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Observations of deep convective influence on stratospheric water vapor and its isotopic composition

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[1] In situ observations of H_2O and HDO in the midlatitude stratosphere are used to evaluate the role of convection in determining the stratospheric water budget. The observations show that water vapor in the overworld stratosphere (potential temperature > 380 K) is isotopically heavier than expected. Measurements in an airmass with anomalously high concentrations of water vapor show isotopic water signatures that are characteristic of evaporated ice lofted from the troposphere during convective storms. Observed H_2O and HDO concentrations in the plume of enhanced water and in the background stratosphere suggest that extratropical convection can account for a significant fraction of the observed water vapor in the summertime overworld stratosphere above the mid-North American continent. **Citation:** Hanisco, T. F., et al. (2007), Observations of deep convective influence on stratospheric water vapor and its isotopic composition, *Geophys. Res. Lett.*, *34*, L04814, doi:10.1029/2006GL027899.

1. Introduction

[2] Stratospheric water vapor (hereafter H_2O) plays a critical role in the Earth's climate system. Reliable long-term climate forecasts therefore require accurate descriptions of the response of stratospheric H_2O to changes in climate forcing [*Shindell*, 2001; *Forster and Shine*, 2002]. While the exact nature of this response depends on the mechanism that controls the transport of water into the middle/upper troposphere and lower stratosphere, the mechanisms that control this response remain poorly understood [*Rosenlof*, 2003].

[3] In this paper we investigate the role of summertime continental convection in regulating the chemical composition of the extratropical overworld (the region above the 380 K potential temperature (θ) surface [e.g., *Holton et al.*, 1995]). There is abundant evidence that extratropical convection does penetrate well into the lowermost stratosphere [e.g., *Poulida et al.*, 1996; *Fischer et al.*, 2003], and even into the overworld [*Fromm and Servranckx*, 2003; *Jost et al.*, 2004; *Livesey et al.*, 2004; *Ray et al.*, 2004]. *Dessler and Sherwood* [2004] used measurements of H_2O abundance to show that extratropical convection plays an important role in determining the global-scale extratropical H_2O distribution at θ = 380 K. In this paper we revisit this issue using measurements of deuterated water, HDO. HDO is preferentially removed compared to H_2O as condensation occurs, resulting in dramatic reductions in the ratio of HDO/H_2O as air ascends from the boundary layer to the tropopause. Observations of this ratio provide insight and quantitative constraints on the mechanisms that regulate H_2O that cannot be obtained by observations of H_2O alone [*Moyer et al.*, 1996; *Keith*, 2000]. In this paper we present new measurements of HDO to show that the chemical signature of extratropical convection can be observed well into the overworld.

2. Observations

[4] This paper uses data from two instruments developed simultaneously at Harvard. Integrated cavity output spectroscopy (ICOS) is a cavity absorption technique that uses a tunable quantum cascade laser to probe individual rotational lines of H_2O, HDO, $H_2^{17}O$, $H_2^{18}O$, and CH_4 in the mid-infrared (λ = 6.74 μm) [*Sayres*, 2006]. The technique is similar to diode laser absorption spectroscopy that uses a Herriot cell, but with a far greater optical path length because of the use of a high finesse cavity instead of a multipass cell. The Hoxotope instrument combines a new water photolysis system with our pre-existing instrument for laser induced fluorescence detection of OH. Water is photolyzed with an excimer lamp at 172 nm, producing ground state OH and OD radicals. The radicals are detected with rotational state selective laser excitation at 287 nm and fluorescence at 309 nm (J. M. St. Clair et al., A new photolysis laser induced fluorescence technique for the detection of HDO and H_2O in the lower stratosphere, manuscript in preparation, 2007). Each instrument uses a separate rear-facing inlet to sample vapor phase water only.

[5] These two new water isotope instruments flew on the NASA WB-57 as part of the Aura Validation Experiment Water Isotope Intercomparison Flight (AVE_WIIF) campaign during June/July 2005. Three five-hour flights sampled the upper troposphere and lower stratosphere with level legs between 10 and 19 km in the region near Ellington Field in Houston, TX. In this paper, we will use data from the Hoxotope instrument (H_2O and HDO), the

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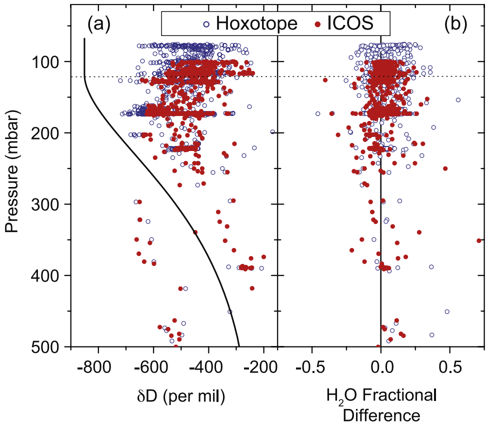

Figure 1. (a) HDO depletion measured by Hoxotope (open circles) and ICOS (closed circles) and predicted by Rayleigh distillation (solid line) expressed in units of per mil. (b) The fractional difference between the H_2O mixing ratio measured by Hoxotope (open circles) and ICOS (solid circles) and the Ly-α instrument, $[H_2O]_{\mathrm{Hox\ or\ ICOS}}/[H_2O]_{\mathrm{Ly}-\alpha} - 1$. The profiles were obtained on 3, 5, and 7 July, 2005 on the NASA WB-57 during flights based at Ellington Field, Texas. The observations are averaged to 30 s. An approximate tropopause is shown.

ICOS instrument (H_2O, HDO, and CH_4); the Harvard Lyman-α hygrometer (H_2O) [*Weinstock et al.*, 1994], and the Meteorological Measurement System (MMS) (pressure, temperature, and wind speed) [*Scott et al.*, 1990].

[6] The Hoxotope and ICOS observations are shown in Figure 1 along with a comparison of the H_2O measured by these new instruments and the well-established Lyman-α instrument. The ratio of HDO/H_2O is expressed in terms of δ notation: $\delta D(‰) = 1000 \times ([HDO]/[H_2O]/3.115 \times 10^{-4} - 1)$, where 3.115×10^{-4} is the ratio of HDO/H_2O in Vienna standard mean ocean water (VSMOW). The agreement between the ICOS and Hoxotope δD measurements is excellent: the average absolute difference between $\delta D_{\mathrm{Hoxotope}}$ and δD_{ICOS} is 15‰, well within the calibration uncertainties of ±50‰. Likewise, each isotope instrument shows excellent agreement with the Ly-α instrument, with a few outliers due to asynchronous sampling when H_2O changes rapidly. The average fractional difference between $[H_2O]_{\mathrm{Hoxotope}}$ and $[H_2O]_{\mathrm{Ly}-\alpha}$ is 0.02 ± 0.11; between $[H_2O]_{\mathrm{ICOS}}$ and $[H_2O]_{\mathrm{Ly}-\alpha}$ the difference is 0.01 ± 0.13. Though not shown, the agreement between these observations and those of the JPL hygrometer and the Harvard total water instrument, both of which also flew on AVE_WIIF, is similar.

[7] In agreement with previous in situ observations [*Webster and Heymsfield*, 2003], we see that the extratropical stratosphere is significantly underdepleted, meaning that the air is isotopically heavier than predicted by Rayleigh distillation [*Jouzel*, 1986]. Our measurements indicate stratospheric δD is −400 to −600‰. Webster and Heymsfield measured values between 0 and −900‰ during July 2002 over Florida in aircraft flights that intentionally sampled the environment around deep convection. Remote observations in the lower to mid-stratosphere indicate that the annual average overworld entry value (δD_e) is around −650‰ [*Moyer et al.*, 1996; *Johnson et al.*, 2001; *Kuang et al.*, 2003; *McCarthy et al.*, 2004]. Our measurements show less depletion than those δD_e values throughout the lowermost stratosphere. If both measurements are accurate, the subtropical and midlatitudes airmasses sampled in AVE_WIIF must have experienced significant perturbations to δD since crossing the tropical tropopause.

[8] It is also possible that one or both of the datasets is subject to measurement bias. There is no clear indication, however, that bias exists. The observations of the δD measured by the two in situ instruments agree with each other and the H_2O measured by these two instruments agree with the more established Harvard Ly-α and JPL hygrometers. The in situ measurements cannot be directly compared with the remote measurements, which were obtained in different years, seasons, and locations. Remote observations are also somewhat problematic in the lowermost stratosphere where atmospheric variability is high and instrument precision is low [e.g., *Johnson et al.*, 2001]. However, the value for the δD_e is determined from regressions of δD and CH_4 obtained in the stratosphere where the remote observations are most robust [*Moyer et al.*, 1996; *Johnson et al.*, 2001; *McCarthy et al.*, 2004].

[9] In the rest of the paper we focus on the overworld in order to investigate the effect of convection on the δD values observed by the in situ observations. Figure 2 shows a profile of δD in the overworld, with data binned in θ ranging from $\delta D = -450$ to -500‰. Also shown are measurements of H_2O obtained with (July flights) and without (June flights) simultaneous water isotope observations. Most of the water measurements show abundances of 5–7 ppmv between $\theta = 380$ and 430 K, values typical of the overworld. However, four flights sampled water vapor significantly above this background value with large enhancements observed at $\theta = 380$ and 400 K ($H_2O > 13$ ppmv), and smaller enhancements at $\theta = 420 - 430$ K ($H_2O > 8$ ppmv). Values of H_2O this high are not consistent with large-scale ascent through the tropical tropopause. The absence of perturbations to O_3 and CO_2 indicate the enhancements result from the injection of an air mass with a very high total water content, e.g. ice.

[10] The isotopic water data give us additional insight into these surprising observations. At the same time we see the enhancement in H_2O on the 7 July, 2005 flight, the two water isotope instruments observed large increases in δD compared to the background overworld. The heavy isotopic water is consistent with enhancement from the lofting and evaporation of ice particles from convective outflow of powerful continental thunderstorms [e.g., *Wang*, 2003]. The isotopic ratio of the convective outflow is determined assuming that evaporated ice mixed with the background stratospheric water. The isotopic ratio is calculated from the difference between the H_2O and HDO in the plume and the background stratosphere, summarized in Table 1. The mean calculated isotopic composition for the convective outflow is $\delta D_c = -210 \pm 60$‰. The isotopic values are similar to those used in modeling studies of ice lofting in the upper troposphere [*Gettelman and Webster*, 2005].

[11] The four encounters with enhanced H_2O provide evidence that lofting events that penetrate into the overworld stratosphere occur during the convective season over North America. The water isotopic composition can help

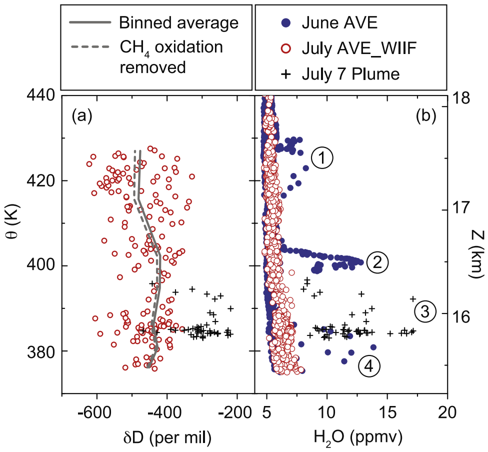

Figure 2. Profiles of (a) the depletion of HDO measured in the overworld during AVE_WIIF and (b) Ly-α water vapor. The δD values are an average of the Hoxotope and ICOS observations averaged to 60 s. The data are restricted to observations of $[H_2O]_{\text{Hoxotope or ICOS}}$ within ±5% of the Ly-α observations. The average δD profile in Figure 2a shows a binned average (every 4K in θ) for δD and for δD corrected for the oxidation of CH_4 [*McCarthy et al.*, 2004]. The H_2O profile in Figure 2b includes measurements obtained during the AVE campaign (solid circles) in June, 2005 in Houston, TX just prior to the AVE_WIIF campaign in July. The four plumes are labelled: (1) 17 June (37°N, 99°W), (2) 22 June (37°N, 99°W), (3) 7 July (34°N, 95°W) and (4) 21 June (26°N, 93°W). The δD was only obtained in the plume sampled on 7 July, highlighted with crosses. The altitude is approximate (±0.5 km).

quantify the integrated effect of these events. If the isotopic depletion of the air entering the stratosphere through the tropical tropopause is $\delta D_e = -650‰$, a large amount of lofted moisture is necessary to enrich the stratospheric water to the $\delta D = -450‰$ observed in these flights. In the limit that the H_2O abundance in the overworld stratosphere is a combination of two sources – a tropical source and a convective source – we can calculate the relative contribution of each source.

$$\frac{[HDO]_{Obs}}{[H_2O]_{Obs}} = \frac{\chi_D[HDO]_c + (1-\chi_D)[HDO]_e}{\chi_H[H_2O]_c + (1-\chi_H)[H_2O]_e} \qquad (1)$$

We use the average depletion value above θ = 380 K, $\delta D_{Obs} = -450‰$, $\delta D_c = -210‰$ for the convective outflow, and $\delta D_e = -650‰$ for the tropical source [*McCarthy et al.*, 2004]. The fractional contribution of convected water to the total budget of this part of the stratospheric overworld determined with these inputs is $\chi_H = 0.45 \pm 0.13$.

[12] The true value of χ_H may be considerably lower. It is possible that there are seasonal and geographic variations in δD_e and it is known that convected ice has variable isotopic composition [*Gettelman and Webster*, 2005]. If we assume a heavier tropical entry value ($\delta D_e = -550‰$) Equation (1) yields $\chi_H = 0.29$; if we assume a heavier ice composition ($\delta D_c = 0‰$) $\chi_H = 0.31$; and if we substitute both $\delta D_e = -550‰$ and $\delta D_c = 0‰$, $\chi_H = 0.18$. It is also possible that systematic biases in the remote and/or in situ data sets lead to an overestimate of χ_H. For example, if the remote data sets were biased low by their 2σ uncertainty (~50‰) and the in situ observations were biased high by 2σ (100‰), correction for these hypothetical biases in equation (1) yields $\chi_H = 0.12$.

[13] Another consideration is that convection in other regions of the globe, e.g. the Asian monsoon, can potentially contribute moisture and isotopic enrichment to the overworld stratosphere [*Gettelman et al.*, 2004; *Fu et al.*, 2006]. The convective signature in water vapor in this case will dissipate before reaching North America but will still enhance the background water vapor and water isotope values. Thus, while up to 45% of the summertime overworld stratospheric water vapor above North America can be attributed to convection, the source should not be considered entirely local.

3. Discussion

[14] Convective influence on the summertime extratropical stratosphere is also indicated by remote measurements of water vapor. Observations by the microwave limb sounder (MLS) on the AURA satellite [*Livesey et al.*, 2005; *Froideveaux et al.*, 2006] show enhancements in water vapor over continental regions in summertime at θ = 380 K (Figure 3). A modeling study has shown that observations similar to those shown in Figure 3 are consistent with moistening of the lower part of the overworld stratosphere by deep continental convection [*Dessler and Sherwood*, 2004]. In that analysis the maxima in water vapor abundance (over Asia and North America) result from a combination of relatively frequent deep convection and relatively warm temperatures at the 380 K surface that allow for ice evaporation into undersaturated air. (There is no maximum over Indonesia despite frequent deep convection because low temperatures and saturated air inhibit ice evaporation.) Though these regions of convective activity are small, isentropic mixing distributes the convective signature throughout the midlatitudes.

[15] We expect the distribution of water vapor isotopic enhancement to follow that of the convective water vapor enhancements shown in Figure 3. In regions where large contributions from convection are observed we expect isotopically heavy observations. In regions with little or no convective influence we expect lighter values. We would also expect lighter isotopic values in seasons less influenced by convection. Remote observations in the fall, winter, and spring have not shown isotopic signatures that would

Table 1. Observations During the Enhanced Water Plume Encountered on 7 July, 2005[a]

	$[H_2O]$, ppmv	[HDO]*, ppmv	δD, ‰
Plume	15.6 ± 0.9	11.2 ± 1.3	−280 ± 50
Background	6.3 ± 0.3	3.8 ± 0.7	−460 ± 90
Difference	9.3 ± 0.8	7.4 ± 1.7	−210 ± 60

[a] Average conditions during the encounter: altitude 15.7 km, θ = 390 K, T = 208 K, P = 115 mbar, Lat = 34°, Longitude = 265°. The transect of the plume was roughly 0.5 km thick and 17 km wide. The HDO* is HDO is normalized to VSMOW = 3.115×10^{-4}. The values determined for the plume are the average of observations where H_2O > 14 ppmv.

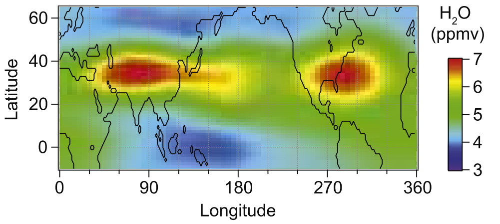

Figure 3. Observations of monthly averaged H_2O from the Aura MLS for July 2005 at $\theta = 380$ K. The θ of each MLS measurement is determined using daily temperature and pressure fields from the United Kingdom Meteorological Office (UKMO) [*Swinbank and O'Neill*, 1994]. Linear interpolation is used to determine H_2O at 380 K.

indicate convective influence [*Moyer et al.*, 1996; *Johnson et al.*, 2001].

[16] The remote MLS data and the in situ water isotope data provide a consistent picture: the overworld stratosphere over North America is significantly influenced by deep convection. At the same time, some observations remain unexplained. The in situ observations show a very small gradient in isotopic enhancement with altitude in the lowermost stratosphere, whereas the influence of deep convection is expected to decrease strongly with altitude. If the entry value for tropical air is taken as $\delta D_e = -650‰$, this implies significant convective influence at $\theta = 430$ K. One possible explanation of this is that $\delta D_e = -650‰$ is not an appropriate assumption due to seasonal and geographic variations or to measurement error. If δD_e were $-500‰$, then the AVE_WIIF observations would suggest convective influence that falls off from the tropopause to essentially zero at $\theta = 430$ K. Another possibility is that the convective outflow at $\theta = 380$ K in the Asian monsoon region is transported equatorward and joins into the general circulation of the stratosphere [*Gettelman et al.*, 2004; *Fu et al.*, 2006]. Finally, it is possible that the vertical extent of convective influence extends above the levels inferred from cloud top height estimates. Recent analysis of remote observations of water vapor suggest that convective influence of stratospheric water vapor may extend to $\theta \sim 460$ K over the Asian monsoon and ~ 410 K over North America [*Dessler*, 2006]. In situ observations at higher θ and in the tropics will help resolve these uncertainties and allow us to quantify the water budget of the stratosphere more accurately.

4. Conclusions

[17] The in situ observations presented here provide evidence for significant convective influence on the H_2O and HDO budget of the extratropical overworld ($\theta > 380$ K). Observations of water vapor plumes in the overworld indicate detrainment from convective storms occurs in the mid-latitude summertime over North America. The observations of HDO suggest that a measurable amount of water vapor is transported into the overworld stratosphere without passing through the tropical tropopause. The contribution appears substantial in the midlatitudes and may constitute a significant fraction of the stratospheric water vapor budget.

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