Tropical Composition and Climate Coupling (TC$^3$) Experiment

Draft White Paper

James R. Holton$^1$, Ross J. Salawitch$^2$, Darryn W. Waugh$^3$, Paul O. Wennberg$^4$

$^1$Department of Atmospheric Sciences, University of Washington, Seattle, WA 98195

$^2$Atmospheric Chemistry Research Element, Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA 91109

$^3$Department of Earth and Planetary Science, Johns Hopkins University, Baltimore, MD 21218

$^4$Division of Geological and Planetary Sciences, California Institute of Technology, Pasadena CA 91125

Please send comments to:

holton@atmos.washington.edu
rjs@caesar.jpl.nasa.gov
waugh@jhu.edu
wennberg@gps.caltech.edu

Version 1.0, September 2000
The composition of air transported from the upper tropical troposphere defines the chemical boundary condition for the global stratosphere and for the upper troposphere in the subtropics. These boundary conditions influence the composition and climate of the entire middle atmosphere. The recovery of stratospheric ozone due to the ban on CFCs will be influenced by how these boundary conditions may change in the coming decades. For example, an increase in the abundance of stratospheric water will, all else being equal, lead to a decrease in the concentration of ozone. In a broader sense, understanding the physical processes that control water in the tropical upper troposphere is critical for assessing how surface temperatures will be affected by rising concentrations of greenhouse gases. The upper tropical troposphere is perhaps the most important region for understanding how stratospheric composition and climate are coupled.

The Tropical Composition and Climate Coupling Experiment (TC³) will obtain, over several years, a comprehensive set of observations to investigate the physics and chemistry in the region of the tropical tropopause. A two-site Pacific aircraft campaign is proposed with flights from Guam (13.4°N, 144.4°E) and Hawaii (21.3°N, 157.9°W) coupled with balloon-borne observations from a yet-to-be-determined tropical site. The science goals of TC³ will benefit directly from the availability of AURA measurements that will place the local observations within a global and long-term context. In return, TC³ is designed to maximize the availability of co-located profiles for AURA validation.

TC³ will address the following questions:

- What are the physical mechanisms that control the humidity of the stratosphere?

The increase of greenhouse gases is expected to alter the amount of water entering the stratosphere. Such changes can profoundly affect stratospheric chemistry. At mid-latitudes, H₂O supplies the HO₅ radicals that directly catalyze removal of O₃. Higher levels of H₂O exacerbate O₃ loss by promoting more efficient heterogeneous chlorine activation at mid-latitudes and in polar regions during winter. The ability to understand how stratospheric water may change in the
coming decades is limited, however, by our uncertainty about the precise mechanisms that dehydrate air entering the stratosphere.

- **What is the chemical fate of short-lived compounds transported into the upper troposphere?**

  The chemical properties of tropical upper troposphere define the chemical boundary condition for the stratosphere. Despite a decade of tremendous progress in tropospheric and stratospheric chemistry, the photochemistry of the upper troposphere remains poorly constrained. This uncertainty limits our ability to understand the fates of numerous compounds (and their degradation products) transported into the near tropopause region. Such knowledge is required if we are to understand the impact of climate change on the stratosphere in general and ozone in particular.

- **What mechanisms maintain the humidity of the tropics and subtropics?**

  The response of the hydrological cycle to changes in the concentration of greenhouse gases is perhaps the single most important source of uncertainty in predicting future changes to Earth's climate and composition. The small amounts of water vapor in the upper troposphere (UT) exert enormous leverage on Earth's radiative balance. Of particular importance is the moisture in the subtropical regions. These dry regions have a large cooling effect on the whole tropics. Understanding the mechanism that controls the humidity of the subtropics is key to determining the nature of the water vapor feedback on climate.

These three key questions are related by a common observational strategy. TC³ will provide a large suite of in situ and remote measurements of tracers of atmospheric motion and chemistry. The same tracers can be used to test the series of hypotheses outlined below regarding the physical and chemical mechanisms that control the distribution of ozone, water vapor and other important trace constituents of the lower stratosphere (LS) and upper troposphere (UT). AURA (and other satellite) observations within the UT/LS will provide information on the spatial, seasonal, and interannual variations while the two-site Pacific aircraft campaign is designed to focus on smaller spatial scale process studies. Aircraft and balloon observations obtained during northern winter and summer will enable examination of important seasonal differences in transport and chemistry. The combined data set will provide high vertical scale resolution observations for validation of AURA measurements in the tropics.

2. Tropical Transport and the Tropical Transition Layer (TTL) Hypothesis

When considering transport of trace constituents it is useful to consider the troposphere-stratosphere system divided into regions of rapid “isentropic mixing” that are separated by semi-permeable “mixing barriers” (of which the tropopause is one), as illustrated in
Figure 7-6 of WMO (1999). In this framework, the tropical troposphere is viewed as well-mixed, with convection rapidly transporting air to (and occasionally across) the tropical tropopause. Above the tropical tropopause there is slow ascent, which is primarily in response to extratropical wave driving in the stratosphere (the “stratospheric pump”) (Holton, 1995). In addition to these vertical motions there is isentropic mixing across the subtropical tropopause, which transports lowermost stratospheric air into the tropical UT, and across the stratospheric subtropical barrier (in the (isolated) “tropical pipe” paradigm this mixing is exclusively out of the tropics). While this conceptual picture has been extremely useful for understanding stratospheric transport and its role in determining the distribution of trace constituents, research in recent years indicates that it is incomplete and modifications are needed, particularly near the tropical tropopause.

Although convection has been observed to penetrate the tropical tropopause, this is not a common occurrence and tropical deep convection rarely penetrates above 14 km. So rather than considering the whole tropical troposphere to be well-mixed by convection, it is may be better to consider convection to mix air up to around 14 km, with slow ascent (driven by the stratospheric “pump”) above this altitude (Highwood and Hoskins, 1998; Folkins et al., 1999).

Together with this slow ascent there is also mixing across the subtropical tropopause, driven primarily by tropospheric weather systems (in particular, the summer monsoons) (e.g. Chen, 1996; Postel and Hitchman, 1999). These systems not only mix lowermost stratospheric air and tropical UT air, but they also have an effect in the lowest altitudes of the overworld (where all isentrops exist solely within the stratosphere (Holton, 1995). Analyses of trace gas and aerosol observations indicate there is rapid transport out of the tropics in the lowest 2 km of the stratosphere (Trepte et al., 1993; Rosenlof et al., 1997; Boering et al., 1995; Mote et al., 1998). More importantly for understanding the tropical budgets of constituents, there is mixing of mid-latitude air into the region of tropical ascent [especially below ~20 km (440K)] (e.g., Avallone and Prather, 1995; Minschwaner et al., 1996, Volk et al., 1996; Flocke et al., 1999).

Taken together the above studies imply that, rather than thinking of a definite boundary between the troposphere and stratosphere at the tropical tropopause, it is more appropriate to think of a tropical transition layer (“TTL”) that extends from around 2 km below the tropical tropopause (i.e. top of typical deep convection) to around 2 km above the tropopause (Highwood and Hoskins, 1998; Folkins et al., 1999). The TTL is illustrated schematically in Figure 2.1. Within the TTL there is slow ascent as well as quasi-horizontal mixing with the extratropical stratosphere (both in the middle and overworld), and some (probably small) degree of convective overshooting and mixing. Such a transition layer, with slow ascent and some mixing of stratospheric air into the tropical UT is consistent with observations of a “standing reserve” of stratospheric air in the tropical UT (Tuck et al., 1997) and with observed vertical profiles of total particle mixing ratios in the tropics (Brock et al., 1995).

The above picture is based on a zonally averaged view of the circulation and constituent distributions, but there are notable zonal asymmetries within the TTL (Figures 2.2 and
The most obvious spatial variations are due to the occurrence of regions of strong convection, which result in large spatial, and seasonal, variations in the ascent into (and temperature within) the TTL. However, there are also important variations in horizontal flow and lateral transport, which are linked to above variations in vertical motion and regional tropospheric systems. Of particular note are the summer subtropical monsoons, and the equatorial westerly ducts that exist during northern winter.

The summer subtropical monsoons have a strong impact on the transport and composition of the TTL. Several studies have shown that upper-level anticyclones associated with the monsoons are very important for transport across the subtropical tropopause (Chen, 1996; Postel and Hitchman, 1999; Dethof et al., 1999). In particular, there is more transport across the tropopause during summer, with the transport (wave breaking) occurring predominantly downstream of the monsoons. The monsoon anticyclones extend into the stratosphere and not only influence STE but also tropical-extratropical exchange in lower stratosphere (Trepte et al., 1993; Horinouchi et al., 2000), i.e., the monsoons can have an influence throughout the depth of the TTL. As discussed below, the monsoons may have a particularly large impact on both UT and LS water vapor.

Another important regional circulation feature is the occurrence of equatorial westerlies over the Pacific and Atlantic Oceans, at all altitudes within the TTL, during northern winter (Tomas and Webster, 1994), as shown in Figure 2.2. [During northern summer there are easterlies at all longitudes (Figure 2.3)]. These “westerly ducts” are preferred regions for tropical-extratropical interactions (e.g., Tomas and Webster, 1994), and in particular appear to be preferred regions for mixing into and within the TTL (Waugh and Polvani, 2000; Horinouchi et al., 2000). Furthermore, west of these ducts there is an anticyclonic circulation between the equatorial easterlies and subtropical westerly jets, which transports air into and out of the moist regions in UT and the tropopause cold pool over Indonesia and Western Pacific (and hence will impact both the UT subtropical moisture and stratospheric water vapor, see below).

In addition to the seasonal variations discussed above there are also interannual variations. There is likely to be a strong influence of both the QBO and ENSO on the TTL (through location of tropical convection as well strength of monsoons and the equatorial westerly ducts). Furthermore, changes in the strength of the stratospheric circulation (such as those inferred from trends in upper stratospheric CH₄, e.g., Nedoluha et al., 1998; Randel et al., 1999) are also likely to influence the transport and composition within the TTL.

In summary the TTL hypothesis for transport within tropical UT and LS is:

1. There is a layer between approximately 14 and 20 km in which there is slow ascent (driven by extratropical wave driving in the stratosphere) and quasi-horizontal mixing with the extratropics.
2. The lateral mixing into/out of/within this layer is limited to certain regions and seasons: downstream of the summer monsoons, and within the northern winter westerly ducts (over the Pacific and Atlantic oceans).

If this hypothesis is correct,

- The composition in the upper 2 to 3 km of the tropical troposphere will not be uniform, and will be a mixture of air from different locations (the surface, the stratosphere, and the free troposphere).
- There will be a significant signature of middle latitude air in the first 2 km above the tropical tropopause.
- There will be seasonal and longitudinal variations in trace constituents within the tropical UT and LS that are strongly coupled to seasonally varying, regional tropospheric systems.

The measurement strategy proposed in the following sections to address issues regarding stratospheric and upper tropospheric water vapor and the photochemistry in the UT/LS will enable the above issues to be tested. Measurements of tracers with a range of lifetimes will provide (via both profiles and tracer-tracer relations) information on dominant transport pathways to the different regions (i.e. the fractions of air in a given region that has been mixed rapidly from the surface or lateral mixed into the region).

3. Stratospheric Water Vapor

3.1. Introduction

A significant fraction of the observed ozone depletion during the past 2 decades in polar regions and at mid-latitudes is clearly associated with the rise in chlorine and bromine due to anthropogenic release of CFCs, halons, and other halocarbons (WMO, 1999: Chapter 12). Industrial production of these pollutants has essentially been banned by the Montreal Protocol and its amendments. If “all else remains equal”, the ozone layer will recover as chlorine and bromine levels fall. As discussed below, increases in stratospheric H$_2$O may complicate the expected recovery of the O$_3$ layer.

Observations of H$_2$O by ground based microwave instruments (Peter, 1998), by balloon-borne frost point hygrometers (Oltmans and Hoffman, 1995), and by the Halogen Occultation Experiment (HALOE) satellite instrument (Evans et al., 1998) all show evidence for a rise in the mixing ratio of H$_2$O during the 1990s. The time series for H$_2$O exhibits non-linear variations due to numerous forcings and the computed linear trend for H$_2$O is sensitive to the time period of the data record, the range of altitudes and latitudes under consideration, and to the statistical technique used to extract the linear component from the signal. The HALOE observations reveal a rise in H$_2$O between 1992 and 1996 of 55 to 150 ppbv/yr, as shown in Figure 3.1. This change is much larger than can be
attributed to the slow rise in tropospheric CH$_4$ (WMO, 1999: Chapter 6). The rate of 
increase of stratospheric H$_2$O observed by HALOE has slowed subsequent to 1996, but 
HALOE still records a substantial increase between 1992 and 1999 (Figure 3.2) (Smith et 
al., 2000).

The record for H$_2$O is not clear, however. Observations from SAGE II show a slight 
decrease in stratospheric H$_2$O over the 1990s (Rosenlof et al., 2000). Observations of 
H$_2$O+2CH$_4$ from instruments aboard the ER-2 also indicate the lack of a significant trend 
between 1993 and 1997 (Figure 3.3) (Hurst et al., 1999). Most interestingly, the HALOE 
trend for H$_2$O in the lower stratosphere between 1992 and 1999 shown in Figure 3.2 is 
rather small, perhaps consistent with the ER-2 measurements. Perhaps the larger trend for 
H$_2$O at higher altitudes reported by HALOE reflects conditions for older air than was 
sampled by the ER-2.

An increase in the abundance of stratospheric H$_2$O will, all else being equal, lead to a 
decrease in the concentration of O$_3$. At mid-latitudes, H$_2$O supplies HO$_x$ radicals that 
directly catalyze removal of O$_3$ (e.g., McElroy et al., 1992; Wennberg et al., 1994). 
Higher levels of H$_2$O exacerbate O$_3$ loss by promoting more efficient chlorine activation 
by sulfate aerosols in the lower stratosphere and by promoting formation of polar 
stratospheric clouds at warmer temperatures in both the Antarctic and Arctic circumpolar 
vortices (e.g., Kirk-Davidoff et al., 1999). If a 100 ppbv/yr trend in H$_2$O (the trend 
oberved between 1992 and 1996) were to persist until 2010, the expected recovery of 
stratospheric O$_3$ due to the ~15% decline in chlorine loading would be negated by 
increased O$_3$ loss from HO$_x$ radicals (Jucks and Salawitch, 2000).

In general terms the aridity of the stratosphere is clearly due to the cold tropical 
tropopause (Brewer, 1949). However, the precise physical mechanism(s) responsible for 
the aridity of the lower stratosphere are unclear. Various “Hypotheses of Stratospheric 
Aridity” are described in the following section, and proposed observational tests of these 
hypotheses are given in Section 3.5.

One of the motivations of the proposed field campaign is that the cause of the recent rise 
in stratospheric H$_2$O is not understood. A simple explanation for this rise is a warming of 
the coldest regions of the tropical tropopause since 1992 (e.g., Evans et al., 1998). 
However, an analysis of a time series (from 1979 to present) of temperature near the 
tropical tropopause, shown in Figure 3.4, reveals a general cooling trend from 1992 to 
1998 throughout the tropics and a slight cooling trend in the Western Pacific (Simmons et 
al. 1999). The tropical-mean trend is most pronounced between 1992 and 1996, when 
ground-based and HALOE observations suggest stratospheric H$_2$O increased quite 
rapidly. In section 3.3, we describe several “Hypotheses of Stratospheric H$_2$O Change” 
that are decoupled from minimum tropopause temperature. Since future changes in 
stratospheric O$_3$ cannot be forecast reliably until we have a better understanding of the 
processes that regulate stratospheric H$_2$O, it is essential that observational tests of these 
hypotheses be designed and carried out.
3.2 Hypotheses of Stratospheric Aridity

The mixing ratio of stratospheric H$_2$O is controlled by processes in the tropical upper troposphere and lowermost stratosphere. In general terms the aridity of the stratosphere is clearly due to the cold tropical tropopause. However, the precise physical mechanism(s) responsible for the aridity of the lower stratosphere are unclear.

Various hypotheses have been proposed to account for details of stratospheric aridity. There is no commonly accepted set of definitions or nomenclature for these hypotheses. The definitions and names given below represent an attempt to distill the essence of hundreds of papers that have been written on this topic during the past 50 years and are used to motivate the observational plans described in the following sections. It is beyond the scope of this document to delve into great detail regarding the numerous observational details in support of, or refuting, the various hypotheses. Summaries of these observational details are given by Holton et al. (1995), Mote et al. (1996), Moyer et al. (1996), Keith (2000), and Sherwood and Dessler (2000). Suffice to say there is no commonly accepted theory for how the aridity of the stratosphere is maintained.

Class I: One dimensional models of stratospheric H$_2$O. Each of these hypotheses treats the entry of air into the stratosphere, and the desiccation of stratospheric air, as events that happen essentially simultaneously during vertical ascent in the tropics.

1) **Cold Trap** - slow, large-scale upwelling through the cold tropopause dehydrates ascending air (Brewer, 1949)

   1a) **Fountain** - air enters the stratosphere only in the Western Pacific, the coldest regions of the tropopause (Newell and Gould-Stewart, 1981)

2) **Convective Overshoot** - Convective turrets ascend rapidly from near the ground and penetrate into the stratosphere, where they induce rapid dehydration due to precipitation of ice (Danielsen, 1982)

   2a) **Ice Lofting** - air entering the stratosphere contains significant amounts of ice, some of which precipitates and some of which evaporates (Johnston and Solomon, 1979).

   2b) **Supersaturation** - air entering the stratosphere is highly supersaturated (e.g., 150%) with respect to cloud top temperature (Moyer et al., 1996)

Class II: Three dimensional models of stratospheric H$_2$O. These hypotheses treat the entry of air into the stratosphere, and the desiccation of stratospheric air, as events that happen at different times and in different places. A feature of these hypotheses is recognition that processes (such as precipitation and mixing) within the TTL may play an important role in determining stratospheric H$_2$O.
3) **TTL Cold Trap** - air enters the TTL to the west of the region of coldest temperatures in an area of forced upwelling and parcels are advected to the region of coldest TTL temperatures by an anti-cyclonic flow. As the air approaches the region of coldest temperature, it sinks, cools, and desiccates over relatively long time scales (several days). Downstream of the TTL cold trap, the air is heated diabatically and rises, eventually mixing into the stratosphere (J. R. Holton, private communication, 2000).

4) **TTL Overshoot** - convective turrets penetrate the TTL. The more energetic turrets penetrate to higher levels and become rapidly (time scales of less than a day) dehydrated due to sedimentation of ice. The resulting humidity of the TTL is caused by mixtures of air parcels that detrain at different potential temperatures. Slow, zonally uniform ascent carries desiccated TTL air into the stratosphere (Sherwood and Dessler, 2000).

5) **Stratospheric Dehydration** - Dehydration in the lowermost stratosphere is induced by meso or synoptic-scale wave activity that lifts and cools air masses in the lower stratosphere (Fritsch and Brown, 1982; Teitelbaum *et al.*, 2000).

The **Cold Trap** theory was proposed to explain the overall aridity of the stratosphere (Brewer, 1949). It was later realized that the water vapor mixing ratio of air entering the stratosphere (currently considered to be about 3.8 to 4.0 ppmv: Abbas *et al.*, 1996; Dessler, 1998) is drier than can be accounted for by the mean temperature of the tropical tropopause, prompting the proposed *Fountain* corollary to the original theory (Newell and Gould-Stewart, 1981). In both cases, stratospheric aridity is maintained by exposure of air to the coldest regions of the tropical tropopause as these parcels first enter the stratosphere.

The **Convective Overshoot** theory was based on the recognition that some cumulonimbus clouds penetrate the tropopause and can irreversibly mix with stratospheric air (Danielsen, 1982). Data from the NASA STEP (Stratosphere-Troposphere Exchange Program) mission provided evidence for a single occurrence of such an event (Danielsen, 1993). The *Ice Lofting* and *Supersaturation* corollaries account for the observation that the water vapor mixing ratio of air entering the stratosphere is typically wetter than can be accounted for by the exceedingly cold temperatures at the top of convective turrets.

Both the **TTL Cold Trap** (J. R. Holton, private communication, 2000) and the **TTL Overshoot** (Sherwood and Dessler, 2000) hypotheses were motivated in part by recent meteorological analyses that indicate the cold tropical Western Pacific region is an area of net subsidence near the tropopause (Gettelman *et al.*, 2000; Sherwood, 2000). This is the same geographic area for which Newell and Gould-Stewart (1981) hypothesized upwelling as a key part of the *Fountain* corollary. **One of the primary goals of our proposed mission is to test this recent suggestion of mean downward motion in the Western Pacific tropopause region by measuring mixing ratio profiles of short lived tracers during N.H. winter** (the time of coldest temperature).
The energetic turrets of the **TTL Overshoot** penetrate the bottom of the TTL, but do not reach the true stratosphere (the overworld). Dehydration occurs rapidly by sedimentation of ice as air ascends vertically: rapid dehydration is essential because the overshooting turrets remain exceedingly cold for only brief periods of time. In contrast, the **TTL Cold Trap** theory hypothesizes that dehydration occurs much more slowly (e.g., over a period of several days) as air is advected horizontally (quasi-isentropically) through a standing cold pool. Consequently, stratospheric H\textsubscript{2}O is most likely sensitive to cloud microphysics according to the **TTL Overshoot** theory and to details of transport according to the **TTL Cold Trap** scenario. For both hypotheses, dehydration of air occurs prior to its arrival into the overworld.

Finally, the **Stratospheric Dehydration** theory was proposed in part to account for the observation that the minimum mixing ratio of stratospheric water often occurs 2 to 3 km above the tropopause in both the tropics and mid-latitudes (e.g., Kley et al., 1979). Mote et al. (1996) have shown, however, that this feature can also be accounted for by the seasonal cycle of tropopause temperature being “recorded” onto the vertical profile of H\textsubscript{2}O due to slow uplift in the tropical lowermost stratosphere.

Before offering a list of “Hypotheses of Stratospheric H\textsubscript{2}O Change”, we digress to consider how stratospheric H\textsubscript{2}O might respond to various forcings for each of the “Hypotheses of Stratospheric Aridity” defined above. This digression is meant to motivate the usefulness of pursuing tests of the various “Hypotheses of Stratospheric Aridity” as a first step for understanding the cause of recent changes to stratospheric H\textsubscript{2}O.

For the various hypotheses, we expect:

**Cold Trap** - stratospheric H\textsubscript{2}O will be most sensitive to tropopause temperature

**Convective Overshoot** - stratospheric H\textsubscript{2}O will be most sensitive to surface temperature and humidity (which determine turret height), the amount of ice lofted across the tropopause, perhaps ice physics, and only indirectly to tropopause temperature

**TTL Cold Trap** - stratospheric H\textsubscript{2}O will be most sensitive to temperature in the TTL, circulation patterns within the TTL, and transport of air into and out of the TTL.

**TTL Overshoot** - stratospheric H\textsubscript{2}O will be most sensitive to surface temperature and humidity (which determine turret height), perhaps ice physics, circulation patterns within the TTL, and transport of air into and out of the TTL, and only indirectly to tropopause temperature.

**Stratospheric Dehydration** - stratospheric H\textsubscript{2}O will be most sensitive to lower stratospheric temperature and transport process in the lower stratosphere.
3.3 Hypotheses of Stratospheric H$_2$O Change

The various “Hypotheses of Stratospheric Aridity” described above have different consequences for how stratospheric H$_2$O is likely to respond to changing climate. Understanding precisely how stratospheric aridity is maintained for a brief period in time is not necessarily sufficient for understanding how stratospheric H$_2$O may change over multi-year time periods. The proposal to conduct this mission over an extended period of time is motivated by the need to gain insight into the seasonal and interannual variations in the transport of H$_2$O within the TTL.

The following hypotheses have been proposed to explain the recent rise in stratospheric H$_2$O. For convenience of distinguishing these hypotheses from the above mentioned “Hypotheses of Stratospheric Aridity”, the numbering sequence starts with “6”:

6) **Rising Cold Trap Temperature** - Stratospheric H$_2$O is responding directly to a warming of the tropopause region (Evans et al., 1998).

7) **Increasing Asian Monsoon** - An increase in the extremity of the Asian monsoon over Indonesia leads to a moister upper troposphere, resulting in greater abundance of H$_2$O within the TTL for air parcels that do not pass directly through a cold trap (Evans et al., 1998; Smith et al., 2000).

8) **Changing Ice Physics** - Convective turrets become less efficiently dehydrated over time due to subtle changes in the microphysics of ice (an adaptation of Sherwood and Dessler, 2000).

9) **Reduced Brewer-Dobson Circulation** - Stratospheric wave activity in the NH is reduced, resulting in increased fraction of air entering the TTL during NH summer (the warm tropopause period) (this white paper).

The radiosonde analyses presented by Simmons et al. (1999) appear to contradict the **Rising Cold Trap Temperature** theory. Smith et al. (2000) present analyses of HALOE data that indicate a considerable moistening of the tropical upper troposphere in September-October-November between 1992 and 1999 that support the **Increasing Asian Monsoon** theory. However, it is an open question whether or not this moistening of the upper troposphere has a direct effect on stratospheric H$_2$O. It is unclear why or how the microphysics of ice would change from year to year, and Sherwood and Dessler (2000) mention the sensitivity of stratospheric H$_2$O to ice physics only “in passing”. The extreme year-to-year variability in the extent of denitrification of the Arctic polar vortex suggests ice physics could play a role in year-to-year variations of stratospheric H$_2$O.

Finally, the **Reduced Brewer-Dobson Circulation** theory is offered as another mechanism for increasing stratospheric H$_2$O. It is conceivable that rising concentrations of greenhouse gases may have induced a reduction in the “extratropical pump” (Holton et al., 1995) altering the Brewer circulation for essentially the same reasons as discussed by
Shindell et al. (1998) for the polar vortex extension of this circulation. According to the framework of Yulaeva et al. (1994), a slow down of the Brewer-Dobson circulation should be associated with tropopause cooling. However, it is unclear whether tropopause temperature is governed more strongly by local thermodynamics (e.g., Kirk-Davidoff et al., 1999) or by large-scale transport. Stratospheric H₂O is likely the result of an admixture of processes that occur mainly during NH winter but also somewhat during summer. A slight increase in the relative contribution of summertime processes (when temperature in the tropical upper troposphere is warmer) to resulting overworld air, perhaps due to a change in the seasonal variation of the Brewer-Dobson circulation, could contribute to the recent rise in stratospheric H₂O.

3.4 The Importance of Isotopic Measurements for testing the hypotheses

Measurements of the isotopic composition of water vapor comprise a key aspect of our proposed strategy for testing the various hypotheses of stratospheric aridity described above. Here we describe how measurements of water vapor isotopic composition will be used. The material in this section is based largely on the work of Moyer et al. (1996) and Webster et al. (2000).

The isotopic composition of H₂O records the condensation and evaporation history experienced by air parcels. In thermodynamic equilibrium, condensates of H₂O are isotopically enriched (e.g., heavier) relative to gas phase H₂O. Consequently, as water condenses and precipitates from an ascending air parcel, the residual vapor becomes progressively lighter. The abundance of HDO relative to H₂O (termed $\delta^D$) and of H$_2^{18}$O relative to H₂O (termed $\delta^{18}$O) can be calculated for an ascending air parcel using fractionation factors between ice and vapor based on laboratory measurements (Merlivat and Nief, 1967; Majoube, 1970), assuming an ambient temperature profile from observation and that $\delta^D$ and $\delta^{18}$O at cloud base equals that of the sea surface (e.g., Moyer et al., 1996; Keith, 2000; Johnson et al., 2000).

Moyer et al. (1996) showed that the expected value of $\delta^D$ at 100 mbar in the tropics is about -900 per mil (Figure 3.5). This calculation assumed thermodynamic equilibrium, desiccation to observed levels of stratospheric aridity, and no transport of condensed material across the tropopause. However, ATMOS measured $\delta^D$ of -670 per mil in the tropical lower stratosphere (Moyer et al., 1996). Johnson et al. (1995), using balloon-borne thermal emission infrared spectrometer measurements obtained at mid-latitudes, estimated a similar value of $\delta^D$ for air entering the stratosphere. Minor contributions to HDO from oxidation of CH₃D have been accounted for in both estimates of $\delta^D$.

The observations of $\delta^D$ appear to be inconsistent with the Cold Trap hypothesis of stratospheric aridity (Moyer et al., 1996). Moyer et al. (1996) suggested two possible explanations of the measurements. First, air parcels in the tropical lower stratosphere may receive contributions to their H₂O budget from isotopically enriched ice lofted across the tropopause that later evaporates (the Convective Overshoot - Ice Lofting hypothesis discussed above). The second possibility is that the actual isotopic
fractionation that occurs in the atmosphere may be weaker than the equilibrium value, which would occur during ice deposition for highly supersaturated conditions (e.g., saturation ratios of 150 to 200%) (the **Convective Overshoot - Supersaturation** hypothesis discussed above).

Keith (2000) extended the work of Moyer et al. (1996) and noted two key points. First, the kinetic fractionation factors used by Moyer et al. (1996) are based on laboratory measurements obtained for temperatures of 240 K and above. Results are extrapolated to colder temperatures. Keith (2000) states “it seems unlikely that errors in the isotopic physics are of sufficient magnitude to resolve the HDO puzzle”. Nonetheless, laboratory measurements must be obtained at colder temperature to better interpret the isotopic measurements proposed here. The second point raised by Keith (2000) is the illustration that an estimate of δ^{18}O for air entering the stratosphere from the mid-latitude balloon measurements of Johnson et al. (2000), when combined with the δD measurements of Moyer et al. (1996) and Johnson et al. (1995), appear to rule out the **Convective Overshoot - Supersaturation** hypothesis of stratospheric aridity. Thus, the **Convective Overshoot - Ice Lofting** theory is the only Class I hypothesis that is consistent with the limited isotopic measurements of H_{2}O.

There are many caveats to the conclusions noted above. The ATMOS measurements of HDO in the tropics are sparse and do not extend to the upper troposphere. Continuity of profiles of δD across the tropopause is essential. The FIRS estimates of δD and δ^{18}O of air entering the stratosphere are based on measurements obtained only at mid-latitudes. Measurements of H_{2}^{18}O in the tropics are needed. Current theoretical models of δD and δ^{18}O only treat stratospheric dehydration in one dimension (e.g., the Class I hypotheses described above). The consequences of condensation, lofting, and mixing within the hypothesized TTL must be incorporated into a more sophisticated theoretical treatment of δD and δ^{18}O. Although the existing observations appear to support the **Convective Overshoot - Ice Lofting** hypothesis, more atmospheric measurements of δD and δ^{18}O are needed. For example, the current remote observations of δD and δ^{18}O have insufficient vertical resolution to address the TTL “Class II” hypotheses of stratospheric aridity.

The discussion so far has focused only on differences in vertical profiles of δD and δ^{18}O predicted by the various “Class I” hypotheses of stratospheric aridity. However, the three “Class II” (e.g., three-dimensional) hypotheses will lead to differences in spatial patterns of δD and δ^{18}O as well. The **TTL Cold Trap** hypothesis should result in considerable horizontal gradients in δD and δ^{18}O as air flows through the cold trap, and relatively gentle vertical gradients across the lower and upper boundary of the TTL. The **TTL Overshoot** hypothesis would be consistent with strong vertical gradients in δD and δ^{18}O at the lower boundary of the TTL, gentle horizontal gradients within the TTL, and gentle vertical gradients near the top of the TTL. The **Stratospheric Dehydration** hypothesis will predict gentle horizontal and vertical gradients in δD and δ^{18}O until truly stratospheric air is encountered.

Perhaps the strongest use of measurements of δD and δ^{18}O, which has not yet been explored in the literature, will be the potential to quantify the effects on H_{2}O of both
thermodynamics and mixing for air parcels with different temperature histories within the hypothesized TTL. Correlation plots of $\text{H}_2\text{O} \text{ vs } \delta D$ and $\text{H}_2\text{O} \text{ vs } \delta^{18}\text{O}$ will be highly non-linear for individual ascending air parcels. Therefore, admixtures of slowly ascending air masses that detrain at different potential temperatures within the TTL (e.g., Sherwood and Dessler, 2000) may best be identified by the appearance of "mixing lines" (e.g., Waugh et al., 1997) in correlation plots of $\text{H}_2\text{O} \text{ vs } \delta D$ and $\text{H}_2\text{O} \text{ vs } \delta^{18}\text{O}$. Since $\delta D$ and $\delta^{18}\text{O}$ essentially preserve a memory of thermodynamic changes in ambient $\text{H}_2\text{O}$ that is strongly dependent on the coldest recent temperature, correlation plots of $\text{H}_2\text{O} \text{ vs its isotopes may ultimately prove to be the best way to quantify the effects on } \text{H}_2\text{O} \text{ of both thermodynamics and mixing."

3.5 Testing the Hypotheses

The observations of $\text{H}_2\text{O}$ and temperature that will be obtained by HIRDLS, MLS and TES in the tropical tropopause region will provide important information on the longitudinal variability of conditions in the upper troposphere and lower stratosphere. The cirrus ice measurement from MLS may provide important constraints on the details of whether significant amounts of precipitable water vapor are lofted across the tropopause. Space-borne observations of $\text{H}_2\text{O}$ will also be obtained in the future by SAGE III. To our knowledge, TES is the only upcoming space-based instrument that will attempt to measure HDO. Other satellites payloads may have some capability (e.g. SCIAMACHY) but have not yet focused their efforts on isotope retrievals.

Our mission has four primary goals for testing hypotheses of both stratospheric aridity and hypotheses of long-term changes in stratospheric $\text{H}_2\text{O}$:

A. Testing the accuracy of the AURA (and other satellite) measurements of $\text{H}_2\text{O}$ and temperature in the tropical tropopause region;

B. Determination of the vertical and horizontal structures of fields of $\text{H}_2\text{O}$, HDO, and temperature in the tropics and whether these structures are suitably resolved by the satellite instruments;

C. Testing the accuracy of the MLS cirrus ice measurement;

D. Measurement at high vertical resolution of the isotopic composition of $\text{H}_2\text{O}$ and certain tracers of vertical motion that have a wide range of lifetimes (e.g., SF$_6$, CO$_2$, CO, CH$_3$I, radon) to complement the satellite measurements of $\text{H}_2\text{O}$, HDO, and other longer-lived tracers.

Note: we have not addressed at all in this draft white paper the issue of the local radiative balance in the tropics. Clearly this is an important issue. We hope to receive feedback from the community regarding the incorporation of measurements of the radiative field into the framework of this proposed mission.
The importance of accurate measurements of H\textsubscript{2}O for testing the various hypotheses of stratospheric aridity is obvious. It is essential for the achievement of goal A that the correlative measurements be obtained in the tropics. A variety of correlative measurement techniques will likely be required. Lidar measurements of H\textsubscript{2}O and microwave temperature profiler measurements of temperature will provide greatest flexibility for obtaining close coincidence in space and time. Airborne in situ measurements of temperature will provide the greatest possible accuracy. Airborne in situ measurements of H\textsubscript{2}O using existing instruments will provide a capability for continuity between AURA and UARS measurements of H\textsubscript{2}O, since there are numerous coincidences between HALOE and MLS measurements of H\textsubscript{2}O and observations from previous in situ campaigns. Balloon measurements of H\textsubscript{2}O and temperature in the tropics are highly desirable because satellite measurements of H\textsubscript{2}O above the cruise altitude of either the ER-2 and WB-57 are of interest (e.g., for assessing the seasonal cycle of H\textsubscript{2}O) and it is not clear the lidar measurements will be sufficiently accurate at these altitudes to be the sole source of validation.

For the achievement of goal A), it will be highly desirable to continue to explore the cause of lingering differences between measurements of H\textsubscript{2}O by various in situ instruments (differences sometimes as high as 15\%). Laboratory intercomparisons of the various in situ instruments should be planned as part of this mission.

Goal B: Testing theories of stratospheric aridity will require accurate knowledge of the vertical and longitudinal variability of H\textsubscript{2}O and HDO in the tropics. Determination of these vertical and horizontal structures, and whether these structures are suitably resolved by the space-based instruments, necessitates obtaining correlative observations over both the Western Pacific (cold tropopause) and the Eastern Pacific (warmer tropopause). It is critical that we assess how well the various satellite instruments can resolve the shape of the vertical profile of H\textsubscript{2}O in the regions where we expect the largest vertical gradients and the minimum abundance of H\textsubscript{2}O to occur (e.g., the Western Pacific). Understanding thermodynamics and transport within the TTL will require testing the ability of the satellite instruments to distinguish subtle differences in composition between the poleward subtropical jet and the equatorial easterly region of the tropical anti-cyclonic flow in the Western Pacific. Consequently, we propose a series of airplane flights from Guam (13.4° N, 144.4° E) and Hawaii (21.3° N, 157.9° W) to provide access to all of the regions of interest.

It is important that we assess how well the various satellite instruments can resolve the shape of the vertical profile of H\textsubscript{2}O in the regions where the minimum abundance of H\textsubscript{2}O is expected to occur (e.g., the Western Pacific). The importance of this validation activity is illustrated by a comparison of vertical profiles of H\textsubscript{2}O obtained by HALOE and an in situ hygrometer in the tropics during October 1994. Figure 1 of Mote et al. (1996) revealed differences of nearly 0.7 ppmv in the minimum mixing ratio of H\textsubscript{2}O (the HALOE measurement of H\textsubscript{2}O exceeded the in situ observation) that appears attributable to the finer altitude resolution of the in situ measurement. This difference compromises the ability of HALOE to accurately identify the H\textsubscript{2}O mixing ratio content of air that enters the stratosphere. Since one of the strongest tests of the various hypotheses of
stratospheric aridity is the relation between minimum tropopause temperature (or minimum TTL temperature) and the H2O mixing ratio of newly minted stratospheric air, it is essential that our validation activities be focused on assessing the reliability of the space-borne measurements of minimum H2O and temperature.

It is important also to obtain measurements of H2O and temperature in the NH summer as well as during the NH winter (coldest tropopause) because the mechanisms for cross tropopause transport may be distinctly different. The humidity of the stratosphere reflects the mixing ratio of water entering the stratosphere weighted by the mass flux through the tropical tropopause. Although ascent is significantly slower in NH summer than during NH winter (e.g., Rosenlof et al., 1997), the much higher water mixing ratio at the tropopause yields a mass flux of water that significantly humidifies the stratosphere. This is seen in the seasonal cycle of water in the region of tropical ascent (Mote et al., 1996). Stratospheric humidity may thus be influenced by interannual changes in the seasonal variation of mass flux through the tropopause, independent of any change in the mixing ratio of water at the tropical tropopause.

The importance of isotopic measurements of H2O for testing hypotheses of stratospheric H2O is described in the previous section. The only H2O isotopic measurement from EOS Aura will be profiles of HDO provided by TES. For validation of this measurement, the existing balloon-borne remote measurement capability is furthest developed. Profiles of HDO have been published by the FIRS group (Johnson et al., 2000). The MkIV instrument can measure profiles of HDO with better precision than ATMOS owing to the longer integration time from a balloon platform (G.C. Toon, private communication, 2000). While balloon-borne observations of HDO should play an important role in testing the accuracy and vertical resolution of the TES observation, these measurements can not be obtained in the Western Pacific region unless a water-landing capability for the gondola is developed.

Various options are available for air-borne measurements of H2O isotopes. A tunable diode laser instrument for measurement of HDO and H218O from the WB-57 is currently under development at JPL (Webster et al., 2000). Measurements of HDO and H218O can also be obtained using an in situ mass spectrometer but we are unaware of any such instrument development for either the ER-2 or WB-57 platforms. Owing to the critical importance of water isotopes for testing the various hypotheses, we suggest that both balloon-borne remote and in situ sampling of H2O isotopes in the tropics be conducted for this mission. The airborne observations will be especially important for defining vertical and horizontal structures in the Western Pacific that will be an important new tool for testing the various hypotheses of stratospheric aridity.

Goal C. The MLS measurement of the ice content of cirrus is a potentially powerful observation for testing the importance of “ice lofting” within both the Class I and Class II hypotheses for stratospheric aridity described above. The primary concern, however, is whether the satellite measurement will provide sufficient vertical resolution. At present, we do not have a means for testing this space-borne observation from either the ER-2 or WB-57 platforms. Such a test would require the development of a new instrument that
could distinguish gas phase from condensed phase H$_2$O. The desired correlative measurement could be achieved by development of an inlet for one of the existing in situ H$_2$O instruments that could, at various times, either include or exclude particulates. Since the existing isotopic measurements of H$_2$O described above appear to suggest that ice lofting is truly important for supplying H$_2$O to the stratosphere, the development of a new in situ capability for measuring condensed phase water should be given high priority both for validation of the MLS measurement of cirrus ice and for fully testing the various hypotheses of stratospheric aridity.

Goal D. The proposed measurements are designed to complement the space-borne observations for further testing of the hypotheses of both stratospheric aridity and hypotheses of long-term changes in stratospheric H$_2$O. Clearly the satellite measurements of H$_2$O, temperature, long-lived tracers of atmospheric transport will be critical for resolving longitudinal gradients throughout the tropics, variations in stratospheric humidity with respect to both altitude and latitude, as well as possible long-term changes in H$_2$O and temperature. Our strategy is to augment the space-borne observations with high vertical and horizontal resolution of a host of other species in the specific region where it has been hypothesized that stratospheric aridity is controlled.

The usefulness of the proposed isotopic measurements for testing the various hypotheses of stratospheric aridity is discussed in Section 3.4.

Measurement of a suite of gases with short photochemical lifetimes (e.g., CH$_3$I, acetone, Ra, $^{210}$Pb, and/or $^{222}$Rn) will provide a strict test of whether air is sinking over the cold Western Pacific, as has been proposed by Gettelman et al. (2000) and Sherwood (2000). Vertical profiles of CO provide quantitative measurements of ascent rates on longer time scales (Herman et al., 1999). Tracer-tracer correlation plots of a suite of tracers with a variety of lifetimes can be used to place additional quantitative constraints on transport in the tropical TTL region (e.g., Flocke et al., 1999).

In situ measurements of the vertical structures of gas phase and particulate H$_2$O at spatial scales that can not possibly be achieved by satellite measurement (e.g., 100 m resolution) will be critical for testing details of the various hypotheses of stratospheric aridity described in Section 3.2. Testing the Class I and Class II Overshoot hypotheses may prove quite difficult using only space-borne observations because of the rapid variation of H$_2$O on small spatial scales (e.g., Sherwood and Dessler, 2000). The TTL Cold Trap hypothesis predicts that lower levels of H$_2$O will be seen downwind of the standing cold pool than upwind of this region. While the satellite observations should be able to resolve such structures, the proposed simultaneous measurement of the numerous short-lived tracers mentioned above (not measured by satellite) will provide an estimate of the “age” of air parcels in the TTL. This information will be key for unraveling the relationship between details of the transport field and desiccation. Finally, measurements of the size distribution of the particulates, using spectrometers based on existing technology that are focused on several-micron size particles, will enable rates of dehydration to be estimated. This will provide a test of another important difference
between the TTL Cold Trap hypothesis (relatively slow desiccation) and the TTL Overshoot hypothesis (relatively rapid dehydration).

Finally, the candidate H₂O instruments (both aircraft and balloon) for this mission have obtained numerous coincident measurements of H₂O with the existing MLS and HALOE instruments on UARS and with SAGE II. Obtaining additional coincidences close in space and time with the various EOS Aura instruments may prove crucial for “bridging the gap” between Aura and UARS and between Aura and SAGE II for studies of long-term changes in stratospheric H₂O, especially if UARS and/or SAGE II are not operational at the time Aura turns on.

4. Photochemistry of the Tropical Lower Stratosphere and Upper Troposphere

4.1 Introduction

The chemical properties of tropical transition layer define the chemical boundary condition for the stratosphere. Despite a decade of tremendous progress in tropospheric and stratospheric chemistry, the photochemistry of this region remains poorly constrained. This limits our ability to understand the fates of numerous compounds (and their degradation products) transported into the near tropopause region. Such knowledge is required if we are to understand the impact of climate change on the stratosphere in general and ozone in particular.

Until recently, the chemical precursors of the stratospheric radicals and aerosol, with the notable exception of water vapor, were thought to be compounds with long tropospheric lifetimes. This greatly simplified defining the chemical boundary condition for the stratosphere because globally-averaged surface measurements of these long-lived compounds could be used. For example, sulfur was thought to be carried solely by OCS, nitrogen by N₂O, and halogens by the relatively long-lived halocarbons. It has become increasing clear, however, that short-lived compounds transported to the tropopause region of the tropics significantly alter the chemistry of the global stratosphere. The amount of OCS transported across the tropopause accounts for no more than half of the sulfur aerosol present in the lower and middle stratosphere (e.g., Weisenstein et al., 1997). Bromine monoxide concentrations in the lower stratosphere appear to reflect the input of very short-lived bromine containing organic and perhaps inorganic compounds (e.g., Ko et al., 1997). The concentration of reactive nitrogen, NO₃, and ozone are non-zero at the tropical tropopause (Strahan, 1999).

Because the upper troposphere in the tropics is arid and cold, until recently, it was considered to be photochemically inactive. The production rate of OH was thought to be determined by the local concentrations of ozone and water vapor. This rate was calculated to be very low both because the yield of O(^1D) in the photolysis of ozone was thought to be small due to the temperature dependence of the quantum yield and because the efficiency of OH production by O(^1D) is reduced at the low humidity. Observations of OH in the upper tropical troposphere have significantly altered this picture (Wennberg
et al., 1997). The measured abundance of OH between 10 km and the tropopause is nearly twice that calculated. It has been suggested that the higher than expected levels of OH result from the transport of short lived HOx precursors into the arid upper troposphere. Oxygenated hydrocarbons such as acetone (Singh et al., 1995) and peroxides (Jaegle et al., 1997) have been suggested as possible sources. Nevertheless, a closure experiment in the tropical upper troposphere has not been accomplished. Independent of the source of these free radicals, the larger concentration of HOx alters the calculated lifetime of CO, SO2, and other compounds and may increase the production rate of ozone in the UT.

The short lifetimes of these compounds significantly impact the design of the TC3 campaign. Because the source strengths of these compounds are likely not steady, reflecting the variability in both the chemistry and the transport from the lower troposphere, the sampling strategy must be optimized to assure it is representative. Satellite observations are key to understanding how representative the in situ data set is of the variable dynamics in the tropics.

Finally, the tropical lower stratosphere offers a unique laboratory to study the photochemical lifetimes of short-lived compounds. This is one of the few regions of the atmosphere where the chemical and dynamical processes are separable on seasonal timescales. Herman et al. (1999) and Weinstock et al. (2000) have shown, for example, that the lifetime of CO can be determined empirically by measurement of the decrease in its mixing ratio with height above the tropical tropopause. The ascent rate can be accurately constrained by coincident CO2 and H2O measurements and the influence of mixing from the subtropics can be accounted for by observations of very long-lived tracers such as N2O. Dessler et al. (1996) have shown that the ozone production rate can also be determined in this manner. This analysis can be extended to any compound with tropospheric sources and irreversible sinks in the lower stratosphere. Target molecules include non-methane hydrocarbons and oxygenated hydrocarbons (such as acetone), PAN, SO2, H2S, etc.

Clearly a suite of measurements of short-lived compounds in the tropical upper troposphere and lower stratosphere will greatly improve our understanding of the chemistry and dynamics of this critical region. Below we outline a set of hypotheses that can be tested through this approach.

4.2 Hypothesis - Sources of Stratospheric Bromine:

Short-lived bromine containing compounds enhance significantly the concentration of bromine monoxide in the upper troposphere and lower stratosphere. As a result, ozone loss rates in the lower stratosphere are significantly faster than expected.

It has become increasing clear that the bromine budget of the stratosphere can not be closed by consideration of longer-lived bromine-containing halocarbons alone. Observations in the lower stratosphere demonstrate that, in the youngest air, bromine monoxide concentrations are significantly larger than expected (Figure 4.1) (Wamsley et
al., 1998). This result is also seen in balloon-borne solar occultation measurements (Pundt et al., 1997, Pfeilsticker et al., 2000) and ground-based slant column observations of BrO (Friess et al., 1999). These bromine monoxide measurements can be explained by assuming a stratospheric boundary condition at the tropical tropopause of 3 to 5 pptv Br$_2$. It is unclear, however, what molecule or molecules carry this additional bromine. Suggestions have included very short-lived bromo-carbons such as bromoform (CHBr$_3$) (Dvortsov et al., 1999; Schauffler et al., 1999; Sturges et al., 2000) as well as the possibility of direct input of Br$_y$ (BrO + HOBr + BrONO$_2$) (Ko et al., 1997; Pfeilsticker et al., 2000). Clearly, understanding the budget and potential trends in bromine require knowledge of the mechanisms for cross tropopause transport. Bromine is an active catalyst in the lower stratosphere accounting for a significant fraction of the chemical ozone loss. Bromine catalysis would account for nearly half of the total photochemical sink of O$_3$ between 14 and 18 km in the mid-latitude stratosphere if the abundance of Br$_y$ is indeed 3 pptv at the tropical tropopause (Figure 4.2).

4.2.1 Testing strategy:

Measurements of bromine monoxide and the largest possible suite of precursors (e.g. Halons, CH$_3$Br, CH$_2$Br$_2$, CHBr$_3$, etc.) in the region of the tropical tropopause will allow closure of the stratospheric bromine budget. Because short-lived compounds are likely involved, a single profile in the tropics will not be adequate. Observations must be obtained in various geographical regions and during several seasons to assure representative sampling. Observations across the subtropical jet will enable better understanding of bromine in the lowermost stratosphere. Measurements of BrO in the upper troposphere with a precision of at least 1 pptv are necessary to constrain the direct input of Br$_y$ to the stratosphere. Aura MLS, SAGE-III, and SCIAMACHY limb observations of BrO can be both validated and used to extend this analysis to the global scale.

4.3 Hypothesis - Iodine and stratospheric ozone:

Iodine monoxide is an important catalyst for the destruction of stratospheric ozone. Solomon et al. (1994) suggested that iodine monoxide (IO) may be an important free radical in the lower stratosphere. Since there are no long-lived iodine containing compounds, any iodine reaching the stratosphere must be carried by short lived organic and inorganic compounds or possibly on aerosol. The catalytic activity of iodine is due mostly to the reactions of IO with HO$_2$. Because this reaction rate is 3-4 times faster than the corresponding rate of BrO, 1 pptv of IO contributes nearly as much as 3 pptv of BrO to the loss rate of ozone in the lower stratosphere.

The stratospheric burden of iodine remains uncertain. Column observations by Wennberg et al. (1997), illustrated in Figure 4.3, suggested that about 0.1 pptv of IO was present in the lower stratosphere (assuming a column weighted near 120 mb). Pundt et al. (1998) obtained an upper limit of 0.1 pptv of IO at 15 km from balloon-borne observations. Recently, however, Wittrock et al. (2000) have observed significant column abundances
of IO in ground-based observations obtained at high latitude. They attribute this signal to a stratospheric mixing ratio of IO equal to 0.65 to 0.8 pptv (Figure 4.4). Consistent with this result, K. Pfeilsticker's group have observed IO in solar transmission spectra obtained by a balloon-borne instrument at float (30-40 km). The retrievals are not yet complete but suggest IO densities similar to Wittrock et al. (K. Pfeilsticker, personal communication, 2000). It remains unclear whether the different results obtained by these researchers represent errors in the earlier measurements of Wennberg et al. and Pundt et al. or uncertainty in the photochemistry of stratospheric iodine. Recently, Bedjanian et al. (1998) have shown that OIO is produced with nearly unit efficiency in the reaction of BrO with IO. Depending on the photochemistry of OIO, this result may significantly alter the fraction of iodine present as IO. If the photolysis of OIO is slow, as suggested by Cox et al. (1999) and partially confirmed by Crowley et al. (2000), the fraction of iodine present as IO in the lower stratosphere may be considerably smaller than earlier models suggested (Solomon et al., 1994; Wennberg et al., 1997). This fraction may also vary significantly depending on the chemical sinks of this compound (with ClO for example).

4.3.1 Testing strategy:

Measurements of iodine monoxide and the largest possible suite of precursors (e.g., CH$_3$I, CH$_2$I$_2$, C$_2$H$_3$I, CF$_3$I) in the region of the tropical tropopause will allow closure of the overworld iodine budget. Because short lived compounds are likely involved, a single profile in the tropics will not be adequate. Observations must be obtained in various geographical regions and during several seasons to assure representative sampling. Observations across the subtropical jet will enable better understanding of iodine in the lower most stratosphere. SAGE-III and SCIAMACHY limb observations of IO can be both validated and used to extend this analysis to the global scale.

4.4 Hypothesis - PAN, Oxygenated Hydrocarbons, and O$_3$ production in the UT:

Oxygenated VOC photolysis both increases HO$_x$ levels and promotes the formation of PAN in the upper troposphere. The enhanced PAN formation significantly reduces the NO$_x$/NO$_y$ ratio. This reduces the ozone formation rate in the upper troposphere and increases the input of NO$_y$ to the stratosphere.

Singh et al. (1995) reported the discovery of high concentrations of oxygenated hydrocarbons in the remote troposphere. One of these compounds, acetone, is expected to significantly influence the photochemistry of the upper tropical troposphere (McKeen et al., 1997). In particular, the photolysis of acetone is a source of odd-hydrogen radicals that can dwarf the traditional O$_3$/H$_2$O source (Figure 4.5) (Singh et al., 1995; Wennberg et al., 1997). Coincident HO$_x$, acetone, and peroxide measurements have never been obtained in the tropical upper troposphere. Because the lifetime of acetone is quite short (10 days), extrapolation of the observations obtained in mid latitudes to the TTL is fraught with uncertainty.
In addition to the production of HO\(_x\) radicals, acetone and other higher aldehydes produce the acetyl radical upon photolysis. Peroxyacetyl nitrate (PAN) formation will occur in the presence of NO\(_2\). The formation of PAN has been suggested to significantly reduce the NO\(_x\) levels in the UT (Keim et al., 1999; Folkins et al., 2000). PAN is not soluble and has a calculated photochemical lifetime in the UT/LS of 1 to 2 months and therefore will likely be efficiently transported across the tropical tropopause. Observations of NO\(_x\) and NO\(_y\) in the tropics are quite limited. During ASHOE/MAESA and STRAT, NO\(_y\) was measured to be ~200-400 pptv at the tropical tropopause. Typically, NO comprised 30-60% of the budget of NO\(_y\).

4.4.1 Testing strategy:

Measurements of NO, NO\(_2\), NO\(_y\), HNO\(_3\), HNO\(_4\), PAN, acetone, and other oxygenated organic compounds in the TTL will provide a critical test of this hypothesis. Because the tropospheric sources of these compounds are inhomogeneous, a single profile in the tropics will not be adequate. Observations must be obtained in various geographical regions and during several seasons to assure representative sampling. TES and HIRDLS are expected to retrieve many of these compounds (e.g. NO\(_2\), HNO\(_3\), PAN) and the aircraft observations can provide validation of the retrievals. AURA and other satellite observations of these compounds can be used to extend the higher spatial resolution \textit{in situ} observations to the global scale.

4.5 Hypothesis - The stratospheric sulfate aerosol layer:

\textit{Short-lived sulfur containing compounds contribute significantly to the background stratospheric aerosol.}

Understanding the mechanisms that determine the aerosol surface area in the lower stratosphere during volcanically-quiescent times is critical for understanding the interactions of the free radical families and therefore ozone. Recent studies have pointed out that the OCS source is insufficient to explain the background stratospheric aerosol (e.g., Weisenstein et al., 1997; Kjellstrom, 1998; Karcher and Solomon, 1999). Other possible organic sources of stratospheric sulfur are CS\(_2\), dimethyl sulfide (DMS), methyl sulfonic acid (MSA). Observations of aerosol number and volume near the tropical tropopause suggest significant aerosol mass is advected through the tropical tropopause (Brock et al., 1995). This result strongly suggests the existence of a TTL. In addition, Murphy et al. (1998) have shown a veritable zoo of particle types in the tropopause region. A 2D model calculation based on a recently determined upper limit for the UV absorption cross section of H\(_2\)SO\(_4\) significantly underestimates the sole atmospheric profile of SO\(_2\) obtained under quiescent conditions (\textit{Figure 4.6}) (Burkholder et al., 2000). This comparison suggests the occurrence of an undetermined mechanism for conversion of H\(_2\)SO\(_4\) to SO\(_2\) or perhaps the transport of significant amounts of SO\(_2\) or organic sulfur species other than OCS across the tropical tropopause. These studies point to unique and interesting aspects of gas phase sulfur and sulfate aerosol chemistry that can be addressed in the tropical upper troposphere and lower stratosphere.
4.5.1 Testing strategy:

Measurements of aerosol properties (e.g., particle size, composition, and the number of condensation nuclei) as well as the abundance of both organic and inorganic precursors of stratospheric sulfur in the TTL will provide a critical test of this hypothesis. The measurements should be focused on improving our understanding of both the chemical and physical processes responsible for aerosol formation in the stratosphere. Profiles of OCS, H$_2$SO$_4$, and SO$_2$ can be measured by the balloon–borne MkIV spectrometer and would be most useful if observed in the tropics. AURA and other satellite observations of aerosol extinction at numerous wavelengths will provide global physical and perhaps chemical characteristics of stratospheric aerosols. The proposed mission would provide validation for these aerosol measurements near the tropical tropopause. The Aura platform will only be able to measure profiles of SO$_2$ under volcanic conditions. If a major eruption were to occur soon after launch of EOS Aura, a flight of MkIV would provide important validation for profiles of SO$_2$, focused of course on a different “hypothesis” than offered here.

5. Tropical Upper Tropospheric Water Vapor

5.1 Introduction

The response of the hydrological cycle to greenhouse gas induced climate forcing is perhaps the single most important source of uncertainty in predicting future changes to Earth’s climate and composition. The small amounts of water vapor in the upper troposphere (UT) exert enormous leverage on the radiative balance and composition of the lower atmosphere. Increases in upper tropospheric H$_2$O within global climate models result in much greater radiative effects at the surface than are caused solely by the buildup of CO$_2$ and other greenhouse gases (e.g., Shine and Sinha, 1991; Ramanathan and Collins, 1991). Of particular importance is the moisture in the dry subtropical regions, which has a large cooling effect on the whole tropics. As the radiation budget is more sensitive to fluctuations for very dry air than for very moist air, changes in subtropical water vapor could have a profound effect on Earth's climate (Spencer and Braswell, 1997). It is therefore important to understand what processes control the subtropical humidity and how climate change will affect these processes. These are key issues in determining the nature of the water vapor feedback on climate.

The standard picture of the tropical troposphere is of large areas of gentle downwelling punctuated by isolated convective regions of extremely rapid ascent, which make up only a small fraction of the total area (only around a third of tropics is subject to direct moistening by nearby deep convection). The outflow from these convective regions is largest around 200 hPa (base of TTL), and as this air subsides the water vapor mixing ratio relaxes to very low values (10 ppmv or less). Observations show that the sub tropics are dry, but not as dry as this simple picture would imply (e.g., Pierrehumbert, 1998).
Hence there must be an additional moisture sources that hydrate the regions of the tropics characterized by descent.

5.2 Hypotheses for supply of moisture to the subtropical free troposphere:

1. Evaporation of precipitation:
   
   Evaporation of hydrometeors provides the main moisture source for the subtropics (Sun and Lindzen, 1993).

2. Evaporation of detrained cloud particles:
   
   Evaporation of dissipating clouds provides the main moisture source for the subtropics.

3. Lateral Transport
   
   Outflow from tropical convective regions occurs over a deep layer (i.e. not restricted to altitudes above 200 hPa) and lateral transport by the large-scale wind field over this layer provides the additional supply of moisture (Pierrehumbert, 2000 and references therein).

Which of the above hypotheses are correct, or more realistically the relative contribution of the three sources to subtropical moisture, has major implications on how subtropical humidity will change in response to climate warming (and hence the water vapor feedback on climate) (e.g., Held and Soden, 2000; Pierrehumbert, 2000). This also has implications on requirement for climate models to accurately simulate tropospheric water vapor and water vapor feedbacks, i.e. the first two hypotheses require accurate representation of microphysical processes, whereas for the third hypothesis accurate representation of large-scale winds and in particular transient wave activity is most important (e.g., Pierrehumbert and Roca, 1998). For the Lateral Transport hypothesis, it is also important to establish from what level(s) in the atmosphere the advection of H$_2$O occurs.

There is compelling evidence that lateral transport is a significant source for subtropical moisture. In particular, simulations of water vapor distribution using analyzed winds are generally in good agreement with observations suggesting that the tropical humidity distribution in the free troposphere can be understood (at least to first order) by following air parcel trajectories backwards to their “temperature of last saturation” (e.g., Sherwood 1996; Salathe and Hartmann, 1997; Pierrehumbert, 1998; Pierrehumbert and Roca, 1998; Dessler and Sherwood, 2000). These simulations indicate that upper tropospheric water vapor distribution will contain many fine-scale features, with moist and dry plumes intertwined, and it has been suggested that the northern subtropics can be viewed as background of dry air interrupted by a few moist plumes (Pierrehumbert, 1998), see
Figure 5.1. Note that fine-scales exist not only in the horizontal, but also in the vertical due to differential advection that creates thin layers as in the stratospheric surf zone.

5.2.1 Testing strategy:

TC\textsuperscript{3} has three major foci for testing the hypotheses of UT water vapor:

A. confirmation of the accuracy of the AURA (and other satellite) measurements of H\textsubscript{2}O and MLS measurements of cirrus ice in the tropical and subtropical UT;

B. determination of the horizontal and vertical scale of structures in UT H\textsubscript{2}O field;

C. measurements of the isotopic composition of H\textsubscript{2}O, together with measurements of tracers with a range of lifetimes, that will help determine the origin of upper tropospheric water vapor (and the relative contribution of above hypotheses to subtropical moisture budget).

Given the importance of water vapor in the radiative budget it is crucial to accurately know the water vapor distribution. Because of lack of data, however, there is currently uncertainty regarding the actual abundance of water vapor in the UT. Measurements from AURA (and other future satellites) will provide measurements of H\textsubscript{2}O in this region, and will greatly improve the situation. However, aircraft measurements in both moist and dry regions are required for validation of the satellite observations of H\textsubscript{2}O.

The MLS measurement of cirrus ice will provide important constraints on the hypotheses for supply of moisture to the subtropical free troposphere. Development of a new in situ capability for measurement of condensed phase H\textsubscript{2}O should be given high priority both for validation of the MLS measurement and for testing these hypotheses on spatial scales (e.g., high vertical resolution) that are inaccessible to the satellite.

Addressing focus B will be an important part of the satellite validation. Because fine-scale structures are ubiquitous in UT H\textsubscript{2}O this variability will need to be considered when validating satellite measurements (with aircraft and other measurements), and also when interpreting the satellite measurements (in particular, measurements near the tropopause). Focus B is also important determining the processes that control subtropical moisture. In particular, if fine-scale features are not ubiquitous then this will cast doubt on lateral mixing being the dominant source of moisture in the subtropics as well as the TTL paradigm.

Each of the scenarios listed above (Hypotheses 1-3) will leave a distinct isotopic signature in H\textsubscript{2}O found in the dry downwelling regions. Focus C, the measurement of the isotopic composition of H\textsubscript{2}O, will provide important new constraints on the origin of upper tropospheric water vapor, and will be vital for distinguishing between different mechanisms for controlling the moisture in each region. The isotopic composition of water vapor preserves a record of the condensation history of an air mass (see section 3.4), and profiles of H\textsubscript{2}O, HDO, and H\textsubscript{2}\textsuperscript{18}O can be used to quantify the role of the various
sources of water vapor for the tropical and mid-latitude upper troposphere (Webster et al., 2000).

The signature for hypothesis 1, Evaporation of Precipitation, will be a relatively constant vertical profile of vapor isotopic composition. The lofted ice particles that supply water to this region will preserve the much heavier isotopic composition of their original condensation level. If these ice particles are the primary source of water, the ambient levels of HDO and H$_2^{18}$O will reflect primarily the composition of the particles, with little fractionation upon evaporation. The signature for hypotheses 2 and 3 will be a sloped profile of vapor isotopic composition that becomes lighter with increasing altitude.

For hypothesis 3, Lateral Transport, the isotopic ratios should act as “conserved tracers” as air parcels from convective and downwelling regions mix. At a specific potential temperature, correlation plots of HDO vs H$_2$O and of H$_2^{18}$O vs H$_2$O should define “mixing lines” that can be used to quantify the relative contribution of each air mass type to the resulting admixture. In regions where profiles of HDO and H$_2^{18}$O vs altitude become progressively lighter with increasing altitude, the failure to observe such mixing lines will suggest that hypothesis 2, Evaporation of Detrained Cloud Particles, is occurring. Comparison of plots of HDO vs H$_2$O can yield information about mixing of air masses from different altitudes. Therefore, measurements of the isotopic composition of H$_2$O may provide quantitative information regarding the atmospheric level(s) from which water is advected according to the Lateral Transport scenario.

To address all the above issues aircraft measurements will be required in both (tropical) moist and (subtropical) dry regions, and key flights will be those that cross between these two types of regions (i.e., flights that cross strong gradients in water vapor). It will also be important (for validation of the lateral mixing hypothesis) to flight within regions where transport between moist and dry regions is predicted to occur. This can be done by the same series of flights proposed below.

6. Observational Strategies and Instrument Development

6.1 Observational Strategies for TC$^3$

The Tropical Composition and Climate Coupling Experiment will combine AURA and other satellite measurements with in situ and remote observations obtained from a high altitude aircraft (the ER-2 and/or WB-57) deployed from two Pacific sites: Guam (13.4°N, 144.4°E) and Hawaii (21.3°N, 157.9°W). NASA aircraft have operated out of both of these sites and transit between the sites can be made in single flights of either the ER-2 or WB-57. These sites will enable observations of the two distinct meteorological regions - areas of active convection and areas of downwelling on both sides of the ITCZ. The following information is based on climatological fields of wind, temperature, outgoing longwave radiation (OLR), upper troposphere H$_2$O from MLS, and lower stratosphere H$_2$O from HALOE shown in Figures 2.2 and 2.3. (See also http://code916.gsfc.nasa.gov/Personnel/people.Newman_Paul_A./easton/easton.html for more information on these climatologies).
During Northern Winter:

Guam is in a non-convective region (low clouds, high OLR), but to the south (and within reach by the ER-2 or WB-57) is a region of strong convection. There is a strong north-south gradient in UT water vapor over Guam (Figure 2.2), with high water vapor to the south over the region of strong convection. Within the neighborhood of Guam there are also important variations of temperature and H$_2$O near the tropopause. The cold pool at 100 hPa is centered just about over Guam, while (according to HALOE) the region of minimum water vapor at the tropopause is slightly north of Guam. There are also strong variations in the horizontal flow, in both the UT and LS (i.e. 200 to 70 hPa), near Guam, which are possibly important for both stratospheric dehydration and redistribution of UT moisture.

Therefore a NH winter deployment with principal base in Guam, with flights to the south and the north, would sample

- convective and non-convective regions
- high and low UT water vapor (and cross steep gradients in UT H$_2$O)
- the cold pool in the tropopause or TTL
- near the tropopause the region of minimum H$_2$O.

The conditions in the region around Hawaii are very different from those over Guam during northern winter, and flights from Hawaii would enable an even larger contrast than the Guam-based flights. Sampling lower UT water vapor and tropopause air away from the cold pool as well as the region where large lateral transport is expected (transport from extratropics into tropics both above and below the tropopause, and transport moist tropical UT into dry subtropics). The Hawaii component of the deployment may only need to consist of 1 flight into Hawaii from Dryden, 1 flight due south and back, and 1 flight to Guam, with reverse for flights back to Dryden.

During Northern Summer:

A corresponding NH summer deployment would provide valuable information on seasonal variations in chemistry, transport, and H$_2$O. As shown in Figure 2.3, the conditions in the summer are very different than winter Summer flights would enable investigation of the influence of the Asian monsoon on the composition of the tropical UT/LS (from Hawaii it is possible to reach the preferred region for exchange across the northern subtropical tropopause, e.g., Figure 4 of Postel and Hitchman 1999) and may provide key information for understanding the cause of the seasonal cycle and apparent long-term rise of stratospheric H$_2$O.

Notes:
- Need to determine whether it is possible to sample “outflow” of Asian monsoon from Guam/Hawaii, both in UT and LS.

- For possible moistening of tropical stratosphere, Sep-Oct may be preferred season.

- Need to talk a bit about the balloon deployment (s) from the tropics: will the site used previously in Brazil be good enough?

Finally: it is beyond the scope of this draft white paper to delve into which high altitude aircraft is preferable. Clearly there are advantages and disadvantages for each. Ideally, the mission will involve both high-altitude aircraft as well as a lower-altitude aircraft that will provide LIDAR measurements and in situ observations of the composition of air that is convected to the upper troposphere. Aircraft issues, as well as details of the payload(s), are better handled after a consensus has been reached on the scientific goals of a tropical mission.

6.2 New Instrumentation Needs for TC3

The previous sections have outlined a number of new instrument needs that will be required to test the various hypotheses. All of these measurements must be obtained at a vertical resolution of order 100 m (e.g., high temporal resolution sampling is required) to test the TTL paradigm and the associated “hypotheses of stratospheric aridity”, “hypothesis of LS and UT photochemistry”, and “hypotheses of UT water vapor”. It is conceivable that some of these measurement requirements could be met by a Whole Air Sampler system. If that is the case, this system must be capable of obtaining more samples than are returned by the current ER-2 system. The new measurement needs are:

- Water Isotopes (both HDO and H$_2$O$^{18}$)
- Condensed Phase H$_2$O
- Short Lived Tracers of Convection (Ra, $^{210}$Pb, $^{222}$Rn, CH$_3$I)
- Oxygenated Hydrocarbons (PAN, acetone), H$_2$O$_2$, and possibly HNO$_4$
- Short Lived Organic Bromine Source Species (CH$_3$Br, CHBr$_3$, CH$_2$Br$_2$, Halons, etc)
- Iodine Monoxide (IO) and its primary Organic Source (CH$_3$I). Measurements of CH$_2$I$_2$, C$_2$H$_5$I, CF$_3$I are also desired.
- Sulfur Dioxide (SO$_2$) (from an airborne platform) and its organic sources CS$_2$, dimethyl sulfide (DMS), methyl sulfonic acid (MSA).
7. Summary of Hypotheses for TC$^3$

The following hypotheses will be directly tested by the proposed tropical high-altitude aircraft flights from Guam and Hawaii, balloon flights from an as yet undetermined tropical site(s), and measurements from the AIRS, HIRDLS, MLS, and OMI instruments on board EOS Aura. An evaluation of these hypotheses will play a central role in gaining quantitative understanding of the couplings between atmospheric composition and climate:

There is a tropopause transition layer (TTL) in the tropics between 14 and 20 km characterized by:
- slow ascent (driven by extratropical wave driving in the stratosphere) and quasi-horizontal mixing with the extratropics;
- lateral mixing into/out of/within this layer is limited to certain regions and seasons: downstream of the summer monsoons, and within the northern winter westerly ducts over the Pacific and Atlantic oceans.

Stratospheric Aridity is maintained either by:

Class I Hypotheses (One dimensional models):
1) **Cold Trap** - slow, large-scale upwelling through the cold tropopause dehydrates ascending air
   1a) **Fountain** - air enters the stratosphere only in the Western Pacific.
2) **Convective Overshoot** - Convective turrets ascend rapidly from near the ground and penetrate into the stratosphere, where they induce rapid dehydration due to precipitation of ice.
   2a) **Ice Lofting** - air entering the stratosphere contains significant amounts of ice, some of which precipitates and some of which evaporates.
   2b) **Supersaturation** - air entering the stratosphere is highly supersaturated (e.g., 150%) with respect to cloud top temperature

Class II Hypotheses (Three dimensional models):
3) **TTL Cold Trap** - air enters the TTL to the west of the region of coldest temperatures in an area of forced upwelling and parcels are advected to the region of coldest TTL temperatures by an anti-cyclonic flow. As the air approaches the region of coldest temperature, it sinks, cools, and desiccates over relatively long time scales (several days).
4) **TTL Overshoot** - convective turrets penetrate the TTL. The more energetic turrets penetrate to higher levels and become rapidly (time scales of less than a day) dehydrated due to sedimentation of ice. The resulting humidity of the TTL is caused by mixtures of air parcels that detrain at different potential temperatures.
5) **Stratospheric Dehydration** - Dehydration in the lowermost stratosphere is induced by meso or synoptic-scale wave activity that lifts and cools air masses in the lower.

Sources of stratospheric bromine:
Short-lived bromine containing compounds enhance significantly the concentration of bromine monoxide in the upper troposphere and lower stratosphere. As a result, ozone loss rates in the lower stratosphere are significantly faster than expected.

Iodine and stratospheric ozone:
Iodine monoxide is an important catalyst for the destruction of stratospheric ozone.
PAN, oxygenated hydrocarbons, and O₃ production in the UT:
Oxygenated VOC photolysis both increases HOₓ levels and promotes the formation of PAN in the upper troposphere. The enhanced PAN formation significantly reduces the NOₓ/NOy ratio. This reduces the ozone formation rate in the upper troposphere and increases the input of NOₓ to the stratosphere.

The stratospheric sulfate aerosol layer:
Short-lived sulfur containing compounds contribute significantly to the background stratospheric aerosol.

The supply of moisture to the subtropical free troposphere is maintained either by:
1. Evaporation of precipitation: Evaporation of hydrometeors provides the main moisture source for the subtropics.
2. Evaporation of detrained cloud particles: Evaporation of dissipating clouds provides the main moisture source for the subtropics.
3. Lateral Transport: Outflow from tropical convective regions occurs over a deep layer (i.e. not restricted to altitudes above 200 hPa) and lateral transport by the large-scale wind field over this layer provides the additional supply of moisture.

Finally, the study of stratospheric aridity is motivated in part by the need to understand the cause of the apparent rise in stratospheric H₂O during the 1990s and to clearly define future changes to stratospheric H₂O that occur during the EOS Aura era. Our mission is focused on elucidating the mechanism(s) that govern stratospheric H₂O and on validating a suite of measurements to be obtained from EOS Aura. Testing the hypotheses of stratospheric aridity will provide insight into the climatic variables (e.g., surface temperature; tropopause temperature; convective energy, etc.) that affect stratospheric H₂O (e.g., Section 3.2). Our proposal to conduct this mission over several seasons, and several years, is designed to provide additional insight into how seasonal and/or interannual variability in processes in the hypothesized tropical TTL affect stratospheric H₂O. In this way, the TC³ mission will lay the ground-work for the use of Aura satellite observations to test the following hypotheses:

An increase in stratospheric H₂O may be caused either by:
Rising Cold Trap Temperature - Stratospheric H₂O responds directly to warming of the tropopause region.
Increasing Asian Monsoon - An increase in the extremity of the Asian monsoon over Indonesia leads to a moister upper troposphere, resulting in greater abundance of H₂O within the TTL for air parcels that do not pass directly through a cold trap.
Changing Ice Physics - Convective turrets become less efficiently dehydrated over time due to subtle changes in the microphysics of ice.
Reduced Brewer-Dobson Circulation - Stratospheric wave activity in the NH is reduced, resulting in increased fraction of air entering the TTL during NH.
8. References


Rosenlof, K.H. et al., 2000: Water vapor trends paper (need title), in press (or, submitted?).


9. List of Figures

The figures referred to in this white paper are saved as individual files on the server containing this document. Captions are included with each figure. Both postscript (*.ps) and “portable document format” (*.pdf) files are provided for each figure. Please contact one of the authors if you are having difficulty accessing or printing the figures. The figures are numbered according to the section of the document where they first appear.

The list of figures is:

2.1 Schematic drawing of the TTL.
2.2 January climatology of wind, outgoing longwave radiation, 100 mbar temperature, and H2O at 215 mbar and at 100 mbar.
2.3 July climatology of wind, outgoing longwave radiation, 100 mbar temperature, and H2O at 215 mbar and at 100 mbar.
3.1 Trends in upper stratospheric H2O from HALOE and from a ground based spectrometer showing a significant rise in the abundance of H2O between 1992 and 1996.
3.2 Trends in stratospheric H2O from HALOE, for 1992 to 1996 and for 1992 to 1999, showing a recent “slow down” of the H2O increase rate and also rather small trends for H2O in the lower stratosphere.
3.3 Time series of H2O+2CH4 in the lower stratosphere from various ER-2 missions, showing evidence for no change in total hydrogen between 1993 and 1997.
3.4 Time series of monthly, tropical mean temperature at 100 hPa, 1979 to 1998, and time series of mean NH winter-time temperature for three tropical stations that suggest the tropopause cooled during the mid-to-late 1990s.
3.5 Measured and modeled values of HDO (δD) in the tropics that indicate stratospheric water is isotopically heavier than expected, assuming thermodynamic equilibrium and no ice lofting across the tropopause.
4.1 Measured and modeled mixing ratios of BrO in the mid-latitude, lower stratosphere that suggest Brγ is several pptv greater than zero near the tropopause.
4.2 The contribution of bromine cycles to ozone loss for two Brγ scenarios, illustrating the importance of several pptv abundances of Brγ near the tropopause.
4.3 Solar spectrum measured at Kitt Peak showing evidence for little or no stratospheric IO.
4.4 Differential slant column density measurements of IO from Ny-Alesund showing evidence for considerable abundances of stratospheric IO.
4.5 Measured and modeled profiles of OH in the tropical upper troposphere and calculated production rates for HOx, illustrating the importance of non-methane hydrocarbon and/or peroxide sources of HOx.
4.6 Measured and modeled profile for SO2 under non-volcanic conditions, showing much larger abundances of stratospheric SO2 than expected.
5.1 Measured and modeled global distributions of H2O in the upper troposphere (330 K surface) for March 1993, illustrating the importance of large-scale advective mixing as a source of sub-tropical tropospheric H2O.