozone from MLS will be used in combination with OMI total ozone column to produce daily maps of tropospheric ozone residual (TOR). This technique was initially demonstrated by Fishman et al. [1990] using seasonal averages of TOMS and SAGE ozone data, and has been pursued by many others using TOMS data alone (mainly possible/demonstrated in the tropics). UARS MLS and HALOE data have been used for similar purposes with TOMS, but are limited by the data quality for the lowermost stratosphere, especially outside the tropics. The improvements in EOS MLS sensitivity to lower stratospheric ozone should largely remove this limitation. The OMI-MLS TOR column product will be compared to the tropospheric ozone columns measured directly by TES. In addition to validation of the TES measurement, the OMI-MLS TOR daily product will supplement the TES measurements, which have an expected 50% duty cycle. We expect similar analyses using OMI-HIRDLS TOR, which will have much better cross-track spatial resolution than OMI-MLS. The three Aura measurements of tropospheric column ozone (TES direct, OMI-MLS TOR, OMI-HIRDLS TOR) will provide a valuable combined dataset for understanding the quality of the various individual measurements and for improving our understanding of tropospheric ozone and its space-time variations.
- **Describing and quantifying the relation between surface pollution and the upper troposphere.** The MLS measurements of CO can provide a general measure of pollution in the upper troposphere, as this gas arises from fossil fuel combustion, methane and non-methane hydrocarbon oxidation, as well as from biomass burning. CO is an ozone precursor and is often positively correlated with ozone [e.g., Watson et al., 1990]. However, high humidity regions (e.g., over southeast Asia, Indonesia, and South America) tend to show lower O₃ abundances in the upper troposphere because of increased HO_x-related loss and vertical transport [Thouret et al., 1998]. Also, stratospheric intrusions will cause a negative correlation between O₃ and CO. Observing such correlations on a global scale as a function of time, even with the limitation of the upper tropospheric data from MLS alone, should help provide some constraints regarding the sources of ozone. Furthermore, the MLS CO and CH₃CN data in combination should provide some discrimination for the different sources of CO, [since CO has both biomass burning and industrial sources \(but CH₃CN enhancements are likely to be biomass burning related\)](#). Comparisons with aircraft data will be useful for validation, for related measurements (such as C₂H₂, another biomass burning tracer, or tracers of urban pollution, such as C₂H₆, C₂Cl₄), and for connecting localized effects to the more global satellite perspective. The multiyear MLS observations of upper tropospheric CO, O₃, H₂O, CH₃CN and HCN should contribute to a better understanding of the influence of various sources on global pollution in the upper troposphere. Simultaneous data gathering from different satellite instruments and various aircraft missions is important for both scientific objectives and validation, especially because of (1) satellite limitations in terms of signal-to-noise, vertical resolution, and horizontal sampling, and (2) aircraft limitations such as horizontal, vertical, and temporal coverage. A more comprehensive dataset will allow for an easier extension

of the analysis range into the lower troposphere. Correlations of the MLS O₃, CO, CH₃CN and HCN measurements will provide useful information regarding the sources of observed O₃ enhancements, biomass burning in particular. Trajectory calculations will be used for tracing the enhancements back to their sources. Such studies will involve use of other Aura products (e.g., high NO₂ columns from OMI, for urban pollution sources of O₃; TES-derived enhancements in O₃, CO, CH₄; aerosol from OMI).

- **Improving knowledge and understanding of the global upper tropospheric ozone distribution and its budget.** The MLS upper tropospheric ozone measurements will be used, along with similar measurements from TES and HIRDLS, to produce a more detailed global description of the upper tropospheric ozone field, its budget, and its variations than has previously been possible. Previous models of aircraft data have shown that both photochemical production and mixing processes can be important for ozone production in the middle/upper troposphere [e.g., Wild et al., 1996]. However, some processes such as convection and transport are not that well represented in tropospheric chemical/transport models [WMO, 1998]. The addition of tropospheric MLS and Aura data on a global basis should dramatically increase the tropospheric database and the number of tropospheric model constraints, especially for ozone (where the best MLS signal-to-noise is expected). The MLS measurements will be particularly valuable in the presence of cirrus that degrade TES and HIRDLS measurements more than they do those of MLS. Stratosphere-troposphere exchange is a non-negligible ozone source for the upper troposphere [Lelieveld and Dentener, 2000], and improvements in model parameterizations of this process should be possible from accurate global profiles of ozone and other species in the upper troposphere and lower stratosphere. The MLS measurements in the presence of cirrus will be an especially important contribution, and HIRDLS cross-track measurements will significantly supplement the MLS along-track measurements. Improvements in estimates of mass flux from stratosphere to troposphere, such as those provided by Gettelman et al. [1997], based on UARS data, are to be expected. The use of aircraft data - such as the Measurement of Ozone and Water Vapor by Airbus In-service Aircraft (MOZAIC) data base [Marenco et al., 1998] - in conjunction with the satellite measurements should allow for better statistical estimates of the global importance of stratosphere-troposphere exchange. (Also see Zahn et al. [2000].) Quantitative analysis of the upper tropospheric ozone budget and more stringent tests of models require, however, knowledge of key chemicals not measured by Aura. Aircraft will be needed for a thorough investigation of the important budget question. HO_x is needed, for example, since O+HO₂ is a major loss mechanism for ozone in the upper troposphere; NO_x and its difficult-to-quantify lightning source is also a variable of significance [e.g., Lamarque et al., 1996].
- **Understanding interannual and longer-term changes in the upper troposphere.** Aside from regional pollution sources and effects on the global troposphere, the short term characteristics of such events (e.g., seasonal effects, relationship to convection), and the upper tropospheric ozone budget, a better understanding of longer-term changes is needed. While seasonal variations of upper tropospheric O₃ seem to be fairly well captured by models [e.g., Wang et al. 1998; Law et al., 1998, 2000], pollution transport is episodic [e.g., Yienger et al., 2000], and requires an understanding on a global, interannual, and long-term basis. This includes issues such as the variability related to El Nino [e.g., Chandra and Ziemke, 1998] and expected long-term increases in pollution from Asia. The added constraints from the MLS and Aura-wide tropospheric data, coupled with related and needed complementary data from aircraft missions, will be critical for a more precise

predictive capability of tropospheric change (e.g., the interactive nature of climate change and tropospheric composition).

4 Climate Variability

The following key questions [NASA, 2000] related to climate variability will be addressed with EOS MLS data.

- *What trends in atmospheric constituents and solar radiation are driving global climate?*
- *How well can transient climate variations be understood and predicted?*
- *How well can long-term climate trends be assessed or predicted?*
- *How well can future atmospheric chemical impacts on ozone and climate be predicted?*

Discussion of specific objectives that will be addressed with the data are divided into the following subsections:

4.1 Climate Processes Involving Upper Tropospheric H₂O;

4.2 Radiational Effects of Stratospheric O₃ and H₂O;

4.3 Climatic Effects of Volcanic SO₂.

4.1 Climate Processes Involving Upper Tropospheric H₂O

A key uncertainty in predicting future changes to Earth's climate is the response of tropospheric water vapor to changes in the concentration of greenhouse gases. Water vapor in the upper troposphere (UT) has a particularly strong infrared radiative effect due to the low temperatures there, and the small amounts of water vapor in the UT exert enormous leverage on Earth's radiative balance. [Increases in upper tropospheric H₂O within global climate models result in much greater radiative effects at the surface than are caused solely by the build-up of CO₂ and other greenhouse gases \[e.g., Shine and Sinha, 1991\]. Of particular importance is the moisture in the dry subtropical regions which has a large cooling effect on the whole tropics \[Spencer and Braswell, 1997\]. Due to lack of data, however, there is large uncertainty regarding the actual abundance of water vapor in this region.](#)

Understanding the mechanisms that control humidity of the tropical troposphere is key to determining the nature of the water vapor feedback on climate, and is thus essential for improving climate change predictions. Observational evidence demonstrates that the subtropics are not as dry as the simple picture of outflow and subsidence from equatorial convective regions would imply – hence there must be additional moisture sources that hydrate the regions of the tropics characterized by descent. There are three hypotheses for the sources of this moisture: (1) evaporation of precipitation, (2) evaporation of detrained cloud particles, and (3) lateral transport. The relative contribution of these three sources to subtropical

moisture has major implications on how subtropical humidity will change in response to climate change and, hence, major implications for the water vapor feedback on climate [e.g., Held and Soden, 2000; Pierrehumbert, 2000]. Improving our understanding of the relative contributions from these sources is crucial for climate models to accurately simulate tropospheric water vapor and water vapor feedbacks.

Key questions related to upper tropospheric water vapor that will be addressed by EOS MLS include those itemized below.

- **What is the distribution of upper tropospheric water vapor, and how does it vary on seasonal and interannual time scales?**

EOS MLS will provide accurate measurements of water vapor in the tropical and subtropical upper troposphere, even in the presence of cirrus where observations by other techniques (infrared, visible, and ultraviolet) can be flawed. These measurements will enable the distribution and temporal variation of upper tropospheric humidity (UTH) to be accurately determined. UARS MLS has already provided valuable information of the distribution and temporal variability of water vapor in this region of the atmosphere. Figure 4.1-1 shows UARS MLS observations of upper tropospheric H₂O at 215 hPa, and how it changes spatially and temporally in relation to El Niño. The better accuracy and precision, and spatial and temporal coverage, of EOS MLS – and its extension of the UARS MLS data set over a longer time period – will further improve our knowledge of UTH.

Figure 4.1-1, now located at end after references, will be inserted somewhere near here.

- **What are the processes controlling upper tropospheric humidity?**

Joint analysis of the MLS UTH measurements together with information on the location and strength of deep convection and the circulation (e.g., from NOAA or other EOS satellite measurements of outgoing longwave radiation) will improve understanding of how deep convection affects upper tropospheric humidity (both in the tropics and subtropics). Consideration of simultaneous measurements of tracers of air motion from Aura instruments (including CO and HCN from MLS) will provide information on the origin of the air mass, and may place further constraints on the hydration paths for upper tropospheric water.

Quantitative measurements of ice water content in cirrus clouds will also provide important information about the supply of water to the upper troposphere. This, for example, will help determine what fraction of water in a convective air parcel remains in condensed form that can rapidly fall out.

Examination of the variation of the observed UTH over seasonal through interannual time scales will provide insight how different forcings, such as El Niño, may affect climate variability over these time scales.

Comparisons of the observed UTH with both process (e.g., trajectory and mesoscale) and general circulation models will help determine how well these models can reproduce the observed distribution, and will also test our understanding of the processes controlling upper tropospheric water vapor. For example, comparison of simple models that determine the humidity by tracing air parcels to the temperature of last saturation with the MLS observations will provide tests of the “lateral transport” hypothesis.

4.2 Radiational Effects of Stratospheric O₃ and H₂O on Climate

- **EOS MLS measurements will provide a better understanding of the radiative effects of O₃ and H₂O on stratospheric climate**

O₃ and H₂O play significant roles in the radiation budget of the stratosphere. O₃ absorbs solar ultraviolet radiation, and absorbs and emits infrared at wavelengths near 9.6 μm. The radiation budget of the middle and upper stratosphere is dominated by the heating effect due to solar radiation absorbed by O₃ and the cooling effect due to infrared radiation emitted by CO₂ and O₃. The existence of the temperature maximum at the stratopause is attributable to the absorption of solar radiation in the upper stratosphere. H₂O also absorbs solar radiation in the near infrared, and absorbs and emits in the thermal infrared [e.g., Andrews et al., 1987]. The radiative effects of H₂O as a fraction of the total heating rate are particularly significant in the lower stratosphere.

Changes in stratospheric O₃ or H₂O are expected to change stratospheric circulation in ways that are difficult to predict. In addition to direct effects (e.g., the influence of the change in heating and cooling rates on both the vertical static stability and the meridional temperature gradients), there is a feedback via geostrophic balance on the magnitude of the zonal mean winds. This impinges on propagation of planetary waves from the troposphere into the stratosphere and the rate at which they are radiatively damped and mechanically dissipated [e.g., Pawson et al., 1992], and will affect the strength of the diabatic circulation. A full understanding of the past and future interaction of these processes entails calculation of detailed three-dimensional heating and cooling fields, which requires accurate knowledge of the distributions of H₂O and O₃ throughout the stratosphere.

Garnier and Shine [1999] – in a study of direct and indirect effects of ozone depletion – concluded that the observed downward trend in stratospheric temperatures is broadly consistent with a radiative link to the changing O₃ amount. There have been, however, several developments since that study. Garnier and Shine's [1999] initial calculations used rather idealized representation of the O₃ changes and did not consider changes in other greenhouse gases. The existence of more refined datasets, and the consideration of other greenhouse gases and the possibilities of chemical interactions, is forcing some re-appraisal of that assessment. Recent analyses [Rosenlof et al., 2001] suggest that stratospheric H₂O has been increasing for more than 45 years. The possible impact of trends in stratospheric H₂O on stratospheric cooling has been estimated by Forster and Shine [1999] as of the same order (but of opposite sign) as that due to trends in stratospheric O₃ over recent decades. Subsequently, other investigators [Smith et al., 2001; Shindell, 2001; Dvortsov and Solomon, 2001; Oinas et al., 2001; Ramaswamy et al., 2001; Ramaswamy and Schwarzkopf, 2002; Forster and Shine, 2002] have assessed stratospheric cooling and radiative forcing by O₃ and H₂O. There is a wide disparity in the values calculated by these investigators. Although some of the variation in the estimates is related to differences in the radiative schemes used by these authors, a major source of the variation is uncertainty in the vertical distribution of the trends in stratospheric H₂O [Shine et al., 2003]. World Meteorological Organization [2003] states that 'changes in ozone, well-mixed greenhouse gases, and stratospheric water vapor can explain the major features of the observed global and annual mean stratospheric cooling over the past two decades.' EOS MLS (and

HIRDLS) will accurately measure the variations of O₃ and H₂O in the region where there is currently most uncertainty and allow the effects of future changes to be more confidently attributed and more accurately predicted.

All trace gases that reach the stratospheric “overworld” (i.e., the part that cannot be reached from the troposphere except by diabatic processes), including the pollutants responsible for destroying ozone, must pass through the tropopause transition layer in the tropics. The vertical motions in and immediately above this layer are therefore important to stratospheric change, but they are small and notoriously difficult to estimate. One of the most promising ways is via heating rate calculations, but these have been hampered by the fact that the net heating rates are small differences between heating and cooling rates that are themselves small. Good estimates of net heating require extreme accuracy in calculation of opposing effects. A major difficulty in the past has been the poor knowledge of the concentrations of radiatively active gases in this region, especially of H₂O. EOS MLS (and HIRDLS) will provide high-quality measurements in this crucial region of the atmosphere and should permit important improvements in our knowledge of the trans-tropopause transport mechanisms.

- **EOS MLS measurements will provide a better understanding of the greenhouse forcing of surface temperature by O₃ and H₂O in the lower stratosphere and around the tropopause**

The greenhouse forcing of surface temperature is significantly affected by O₃ and H₂O in the lower stratosphere and around the tropopause [IPCC, 2001]. (The important role of upper tropospheric H₂O on climate change is covered in section 4.1.) The amount of forcing is sensitive in complex ways to their amounts, which creates difficulties in diagnosing past changes of surface forcing and predicting future changes.

A given fractional change in O₃ has the largest effect on surface forcing when it occurs near the tropopause, but the effect is still important for O₃ changes up to ~23 km [Forster and Shine, 1997]. Increasing stratospheric O₃ will decrease the shortwave radiation, but increase the longwave radiation, reaching the surface. (It is thought that recent decreases in stratospheric O₃ have partially offset the warming due to increase of CO₂ and other greenhouse gases [reference?].) Increasing tropospheric O₃, on the other hand, cools the surface. Consequently, the effects at the surface depend critically upon the transition height between decreasing ozone aloft and increasing ozone below. The O₃ changes above and below the tropopause are poorly characterized at present. As a consequence, scientific understanding of greenhouse forcing by stratospheric O₃ changes is currently classified as only “medium” [IPCC 2001, pages 302 and 358]. The new EOS MLS (and HIRDLS, TES) measurements will reduce this ambiguity for future changes.

Stratospheric O₃ recovery is not expected to be generally detectable much before 2010 [Jackman et al., 1996; Hofmann and Pyle, 1999]. (Hoffman et al. [1997] have argued that recovery in the Antarctic ozone profile may be conclusively detected as early as 2008.) Until then, attempts to attribute interannual stratospheric ozone changes to recovery or to natural variability will need detailed case studies requiring three-dimensional measurements of O₃, which EOS MLS (and the other Aura instruments) will supply. Likewise, an understanding of future changes in upper tropospheric O₃ will need to consider the expected increased O₃ flux from the lower stratosphere into the troposphere as stratospheric O₃ recovers. Neglect of this effect is a known potential defect in the most

recent IPCC [climate change](#) assessment [IPCC, 2001, page 275]. [Aura](#) measurements will allow that flux to be better quantified in monitoring future trends.

The important contribution to surface radiative forcing from H₂O in the tropopause [region](#) and lower stratosphere has been highlighted in a number of studies. Forster and Shine [1999] estimate an increased greenhouse forcing of 0.2 Wm⁻² from stratospheric H₂O since 1980. [They conclude that the impact of trends in stratospheric H₂O on the radiative forcing of tropospheric climate are of the same order as that due to changes in stratospheric O₃ over recent decades.](#) Forster and Shine [2002] have recently revised their earlier estimates of the change since 1980 to 0.29 Wm⁻², and obtained an upper limit of 0.62 Wm⁻² since 1960. [They still conclude that the magnitude of radiative forcing by stratospheric H₂O trends is comparable \(and opposite in sign\) to the radiative forcing due to stratospheric O₃ depletion.](#)

A study of the greenhouse effects of aircraft [IPCC, 1999] pointed out that the possible existence of a substantial fleet of supersonic aircraft in the latter part of this century could have a much larger warming effect (through H₂O deposited in the lower stratosphere) than the subsonic aircraft that they would replace. Again, the present distribution of H₂O in that region is poorly characterised, but will be addressed by the EOS MLS measurements, [and those of the other Aura instruments.](#)

4.3 Climatic Effects of Volcanic SO₂

Large quantities of gaseous SO₂ can be injected into the stratosphere by volcanoes. This SO₂ is converted by reaction with OH into sulfuric acid [McKeen et al., 1984], which –with a time constant of about 1 month [Bluth et al., 1992; Read et al., 1993] – condenses into aerosols. Volcanic aerosols have significant radiative effects: their scattering of incoming shortwave solar radiation leads to surface cooling, and their absorption of upwelling longwave terrestrial radiation leads to [heating in the](#) lower stratosphere [e.g., Michelangeli et al., 1989; McCormick et al., 1995, and references therein]. Temperature increases of 3-4 K at 23-24 km were observed at low latitudes after both the El Chichon and Mt. Pinatubo eruptions [Labitzke et al., 1983; Labitzke and McCormick, 1992].

EOS MLS provides unique capabilities to test, and possibly improve, our understanding of stratospheric aerosol formation from volcanically-injected SO₂, and the short term (on time scales of years) effects of these aerosols on climate. Dense volcanic aerosols have negligible effects on MLS measurements as shown in Figure 3 of Waters et al. [1999], and EOS MLS can measure the vertical profile of SO₂ and its decay as the aerosols form, as was done with UARS MLS following the Pinatubo eruption [Read et al., 1993]. The simultaneous MLS measurements of SO₂ and OH will provide a more stringent test than ever before possible of our understanding of the conversion of stratospheric SO₂ to sulfuric acid. The MLS measurements of temperature, tracers (such as N₂O and O₃) of vertical transport in the lower stratosphere, and geopotential height – all made in the presence of dense volcanic aerosol which cannot be done with other remote sensing techniques – will provide unique information on the short term response of stratospheric temperature and circulation to aerosol increases. Such information should help test models that predict long term climate change due to aerosol variations. Some MLS scientific objectives related to climatic effects of volcanic SO₂, and some expected studies for reaching these objectives, include the following.

- **Determination of the total amount and vertical profile of SO₂ injected into the stratosphere by volcanoes that erupt during the Aura mission, to provide early estimates of the magnitude of their effects on near-term climate.**

The MLS measurements will be compared with similar measurements from OMI (and other instruments) that measure total volcanic SO₂ to check for consistency. UARS MLS detected stratospheric SO₂ abundances more than 100x lower than could be detected from satellite by UV techniques [Read et al., 1993] at the time, so MLS may be able to provide information on SO₂ from much less intense volcanoes than can be observed by other techniques.

- **Quantitative testing of our understanding of the decay of SO₂ leading to formation of sulfuric acid and stratospheric aerosol.**

MLS measurements of the decay of SO₂ as a function of height will be compared quantitatively to the rate of its decay as a function of height calculated using MLS OH and the laboratory-measured rate for the reaction $\text{SO}_2 + \text{OH} + \text{M} \rightarrow \text{HSO}_3 + \text{M}$ (where M indicates a three-body reaction) thought to control the SO₂ decay.

- **Testing of models that predict the short-term response of stratospheric temperature and circulation to an abrupt increase in aerosol.**

MLS measurements of the profiles of temperature, tracers and geopotential height as a function of time during and after the formation of stratospheric aerosol from volcanically-injected SO₂ will be compared with predictions of models (that are expected to use aerosol information from independent measurements) to test the model validity for predicting such phenomena.

5 Summary

Not sure yet what will go here, or even if this section is needed - but will keep in for now as a placeholder.

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FIGURES NOT YET INSERTED IN TEXT

Figures on the following pages will be inserted at appropriate places in the text sometime in the future when the document is more mature.

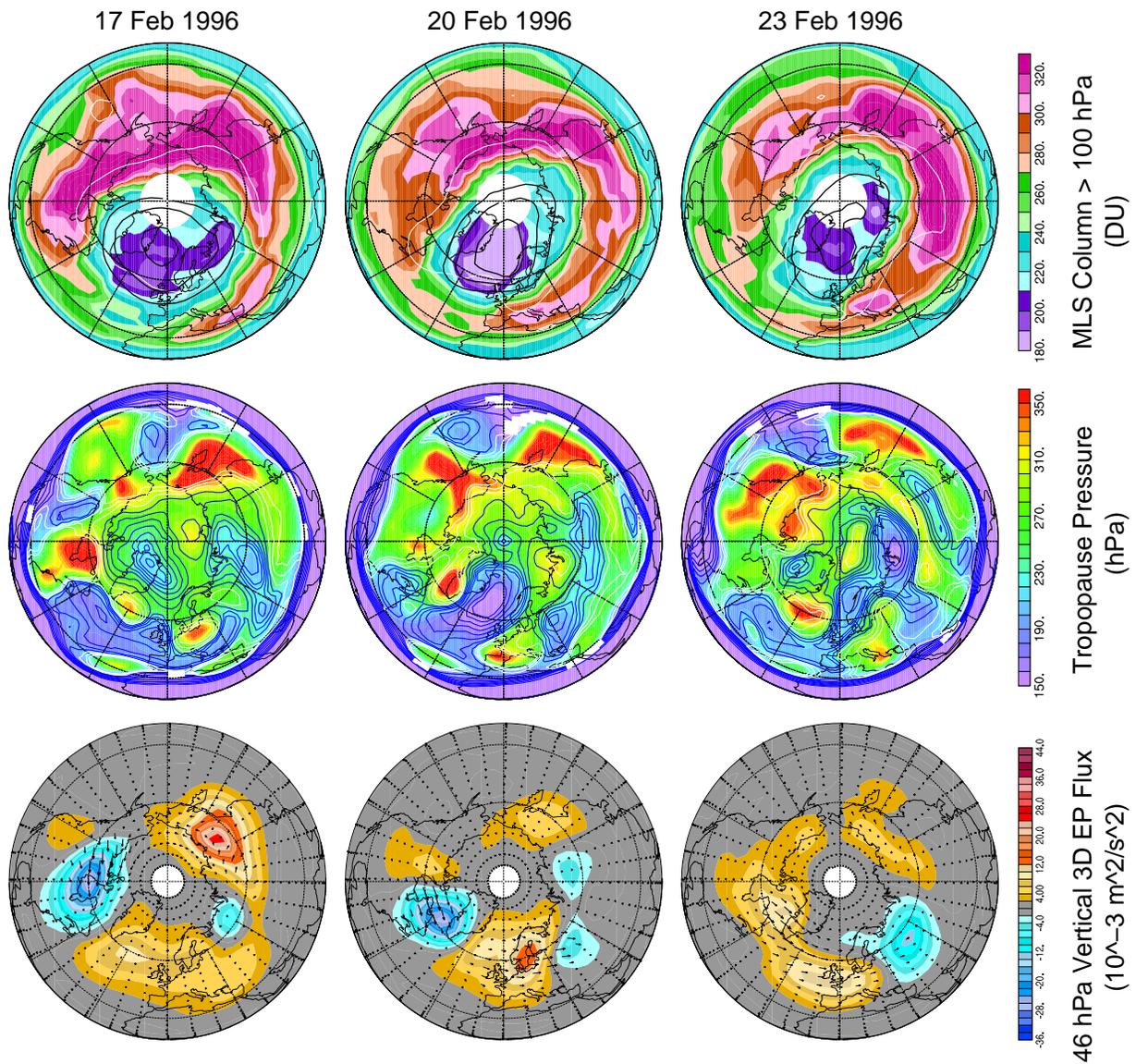


Figure 2.1.5-1. Top: column ozone above 100 hPa from UARS MLS. Center: “dynamical” tropopause pressure (colors) and temperature (overlaid contours, by 2~K, 200-214~K in blue, 216-220~K in white). Bottom: 3D EP fluxes at 46~hPa (colors show vertical component), before, during and after (left to right) an ozone minihole event in February 1996. 190, 195 and 200 K temperature contours (black), and the 1.2 sPV contour (white, near the vortex edge), are overlaid on column ozone plots. A high, cold tropopause develops, associated with an underlying upper tropospheric ridge. Negative (positive) vertical 3D EP fluxes show cooling (warming) and upward (downward) adiabatic motion [Sabutis, et al. 1997, Sabutis and Manney, 2000]. There is cooling/rising motion (western flank of the cold region) upon entering the region of high/cold tropopause, and warming/sinking (southeast edge of cold region) exiting it. The pattern of wave activity “corrals” the low column ozone into a localized region with tighter gradients, and is characteristic of minihole formation. Interannual variability in minihole formation and frequency contributes strongly to interannual variability in midlatitude and Arctic column ozone. [3D EP flux calculations and interpretation courtesy of Joseph Sabutis.]

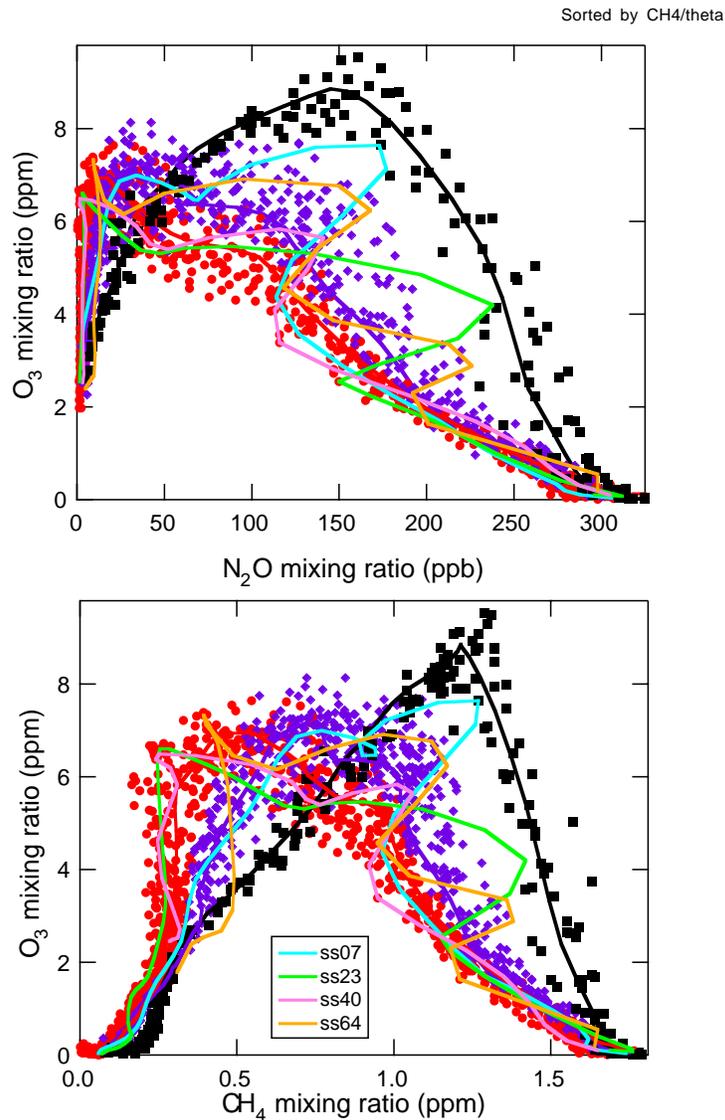


Figure 2.1.5-2. Correlations of ozone versus N₂O (top) and CH₄ (bottom) from ATMOS data in November 1994. Orange, purple and black points represent “protovortex”, midlatitude and tropical correlations, respectively, and same-colored lines their averages. Overlaid light orange, pink, cyan and green lines show correlations for individual profiles that have laminae in them. Changes in the tracer correlations in the regions of laminae indicate the origins of the air causing the laminae: the blue, light orange and green curves show laminae from tropical air in midlatitudes (extensions toward tropical curve); the green and light orange curves also show laminae from protovortex air drawn out into midlatitudes (extensions toward protovortex curve; the pink curve is inside the protovortex with a lamina resulting from midlatitude air intruding into it (extension from protovortex toward midlatitude curve).

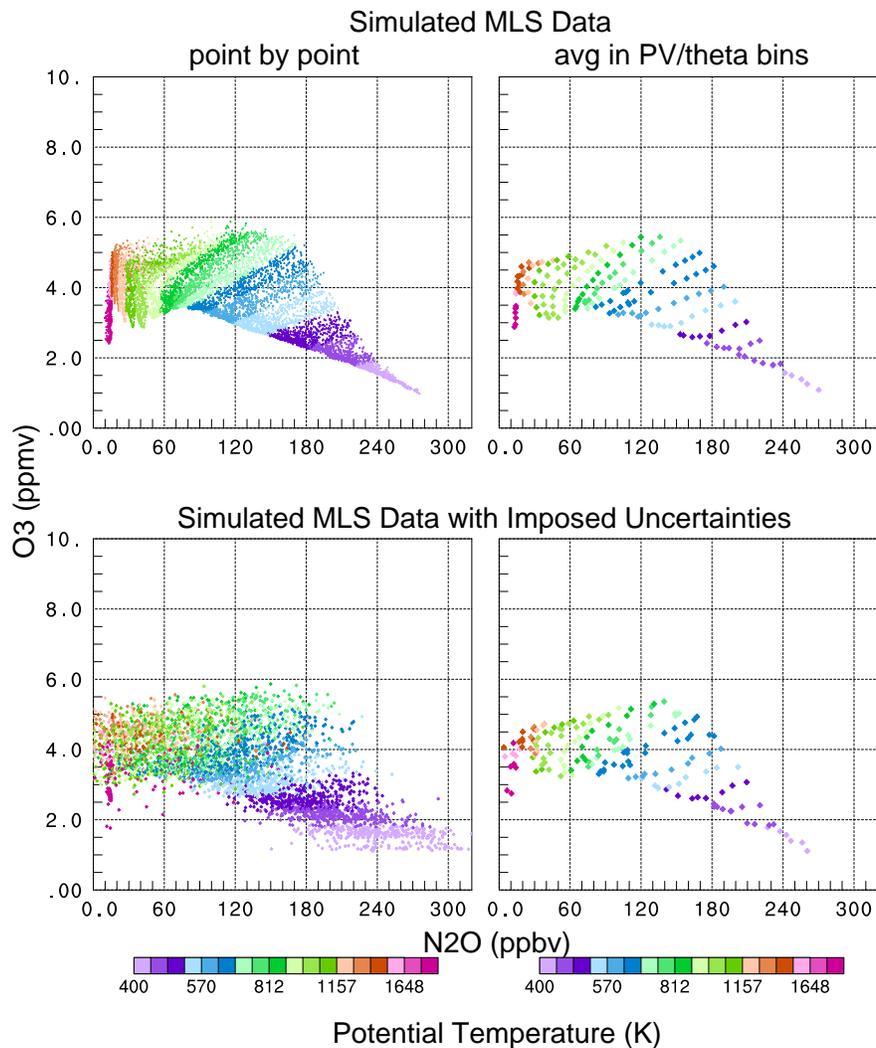


Figure 2.2.5-1. O_3/N_2O Correlations in the polar vortex for 20 Feb 1996 for the simulated dataset used for MLS studies; (a) at each gridpoint throughout the stratosphere from 380 to 2000 K; (b) averaged in PV/theta bins; (c) at each gridpoint, with Gaussianly distributed random errors of magnitude consistent with the EOS MLS precision estimates added to each datapoint; and (d) fields with uncertainties averaged in PV/theta bins. The PV/theta averaging results in an ozone/ N_2O distribution closely resembling the "truth", even though the gridpoint-by-gridpoint correlations are seriously degraded.) (LABELS NEED TO BE FIXED.)

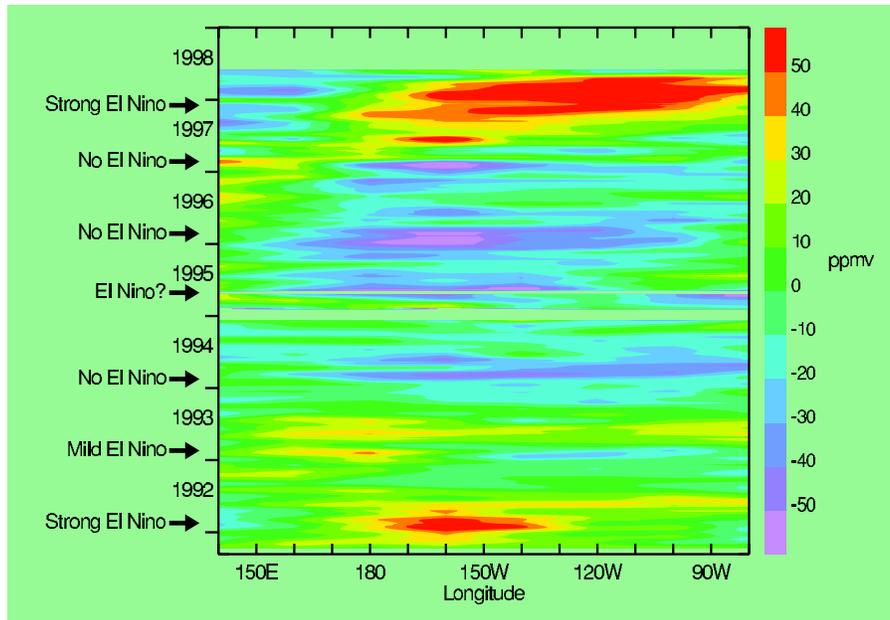
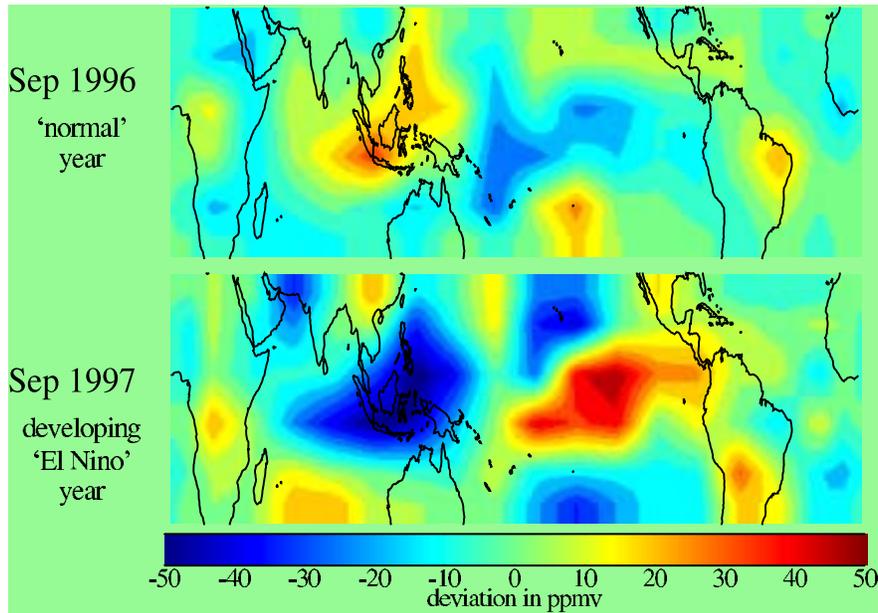


Figure 4.1-1. UARS MLS observations of (top) the different spatial distributions in upper tropospheric 215 hPa H₂O for 'normal' and 'El Niño' periods and (bottom) the temporal evolution of tropical 215 hPa H₂O over a 6-year period and its correlation with El Niño sea surface temperature events.