

## Variations in middle atmospheric water vapor from 2004 to 2013

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[1] We show measurements of middle atmospheric water vapor as measured by two ground-based Water Vapor Millimeter-wave Spectrometer instruments and three satellite-based instruments: the Aura Microwave Limb Sounder, the Atmospheric Chemistry Experiment (ACE), and the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS). We also show CH<sub>4</sub> measurements from the MIPAS and ACE instruments and use these to help interpret the H<sub>2</sub>O variations. We find that interannual changes in stratospheric H<sub>2</sub>O of ~0.5 ppmv, observed from Table Mountain, California, at 26 km and 40 km from 2010 to 2012, are caused primarily by dynamically driven changes in CH<sub>4</sub> during this period. The interannual variations in H<sub>2</sub>O observed over Mauna Loa, Hawaii, are shown to be quite similar to the average variations observed over 50°S–50°N in the lower mesosphere; thus, we conclude that a single ground-based microwave instrument can provide a useful estimate of interannual globally averaged lower mesospheric H<sub>2</sub>O variations, even when such changes are as small as ~0.2–0.3 ppmv. We find that the increase of ~0.2–0.3 ppmv in H<sub>2</sub>O in the lower mesosphere since 2006 is qualitatively consistent with an increase in tropical tropopause temperature since around 2001.

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### 1. Introduction

[2] There have been significant variations in middle atmospheric water vapor over the last two decades, including a large increase observed by the Halogen Occultation Experiment (HALOE) and by the Naval Research Laboratory's Water Vapor Millimeter-wave Spectrometer (WVMS) instruments in the early 1990s [Evans *et al.*, 1998; Nedoluha *et al.*, 1998a], a large decrease observed by several satellite instruments in 2001 [Randel *et al.*, 2004, 2006], and an overall increase observed by balloons since the 1980s [Hurst *et al.*, 2011]. These variations have spawned a series of studies of the effects of changes in stratospheric water vapor on the radiation budget [Forster and Shine, 1999; Maycock *et al.*, 2011; Tandon *et al.*, 2011]. Studies have also shown that regional changes in circulation can result from changes in stratospheric water vapor [Joshi *et al.*, 2006; Maycock *et al.*, 2013]. In addition, Solomon *et al.* [2010] showed that changes in stratospheric water vapor have had a significant effect on global surface climate change over the last decade.

[3] The oxidation of CH<sub>4</sub> in the stratosphere and the lower mesosphere results in the production of H<sub>2</sub>O. Year-to-year changes in water vapor from the middle stratosphere to the lower mesosphere at any particular location are usually caused primarily by dynamical factors which change the amount of CH<sub>4</sub> oxidation experienced by parcels at a particular altitude. Coincident measurements of CH<sub>4</sub> along with H<sub>2</sub>O are therefore extremely useful in helping to understand the causes of interannual H<sub>2</sub>O variations in the middle atmosphere. We note that a significant fraction of the increase in stratospheric H<sub>2</sub>O observed in the early 1990s [Nedoluha *et al.*, 1998a] was a result of dynamically driven changes in CH<sub>4</sub> oxidation [Nedoluha *et al.*, 1998b].

[4] Over time scales of several years, variations in water vapor in the stratosphere seem to be coupled to changes in tropical tropopause temperatures [e.g., Randel *et al.*, 2004], and on time scales of several decades, anthropogenically driven increases in CH<sub>4</sub> (which have never been larger than ~0.015 ppmv/yr) [e.g., Dlugokencky *et al.*, 2011] should result in observable increases in stratospheric water vapor. In this study we will determine whether the WVMS measurements are sensitive to the influence of changes in tropical tropopause temperature on a decadal scale. We will particularly emphasize here the measurements in the lower mesosphere, where the effect of dynamics on CH<sub>4</sub> oxidation (and the resultant effect on H<sub>2</sub>O mixing ratios) is reduced because the CH<sub>4</sub> has almost all been oxidized. We note that in the upper mesosphere, decadal-scale changes in H<sub>2</sub>O are driven primarily by the solar cycle [Nedoluha *et al.*, 2009]; hence, in the upper mesosphere, it becomes more difficult to detect decadal-scale changes which are not driven by the

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solar cycle. Nevertheless, long-term changes in mesospheric H<sub>2</sub>O will affect the occurrence of polar mesospheric clouds [DeLand *et al.*, 2006].

## 2. Middle Atmospheric Measurements of H<sub>2</sub>O and CH<sub>4</sub> Since 2004

[5] Three satellite instruments have provided global water vapor measurements throughout much of the stratosphere and the mesosphere for most of the period from around 2004 to 2013, and two of these instruments also provide CH<sub>4</sub> measurements. Measurements included here are from the Aura Microwave Limb Sounder (MLS), from the Atmospheric Chemistry Experiment (ACE) mission taken with the Fourier transform spectrometer on board the Scisat-1 satellite (ACE-FTS), and from the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) on board the Environmental Satellite (Envisat) research satellite. We shall show comparisons between these satellite measurements and ground-based microwave measurements. We shall also show that although sampling from only a single ground-based location, the mesospheric water vapor measurements over Mauna Loa show interannual variations very similar to those observed in the satellite record on a nearly global scale (50°S–50°N).

### 2.1. Measurement Data Sets

[6] The WVMS instruments have been measuring water vapor since 1992 from sites of the Network for the Detection of Atmospheric Composition Change (NDACC). These measurements make use of the pressure broadening of the 22 GHz water vapor emission line in order to obtain a vertical profile of water vapor. In this study we will make use of WVMS measurements from two NDACC stations.

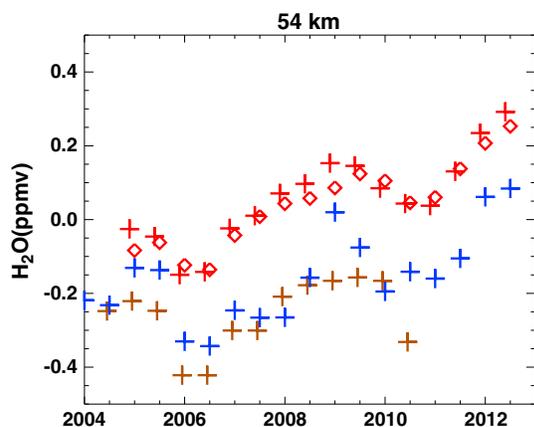
[7] The WVMS measurements to be shown in section 2.2 are those made from Mauna Loa (19.5°N, 204.4°E). Measurements have been made continuously from this site since 1996, but here we will show only measurements since 2004, the year in which Aura MLS began operations. In November 2010, a new generation of WVMS instruments, as described in Gomez *et al.* [2012], was deployed at Mauna Loa. In Nedoluha *et al.* [2013], we showed that this transition was completed without disturbing the long-term data record from this site. The new generation of WVMS instruments extends the useful altitude range of the WVMS measurements from the upper stratosphere down to 26 km, but the new Mauna Loa instrument has not yet remained in a configuration which is sufficiently stable for 26 km retrievals over an extended period (26 km retrievals are particularly sensitive to spectral baseline artifacts which do not necessarily affect higher altitudes); hence, the 26 km data from this instrument will not be shown in this study.

[8] Measurements with a new WVMS instrument at Table Mountain, California (34.4°N, 242.3°E), began in 2008 [Nedoluha *et al.*, 2011] and have been nearly continuous at this site since 2010 [Nedoluha *et al.*, 2013]. Measurements down to 26 km from June 2010 onward at Table Mountain are shown in section 2.4. The measurements from June 2010 through July 2012 are made using a constant (in amplitude, frequency, and phase) single sine-wave baseline. The addition of this constant baseline term can introduce a bias in the retrievals in the middle and lower stratosphere, but, as long as the baseline term remains nearly constant,

the retrievals remain sensitive to variations in water vapor at these levels. In August 2012, the new position of the reference absorber bar [Nedoluha *et al.*, 1995] necessitated the adjustment of the reference angles of the measurements in order to noise-balance the system under conditions with higher tropospheric optical depths. As a result of this change in reference angles, the instrumental baseline changed; hence, 26 km measurements at Table Mountain from August 2012 onward cannot be directly compared to the earlier 26 km measurements. At 40 km and above, single sine-wave baselines of the magnitude applied to any of the WVMS measurements have very little effect on retrievals (~0.02 ppmv) [Nedoluha *et al.*, 2013].

[9] The ACE-FTS instrument (hereafter referred to simply as ACE) is a high-resolution (0.02 cm<sup>-1</sup>) Fourier transform spectrometer which measures atmospheric absorption spectra between 2.2 and 13.3 μm (750–4400 cm<sup>-1</sup>). It performs solar occultation measurements from the middle troposphere up to 150 km [Bernath *et al.*, 2005; Boone *et al.*, 2005]. The altitude sampling of the ACE ranges from a measurement spacing of 6 km to a spacing of less than 2 km. However, for occultations with higher sampling rates, the altitude resolution is limited by the instrument's 1.25 mrad circular field of view, corresponding to 3–4 km. ACE is in a circular, low-Earth (650 km altitude) orbit with an inclination of 74°, resulting in near-global coverage approximately every 2 months. ACE measurements are available from February 2004 to the present, and the version 3.0 data are used for this study. The H<sub>2</sub>O data from ACE have been validated by Carleer *et al.* [2008] by comparisons to H<sub>2</sub>O measurements from several spaceborne instruments. The repeatability of the measurement relative to correlative measurements and its high precision (<5%) make it very suitable to scientific studies and a good reference measurement. The accuracy of the ACE H<sub>2</sub>O data is further supported by a study of Wrotny *et al.* [2009], which shows consistency between H<sub>2</sub>O and CH<sub>4</sub> variations within and between different pressure levels. ACE CH<sub>4</sub> measurements have been validated by De Mazière *et al.* [2008]. ACE data are shown here from 2004 through 2010. After September 2010, there was a problem with the a priori pressure and temperature data used in the analysis, and these data are therefore considered unreliable.

[10] The Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) is a limb emission spectrometer measuring in the middle infrared (4.15–14.6 μm, i.e., 2410–685 cm<sup>-1</sup>) from the Envisat platform in a Sun-synchronous orbit at about 780 km altitude [Fischer *et al.*, 2008]. For the MIPAS operation mode applied during the period relevant to comparison here, about 1300 profiles with 27 altitude steps covering 7 and 72 km altitude each along 14.4 orbits per day were obtained. Here we use the IMK/IAA V5R\_H2O\_220 water vapor product from the Institute for Meteorology and Climate Research, Karlsruhe Institute of Technology. This product has a vertical resolution of 2.5–4.5 km in the relevant altitude range. The precision of another data version with an identical retrieval approach but based on an earlier spectra version has been assessed at about 7% for a single profile, while the total accuracy, including systematic (mainly spectroscopic errors), was between 13% (at 40 km) and 19% (at 25 km) [von Clarmann *et al.*, 2009]. The MIPAS IMK H<sub>2</sub>O data have been validated within the MOHAVE-2009 campaign [Leblanc *et al.*, 2011] by comparison to a number of ground-based,



**Figure 1.** Annual water vapor mixing ratio anomalies relative to an MLS climatology. Symbols are shown for January–December and July–June; thus, each measurement is included in two annual anomalies. Measurements over Mauna Loa are indicated by crosses. Measurements are from MLS (red), WVMS (blue), and ACE (brown). Coincident satellite data have been convolved with WVMS averaging kernels. Also shown are MLS anomalies from 50°S–50°N at 0.46 hPa (~54 km) which have not been convolved with WVMS averaging kernels (red diamonds).

balloon-borne, and satellite instruments. The deviation of MIPAS was within plus and minus 10% of all other instruments, with a slight tendency toward a systematic high bias of <10% between 35 and 45 km [Stiller *et al.*, 2012]. Similar to ACE, the study by Wrotny *et al.* [2009] supported the high quality of MIPAS data by demonstrating the consistency between H<sub>2</sub>O and CH<sub>4</sub> variations within and between different pressure levels. Methane is retrieved from MIPAS spectra in the 1220–1305 cm<sup>-1</sup> wave number region, simultaneously with N<sub>2</sub>O. The vertical resolution ranges from 2 km in the lower stratosphere to 5 km in the mesosphere. The data version used here, i.e., V5R\_CH4\_222/223, has been validated against four different satellite data records, a balloon-borne in situ cryosampler instrument, and the MkIV FTIR instrument (A. Laeng *et al.*, Validation of MIPAS IMK/IAA methane version V5R CH4 222 profiles, submitted to *Remote Sensing of Environment*, 2013). The MIPAS measurements shown here are from 2005 until April 2012, when contact to Envisat was lost.

[11] The Aura MLS H<sub>2</sub>O water vapor product is retrieved from the radiances measured by the radiometers centered near 190 GHz [Lambert *et al.*, 2007; Read *et al.*, 2007]. The instrument began producing science observations on 13 August 2004. The version 3.3 (v3.3) stratospheric and mesospheric water vapor data used here are the latest update to the v2.2 version, which is validated and described in Lambert *et al.* [2007]. The MLS v3.3 data have significant improvements for a number of species [Livesey *et al.*, 2011], including the H<sub>2</sub>O product. Correlative measurement comparisons show a fine-scale oscillation in the v2.2 H<sub>2</sub>O retrievals, and this retrieval artifact has been eliminated in the v3.3 retrievals. The accuracy is estimated to be 0.2–0.5 ppmv (4–11%) for the pressure range 68–0.01 hPa. The scientifically useful range of the H<sub>2</sub>O data is from 316 to 0.002 hPa. Aura MLS measurements shown here are from August 2004 until February 2013.

[12] Extended comparisons between WVMS and MLS data have been presented in many previous studies, and the measurements have always shown good agreement in temporal variations on time scales of months to years [Nedoluha *et al.*, 2007, 2009, 2011]. The Aura MLS data are particularly valuable to this study since daily measurements throughout the WVMS altitude range are available coincident with both Mauna Loa and Table Mountain. We use Aura MLS measurements within ±2° latitude and ±30° longitude of each site from August 2004 to March 2011 and averaged over ±5 days for each target day-of-year to calculate daily climatological values which provide daily a priori profiles for the WVMS water vapor retrievals. With the exception of the lower stratosphere (where National Centers for Environmental Prediction data are used) and the thermosphere (where the MSIS model is used), a similarly calculated MLS temperature climatology provides the required background temperature for the retrieval.

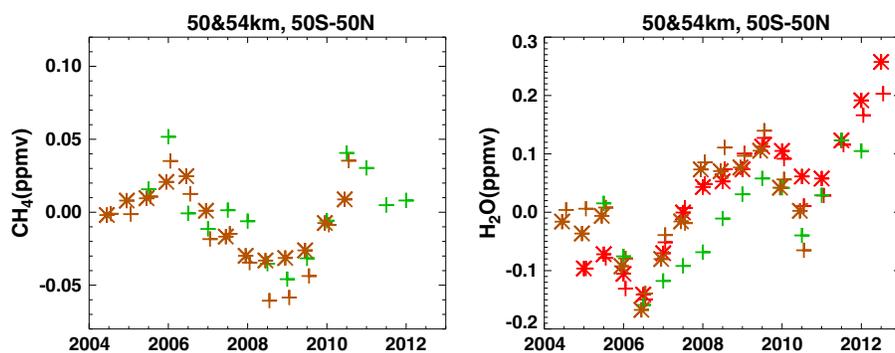
## 2.2. Observed Variations Near the Stratopause

[13] In Figure 1, we compare annual water vapor anomalies (deviations from a mean value of ~7 ppmv) from ACE, MLS, and WVMS at 54 km. The anomalies in Figure 1 are calculated by differencing all measurements from the daily climatology created from MLS data. Because of the relatively good absolute agreement of the ACE, MLS, and WVMS measurements at this altitude (all within ~3% of each other), it is possible to plot the absolute differences and still clearly see the interannual variations. Comparisons of absolute H<sub>2</sub>O measurements for the measurements presented here are given by Carleer *et al.* [2008], Stiller *et al.* [2012], and Nedoluha *et al.* [2007]. The causes of absolute differences between H<sub>2</sub>O measurements are often not understood, and such differences can be large when compared with the ability of any single instrument to accurately measure variations.

[14] The sensitivity of MIPAS retrievals decreases above ~50 km where the retrieval is increasingly dependent on non-local thermodynamic equilibrium effects [Stiller *et al.*, 2012]; hence, MIPAS measurements are not shown in Figure 1. At this altitude, the variation in water vapor caused by variations in Lyman  $\alpha$  radiation related to the solar cycle is statistically insignificant at the ~1% level (~0.07 ppmv) [Nedoluha *et al.*, 2009]. Below this altitude, dynamically driven variations in H<sub>2</sub>O from CH<sub>4</sub> oxidation rapidly increase, making it increasingly difficult to identify long-term changes in H<sub>2</sub>O without coincident CH<sub>4</sub> measurements.

[15] Three of the sets of measurements shown in Figure 1 are over Mauna Loa. The coincident satellite measurements (within ±2° latitude and ±30° longitude for MLS; within ±5° latitude for the more sparse ACE data) are convolved with the averaging kernels from the WVMS measurements. These show similar variations, with the eight full calendar year (2005–2012) anomalies from WVMS and MLS having a correlation coefficient of 0.94 (the standard deviation of the difference is 0.10 ppmv).

[16] In addition to the WVMS measurements at Mauna Loa, and to the MLS and ACE measurements coincident with Mauna Loa and convolved to the WVMS resolution, we show in Figure 1 MLS anomalies on a more global scale. This second set of MLS annual anomalies is calculated from measurements which are not convolved and which are given on a constant pressure level from the native MLS retrieval



**Figure 2.** Annual (left) methane and (right) water vapor mixing ratio anomalies. Symbols are shown for January–December and July–June; thus, each measurement is included in two annual symbols. Measurements are from MLS (red), ACE (brown), and MIPAS (green). Measurements at 54 km (0.46 hPa for MLS) are indicated by stars, whereas measurements at 50 km (0.68 hPa for MLS) are indicated by crosses.

grid (0.46 hPa; which represents an average altitude of  $\sim 54$  km). In order to derive these annual median MLS values, we first calculate daily median anomalies from the MLS climatology in  $2^\circ$  latitude bands  $50^\circ\text{S}$ – $50^\circ\text{N}$  through March 2011 (hence, the time period used for this global climatology is consistent with that used for the WVMS retrievals). We then calculate monthly median anomaly values for each latitude bin and then calculate a  $50^\circ\text{S}$ – $50^\circ\text{N}$  value based on the average of these latitude bins (area weighting of the latitude bins does not perceptibly affect the results). Finally, we average together 12 monthly values to obtain an annual average. As shown in Figure 1, the annual anomalies from the global, constant pressure-level MLS data are only slightly different from the convolved, altitude-based MLS anomalies coincident with Mauna Loa. The correlation coefficient between these two analyses of MLS data is 0.98, while between the WVMS and the  $50^\circ\text{S}$ – $50^\circ\text{N}$  pressure-level MLS data set, the correlation coefficient is 0.92. Based on the similarity of the two analyses of MLS data, we conclude that at least at this particular altitude, the ground-based measurements provide a good indication of average  $\text{H}_2\text{O}$  changes over a large fraction of the atmosphere.

[17] In Figure 2, we show annual methane and water vapor mixing ratio anomalies for MLS, ACE, and MIPAS for  $50^\circ\text{S}$ – $50^\circ\text{N}$ . The ACE and MIPAS values shown in Figure 2 are not calculated as anomalies from the MLS climatology as in Figure 1. Such a subtraction would cause the aliasing of latitude-dependent difference biases into time-dependent biases when sampling changes occur in the ACE and MIPAS measurements (the former of which are sparse, while the latter have some measurement gaps). A second possible choice would have been to calculate separate latitude-dependent climatologies for each of these data sets, but this was clearly problematic especially for the sparse ACE data set. In order to determine the MIPAS and ACE anomalies, we therefore first fit a function with a constant term plus annual and semiannual terms to all measurements within each  $10^\circ$  latitude band from  $50^\circ\text{S}$ – $50^\circ\text{N}$ . We then used the anomalies from this fit in the same manner as for the MLS global anomalies, first calculating monthly median anomaly values for each latitude bin and then averaging together the latitudes to get a monthly global anomaly, and, finally, averaging together the monthly global anomalies to get

an annual anomaly. Note that unlike Figure 1, which is presented as an offset from an MLS climatology near Mauna Loa, in Figure 2, each instrument is compared to its own annual fit; hence, Figure 2 contains no information regarding the mean offset between the instruments.

[18] Both ACE and MIPAS retrievals are calculated on an altitude grid. ACE anomalies are shown at 50 km and 54 km, while MIPAS anomalies are shown here only at 50 km. While the anomalies at 50 km and 54 km are similar, we note that there is slightly more variation at the lower altitude. The reason for this is apparent in Figure 2, which shows the  $\text{CH}_4$  anomalies from ACE and MIPAS (MLS does not measure  $\text{CH}_4$ ). The annual anomalies of  $\text{H}_2\text{O}$  at 50 km are more variable than at 54 km because the amount of  $\text{CH}_4$  which is oxidized to form  $\text{H}_2\text{O}$  is more variable at 50 km. This makes it more difficult, in the absence of  $\text{CH}_4$  measurements, to interpret long-term changes in  $\text{H}_2\text{O}$  that are not caused by the shorter-term dynamical effects which affect the amount of  $\text{CH}_4$  which is oxidized to form  $\text{H}_2\text{O}$ .

[19] In Table 1, we show the standard deviations of the differences of the annual averages of pairs of data sets. The largest such standard deviation shown is 0.068 ppmv ( $\sim 1\%$ ). For cases where three similar measurement data sets are available (e.g., MLS at 0.68 hPa, ACE at 50 km, and MIPAS 50 km), the standard deviation is calculated using one instrument compared to the average of the other two instruments. The last entry shows the standard deviation between the annual anomalies calculated from the difference relative to the daily MLS climatology in  $2^\circ$  latitude bins and the annual anomalies calculated using  $10^\circ$  latitude bins and fitting for an annual and a semiannual cycle. While this standard deviation is lower than the standard deviation for the other  $\text{H}_2\text{O}$  comparisons, it is not insignificant and points out that at the  $\sim 1/3\%$  level, different seemingly reasonable methods will give different answers for these variations.

[20] Figures 1 and 2 both show that  $\text{H}_2\text{O}$  near the stratopause was at a minimum, both globally and at Mauna Loa, in 2006. From 2006 through 2008, Figure 2 shows that there was a decrease in  $\text{CH}_4$  at 50 km and 54 km, and this certainly plays an important part in the rapid increase in  $\text{H}_2\text{O}$  during this period. The number of  $\text{H}_2\text{O}$  molecules resulting from the oxidation of a single  $\text{CH}_4$  molecule in the stratosphere is not precisely a factor of 2, because  $\text{H}_2$  oxidation also

**Table 1.** Annual Variation Standard Deviations (H<sub>2</sub>O; 50°S–50°N Unless Otherwise Indicated)

Comparison Datasets (levels)	Years	Standard Deviation
WVMS Mauna Loa (54 km) versus convolved MLS Mauna Loa (54 km)	2005–2012	0.068 ppmv
WVMS Mauna Loa (54 km) versus MLS (0.46 hPa)	2005–2012	0.059 ppmv
MLS (0.46 hPa) versus ACE (54 km)	2006–2010	0.028 ppmv
MLS (0.68 hPa) versus (ACE + MIPAS (50 km))	2006–2010	0.027 ppmv
ACE (50 km) versus (MLS (0.68 hPa) + MIPAS (50 km))	2006–2010	0.048 ppmv
MIPAS (50 km) versus (ACE(50 km) + MLS(0.68 hPa))	2006–2010	0.042 ppmv
MIPAS CH <sub>4</sub> (50 km) versus ACE CH <sub>4</sub> (50 km)	2006–2010	0.014 ppmv
MLS (0.68 hPa) versus MLS sine-wave fit (0.68 hPa)	2005–2012	0.022 ppmv

produces H<sub>2</sub>O, and the relative proportion of H<sub>2</sub>O production by H<sub>2</sub> oxidation compared to the H<sub>2</sub> production from CH<sub>4</sub> oxidation varies with altitude [Le Texier et al., 1988]. Wrotny et al. [2010] found, using vertical profiles of ACE, HALOE, and MIPAS data, that in the upper stratosphere, each oxidized CH<sub>4</sub> molecule results in an increase of ~2–3 H<sub>2</sub>O molecules. The decrease in CH<sub>4</sub> of ~0.08 ppmv, as shown in Figure 2, therefore produces an additional ~0.16–0.24 ppmv of H<sub>2</sub>O from 2006 to 2008. From 2008 to 2011, however, CH<sub>4</sub> increased again, so that the January–December 2011 50 km MIPAS CH<sub>4</sub> data are within ~0.01 ppmv of the January–December 2006 measurement. Changes in CH<sub>4</sub> should therefore have only a limited effect (<0.03 ppmv) on the difference between H<sub>2</sub>O in 2006 and 2011.

[21] In Table 2, we show the difference between annual H<sub>2</sub>O anomalies in 2006 and 2011. This is the last calendar year for which a full year of MIPAS data is available. Based on the standard deviations shown in Table 1, the WVMS, MLS, and MIPAS measurements all show increases in H<sub>2</sub>O which are statistically significant and which agree with each other. The difference in the MIPAS CH<sub>4</sub> measurement at 50 km in 2011 relative to that in 2006 is insignificant based on the MIPAS versus ACE comparison statistics in Table 1.

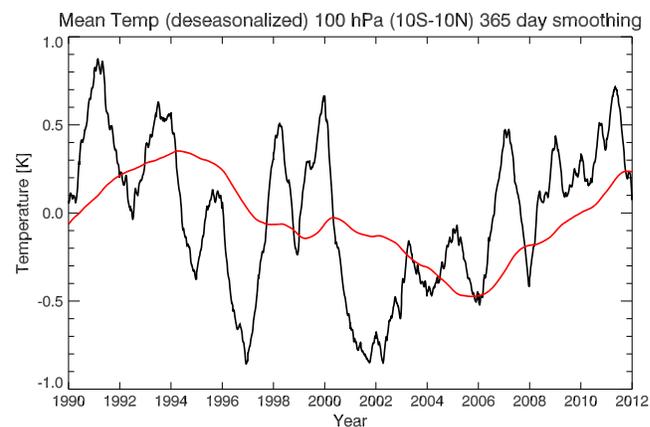
[22] In addition to the dynamically induced variations in H<sub>2</sub>O from CH<sub>4</sub> oxidation, the long-term, but very slow, increase in CH<sub>4</sub> entering the stratosphere should show up primarily as an increase in H<sub>2</sub>O once this air has reached the stratopause. Over the last decade, CH<sub>4</sub> measurements from Hawaii show an increase of ~0.03 ppmv (<http://www.esrl.noaa.gov/gmd/ccgg/flask.html>). An increase in CH<sub>4</sub> of this magnitude could result in an increase in H<sub>2</sub>O from oxidation of ~0.06–0.09 ppmv near the stratopause where almost all of the CH<sub>4</sub> has been oxidized. However, almost all of the increase at the surface in this century has occurred since 2006, and it takes several years for air to reach the stratopause [cf. Waugh and Hall, 2002], so the increase in H<sub>2</sub>O at the stratopause between 2006 and 2011 produced by this source is certainly <0.09 ppmv.

**Table 2.** The 2011 Annual Average Minus the 2006 Annual Average (H<sub>2</sub>O; 50°S–50°N Unless Otherwise Indicated)

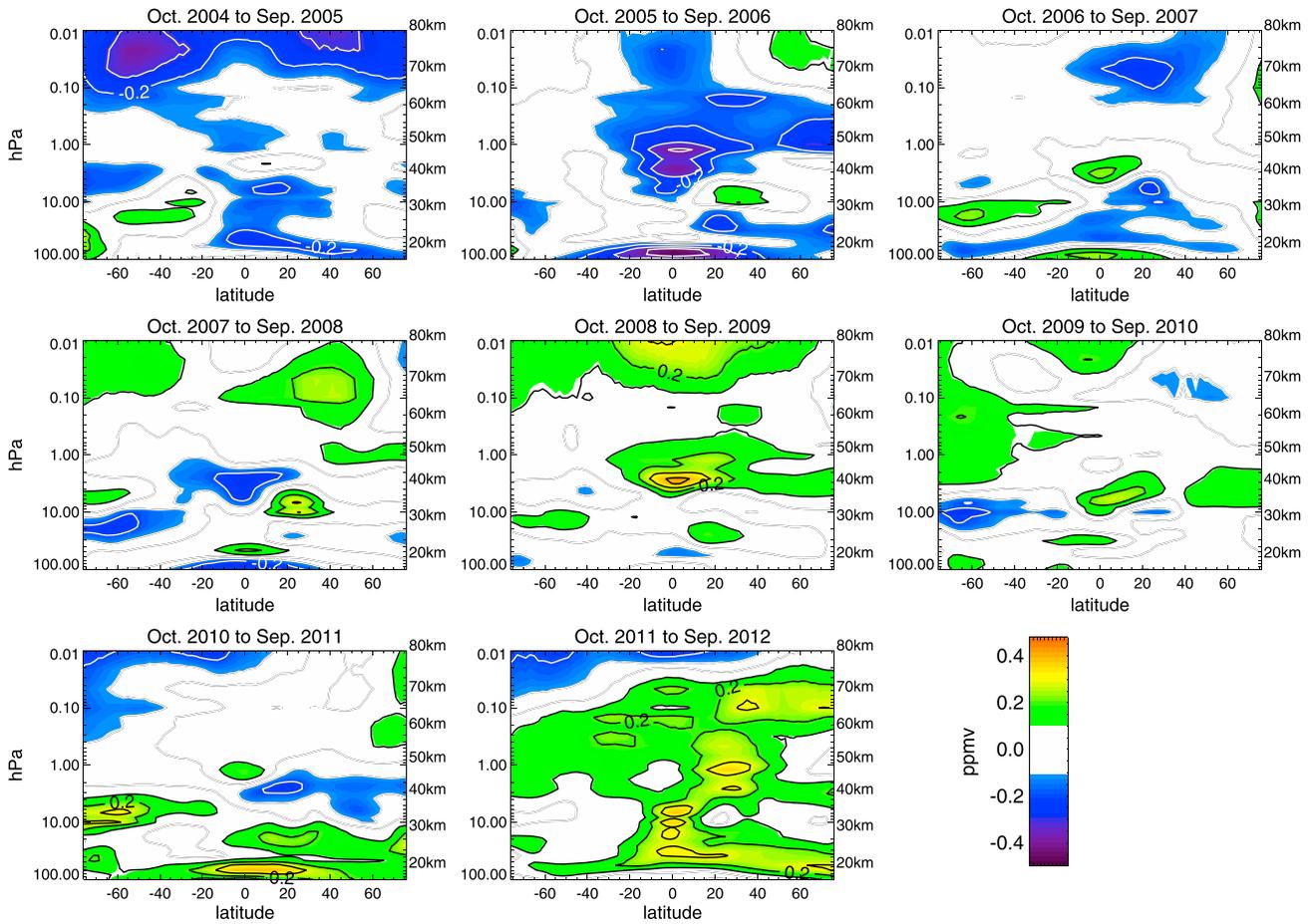
Dataset (level)	2011–2006 Difference
WVMS Mauna Loa (54 km)	0.238 ppmv
MLS (0.46 hPa)	0.264 ppmv
MLS (0.68 hPa)	0.253 ppmv
MIPAS (50 km)	0.282 ppmv
MIPAS CH <sub>4</sub> (50 km)	0.0056 ppmv

[23] Another source for increasing H<sub>2</sub>O at the stratopause is increasing H<sub>2</sub>O entering the stratosphere. In Figure 3, we show the mean 100 hPa temperatures for 10°S–10°N from the Modern-Era Retrospective Analysis for Research and Applications analysis. The mean 100 hPa temperature has certainly increased during the Aura MLS observation period. As has been shown in the frost-point hygrometer balloon measurements from Boulder, CO, water vapor at midlatitudes in the lower stratosphere increased over the period 2005–2010 [Hurst et al., 2011], with a measured increase of 0.35 ppmv at 16–18 km and 0.21 ppmv at 18–20 km. Fujiwara et al. [2010] examined HALOE, MLS, and balloon data and concluded that “observed decadal changes in lower stratospheric water vapor are generally in good agreement with 100 hPa temperature data in the tropical western Pacific from the ERA40, ERA-Interim, and JRA25/JCDAS reanalyses.”

[24] Surface-level CH<sub>4</sub> increases were small from around 1999 to 2005 and began to significantly increase again only in around 2006. However, as shown in Figure 3, tropical tropopause temperatures have been increasing for over a decade since the sharp drop from 2000 to 2001 [Randel et al., 2006]. The precise “age-of-air” in the lower mesosphere is dependent both on the dynamical model and on the type of signal being transported from the tropopause [Waugh and Hall, 2002], but typical values at ~50 km are ~3–5 years. The H<sub>2</sub>O values at 50 km therefore should reflect the effect of variations in tropical tropopause temperatures over a range of previous years. If we average together 5 years of tropical tropopause temperatures, as shown in



**Figure 3.** Mean 100 hPa temperature anomaly (10°S–10°N) shown as a running annual mean (black). Also shown is a running mean averaged over the previous 5 years (red).



**Figure 4.** Annual water vapor anomalies from the MLS measurements. Each contour represents an interval of 0.1 ppmv. Absolute anomalies below 0.1 ppmv are shown in white.

Figure 3, we find an increase of  $\sim 0.6$  K from the 5 years ending in 2005 as compared to the 5 years ending in 2012. The sensitivity of the water vapor saturation mixing ratio to changes in temperature is highly nonlinear, but at  $\sim 100$  hPa and  $\sim 190$  K, it is  $\sim 0.6$  ppmv/K, suggesting an increase in water vapor of 0.36 ppmv over this period. *Randel et al.* [2004], however, showed that while there was a reasonable correlation between tropical radiosonde cold-point tropopause temperatures and variations in  $H_2O$  measured by HALOE, the observed anomalies in HALOE  $H_2O$  were substantially smaller than the variations in saturation mixing ratio. *Fueglistaler et al.* [2013] calculated changes in water vapor entering the stratosphere using ECMWF back trajectories and several temperature products and, using a low-pass filter, found an increase of  $\sim 0.3$  ppmv from 2003 to 2010. The *Fueglistaler et al.* [2013] study corroborates our conclusion that most, but perhaps not all, of the increase in  $H_2O$  near the stratopause is due to an increase in 100 hPa tropical tropopause temperature since 2001.

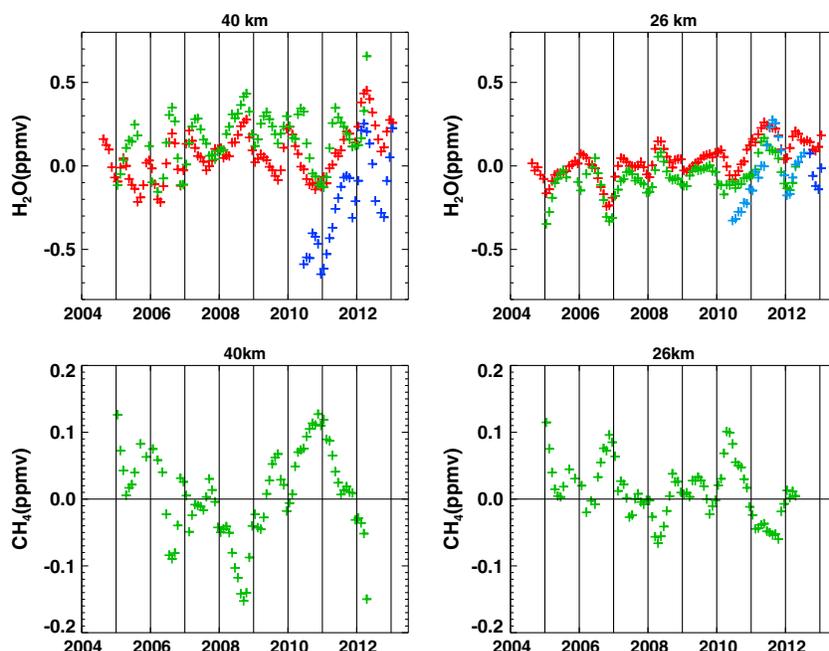
### 2.3. Global Variations Throughout the Stratosphere and Mesosphere

[25] In Figure 4, we show the zonally averaged  $H_2O$  anomalies from MLS measurements for each full year of measurements, from October 2004 to September 2012. From Figure 4, it is clear that the gradual increase of  $H_2O$

mixing ratios during the lifetime of the MLS mission shown in Figures 1 and 2 is indicative of an increase over almost the entire middle atmosphere. The upper mesosphere is the only exception to this, since here the peak values clearly occur in 2008/2009.

[26] In the upper mesosphere, photodissociation by Lyman  $\alpha$  radiation is the primary sink for  $H_2O$ ; hence, decadal-scale  $H_2O$  variations are driven primarily by the solar cycle [*Remsburg, 2010*]. The maximum water vapor mixing ratios in the upper mesosphere are expected when the Lyman  $\alpha$  radiation is near a minimum [*Nicolet, 1981*]. *Nedoluha et al.* [2009] showed, based on WVMS and HALOE measurements coincident with Mauna Loa, that above 60 km, the solar cycle causes significant variations in water vapor, while below 60 km, the variation in water vapor from the solar cycle was  $<2\%$  and became statistically insignificant in both data sets. The minimum in Lyman  $\alpha$  last occurred in 2009 and is consistent with the high water vapor observed by Aura MLS during this period in the upper mesosphere.

[27] Figure 4 shows that the largest range of interannual anomalies generally occur in the tropical middle to upper stratosphere. This is a region which experiences large seasonal and quasi-biennial oscillation-related variations in  $CH_4$  oxidation [cf. *Wrotny et al., 2010*]. Large variations in the annual anomalies do, however, also occur at other latitudes and at a range of altitudes.



**Figure 5.** (top) Monthly water vapor anomalies averaged over three consecutive months over Table Mountain. Results are shown for at (left) 40 km and (right) 26 km. Measurements are from MLS (red), MIPAS (green), and WVMS (blue). The satellite measurements have been convolved with the WVMS averaging kernels. The WVMS measurements at 26 km from July 2012 and before have been shifted by +0.3 ppmv and cannot be directly compared with those from August 2012 onward (they are shown in slightly lighter blue; see text for details). (bottom) Monthly methane anomalies from MIPAS measurements averaged over three consecutive months over Table Mountain. MIPAS anomalies have been convolved with WVMS averaging kernels. Anomalies are shown at (left) 40 km and (right) 26 km.

#### 2.4. Intercomparison of Anomalies in the Stratosphere Over Table Mountain

[28] In section 2.2, we intercompared ground-based water vapor measurements in the lower mesosphere with several satellite data sets since 2004 and provided some interpretation for the observed variations. Ground-based measurements down to 26 km have been only possible since 2010 at Table Mountain, so the intercomparison period with satellite instruments is necessarily shorter than that for the lower mesospheric comparisons. While the ground-based measurements cover just over 2 years, they do show significant changes in water vapor. We compare these recent changes with variations observed over Table Mountain by MLS and MIPAS since 2004.

[29] In Figure 5, we show monthly anomalies, smoothed over 3 months, for ground-based WVMS  $\text{H}_2\text{O}$  measurements from Table Mountain. As discussed in section 2.1, WVMS measurements at 26 km through July 2012 cannot be directly compared to WVMS measurements from August 2012 onward due to a change in instrumental baseline. To aid in displaying the variation within each period, the 26 km retrievals from June 2010 to July 2012 have been shifted by +0.3 ppmv. The 40 km retrievals are unaffected by this baseline change and have not been shifted.

[30] In addition to the WVMS measurements, we show monthly measurements from MLS and MIPAS. Both the MLS and MIPAS data sets generally have retrievals on at least 20 d/month, with the exception of 2005 and 2006. However, for the 21 months from January 2005 until September 2006, all but

4 months had less than 10 days with good MIPAS coincident retrievals; hence, these first few MIPAS monthly anomalies are slightly more uncertain because of this poor sampling.

[31] At 26 km, the MLS, MIPAS, and WVMS measurements show a clear increase from around mid-2010 to around mid-2011. For both MIPAS and MLS, which have been both observed since 2004, the  $\text{H}_2\text{O}$  mixing ratios at this altitude in 2011 are among the highest observed. All three instruments show a decrease in the  $\text{H}_2\text{O}$  anomaly at 26 km from 2011 to 2012. At 40 km, there is a large increase from the start of 2011 until mid-2012 which can be seen in the MLS, MIPAS, and WVMS data. This increase occurs several months after the increase at 26 km.

[32] In Figure 5, we also show the variation in  $\text{CH}_4$  anomalies from MIPAS measurements at 40 km and 26 km. The last MIPAS data points also have a somewhat smaller sample size (they are not smoothed over 3 months) and, at 40 km, serve to emphasize the anticorrelation of the  $\text{H}_2\text{O}$  and  $\text{CH}_4$  variations, but this anticorrelation is clearly present at both altitudes throughout the time series. In particular, the increase in  $\text{H}_2\text{O}$  from 2010 to 2011 at 26 km is clearly related to the decrease in  $\text{CH}_4$  over this period. This suggests that the air in 2011 has a different dynamical history, having experienced more  $\text{CH}_4$  oxidation and subsequent production of  $\text{H}_2\text{O}$  than that in 2010. While we cannot rule out that there may be some component of this 2010–2011 increase at 26 km which may be related to variations in  $\text{H}_2\text{O}$  entering the stratosphere, the magnitude of the  $\text{CH}_4$  variation is itself comparable to that required to explain the entire  $\text{H}_2\text{O}$

variation over this period. However, it is interesting that while the highest 26 km H<sub>2</sub>O values occur in 2011, the largest negative CH<sub>4</sub> anomaly is in 2008. This suggests that H<sub>2</sub>O entering the stratosphere increased between these years, consistent with the 100 hPa tropical MLS measurements shown in Fueglistaler *et al.* [2013, Figure 8a].

[33] Just as for the H<sub>2</sub>O, there appears to be a delay of several months between some of the large-scale variations at 26 km and those at 40 km. There are minima at 26 km in early 2006, early 2008, and late 2011, each of which is accompanied by a minima at 40 km (although no turnaround is yet apparent in the 2013 40 km data). Similarly, the maximum in mid-2010 is followed several months later by a maximum at 40 km. While in the tropics interannual CH<sub>4</sub> variations are much larger at 40 km than at 26 km, it is interesting to note that CH<sub>4</sub> variations at this midlatitude site (and, by implication, the associated variations in H<sub>2</sub>O caused by CH<sub>4</sub> oxidation) are only slightly smaller at 26 km than at 40 km.

[34] While there appears to be some correlation between the 40 km and 26 km CH<sub>4</sub> variations, we have been unable to find a dynamical connection. The temperatures measured by MLS over Table Mountain near 40 km are unusually low at the end of 2012, which may be related to an unusual amount of descent bringing down air with reduced CH<sub>4</sub> mixing ratios. It is possible that variations in descent are also significantly affecting the 26 km CH<sub>4</sub> anomalies, but the vertical gradient in CH<sub>4</sub> mixing ratio at 26 km over Table Mountain is typically ~1/2 that at 40 km, so it is likely that CH<sub>4</sub> mixing ratios in 2012 are also reduced in part due to mixing of low CH<sub>4</sub> air from higher latitudes.

### 3. Conclusions

[35] On interannual time scales, variations in H<sub>2</sub>O from the middle stratosphere to the lower mesosphere are driven primarily by changes in CH<sub>4</sub> oxidation, changes which are primarily driven by variations in stratospheric dynamics. These changes in CH<sub>4</sub> can be presently identified with the use of measurements from satellite-based instruments; however, the ACE-FTS instrument, which has been operating since 2004, is currently the only remaining instrument capable of making such measurements. WVMS measurements in the stratosphere are now possible down to 26 km in the stratosphere, a region in which interannual variations in CH<sub>4</sub> caused by stratospheric dynamics are probably the dominant driver of interannual variations in H<sub>2</sub>O at midlatitudes. The lower end of this altitude range overlaps the altitudes at which balloon measurements provide long-term measurement data sets; however, such interannual variations will only minimally affect multidecadal balloon estimates of trends.

[36] There has been an increase in H<sub>2</sub>O since 2004 near the stratopause and in the lower mesosphere. This increase of ~0.2–0.3 ppmv in H<sub>2</sub>O cannot be explained by variations in CH<sub>4</sub> oxidation but is qualitatively consistent with an increase in tropical tropopause temperature since around 2001. This increase is smaller in magnitude than either the increase of ~0.6–0.7 ppmv detected in the upper stratosphere/lower mesosphere from 1991 to 1996 [Nedoluha *et al.*, 1998a] or the decrease of ~0.4 ppmv in the lower stratosphere detected from 2000 to 2001 [Randel *et al.*, 2006]. The good agreement

between several instruments in their detection of the increase since 2004 is very encouraging. The similarity between the 2004–2011 variations observed by the WVMS instrument at Mauna Loa and the average variations observed over this period from 50°S to 50°N in the lower mesosphere shows that a single ground-based microwave instrument can provide a useful estimate of global changes in this region, even when such changes are as small as ~0.2–0.3 ppmv.

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