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A model of HDO in the tropical tropopause layer

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Abstract

ACPD

3, 4489–4513, 2003

Any theory of water vapor in the tropical tropopause layer (TTL) must explain both the abundance and isotopic composition of water there. In a previous paper, we presented a model of the TTL that simulated the abundance of water vapor as well as the details of the vertical profile. That model included the effects of “overshooting” convection, which injects dry air directly into the TTL. Here, we present results for the model after modifying it to include water’s stable isotopologue HDO (where D represents deuterium, ^2H). We find that the model predicts a nearly uniform HDO depletion throughout the TTL, in agreement with recent measurements. This occurs because the model dehydrates by dilution, which does not fractionate, instead of by condensation. Our model shows that this dehydration by dilution is consistent with other physical constraints on the system. We also show the key role that lofted ice plays in determining the abundance of HDO in the TTL. Such lofted ice requires a complementary source of dry air in the TTL; without that, the TTL will rapidly saturate and the lofted ice will not evaporate.

1. Introduction

In recent years, much progress has been made on the coupled questions of how mass is transported from the troposphere to the stratosphere and how this air is dehydrated to the low mixing ratios of the stratosphere. In particular, it has become apparent that rather than think of the tropopause as an infinitely thin surface, it is better to think of the troposphere and stratosphere as being separated by a layer several kilometers thick (Atticks and Robinson, 1983; Highwood and Hoskins, 1998). Sherwood and Dessler (2000, 2001, hereafter SD00 and SD01) defined this layer, often referred to as the tropical tropopause layer (hereafter, TTL), to run from approximately 14 to 19 km (~ 140 to ~ 70 hPa).

Air is transported into the TTL directly from the planetary boundary layer (PBL) by convection. Only a small fraction of all convection detains at such high altitudes (e.g.

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Folkins et al., 1999), but it is still enough to have an important impact on the composition of the region (Dessler, 2002). Once in the TTL, the air experiences net radiative heating and slowly ascends across the top of the TTL and into the stratosphere. Air passing through the bottom of the TTL in convective updrafts contains several tens of ppmv of water in vapor and condensate, while air slowly rising through the top of the TTL and into the stratosphere contains (on average) 3.85 ppmv of water (Dessler and Kim, 1999; SPARC, 2000). Clearly, dehydration mechanisms in the TTL are removing the majority of the water that enters.

Over the past two decades, two competing concepts of this dehydration mechanism have emerged. The first, which we call “convective dehydration,” posits that air emerges from convection fully dehydrated (on average) to stratospheric values. In this theory, the cold temperatures necessary to generate stratospheric humidities occur in “overshooting” convection (Johnston and Solomon, 1979), so called because this convection rises above or overshoots its level of neutral buoyancy (LNB), typically near 14 km (Folkins, 2002). SD01 used a simple model of the TTL to show how overshooting convection could produce the observed water vapor and ozone distributions throughout the TTL.

The second concept, which we call “gradual dehydration,” posits that air detrains from convection near its LNB at the bottom of the TTL, but with abundant water compared to the stratosphere. As the air slowly ascends towards the stratosphere, it is periodically exposed to temperatures considerably below that of the zonally averaged tropopause. During these exposures, formation and subsequent sedimentation of particles acts to dehydrate the air (Holton and Gettelman, 2001; Jensen et al., 2001). These cold events arise for a number of reasons, such as radiative cooling above deep convection (Hartmann et al., 2001) or atmospheric waves (Boehm and Verlinde, 2000; Potter and Holton, 1995), to name just two.

Both convective and gradual dehydration theories accurately predict many of the details of the water vapor distribution in the TTL and lower stratosphere (SD01, Holton and Gettelman, 2001). In the last few years, however, it has become apparent that

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the abundance of water's isotopologues might be able to discriminate between these theories.

2. Isotopes

The abundance of deuterium in water in a sample is typically expressed as a deviation from a standard, and reported in so-called delta notation:

$$\delta D = 1000 \left(\frac{([HDO]/[H_2O])_{\text{SAMPLE}}}{([HDO]/[H_2O])_{\text{SMOW}}} - 1 \right), \quad (1)$$

where $[HDO]/[H_2O]$ is the ratio of the abundance of HDO to H_2O . $(\dots)_{\text{SAMPLE}}$ is the value of this ratio in the sample, while $(\dots)_{\text{SMOW}}$ denotes the value in Standard Mean Ocean Water, in this case 3.115×10^{-4} (based on the data in Hagemann et al., 1970).

- 10 Except where noted, the sample is the water in the vapor phase of the parcel. It should be noted that Eq. (1) is most often cast as a function of the ratio of deuterium to hydrogen atoms; in that case, the standard value is one half of the value used here. However, we write the equation in terms of the ratio of water abundances for pedagogical clarity. In particular, it is clear from Eq. (1) that δD can be thought of as a measure of the ratio of $[HDO]/[H_2O]$. Therefore, processes that increase or decrease H_2O and HDO in the same proportion (such as the injection of dry air into a parcel) do not change δD . Condensation, on the other hand, preferentially removes the heavy isotopologues and therefore leads to decreases ("depletions") in δD .

- 15 In a Rayleigh distillation process, condensate forms at 100% RH and in equilibrium with the vapor. The condensate is therefore enriched in deuterium with respect to the initial vapor, while the remaining vapor is depleted. After formation, condensate is immediately removed. As condensation continues in this hypothetical parcel, new condensate will be in equilibrium with the now-depleted vapor, and the vapor will continue

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isotopically lightening. The equation describing this is:

$$d\ln R = (\alpha - 1) d\ln q, \quad (2)$$

where $R = [\text{HDO}]/[\text{H}_2\text{O}]$ in the vapor, q is the abundance of H_2O vapor and α is the temperature-dependent fractionation factor. The vertical profile of δD obtained by integrating Eq. (2) assuming a hypothetical saturated air parcel rising through the troposphere is known as the “Rayleigh curve.”

Moyer et al. (1996) first pointed out that the HDO abundance in the stratosphere is higher than predicted by a Rayleigh distillation process, or, as one might say, is “under-depleted”. They hypothesized that lofting of ice formed at lower altitudes (which would be isotopically heavy) followed by evaporation of the ice near the tropopause, could explain the under-depletion. They also speculated that non-equilibrium effects might be responsible. Keith (2000) considered both HDO and H_2^{18}O and concluded that the non-equilibrium effects could not explain the simultaneous under-depletion of both D and ^{18}O . He also showed a calculation demonstrating that ice lofting could plausibly explain the HDO under-depletion.

Both Moyer et al. (1996) and Keith (2000) assumed air entering the stratosphere experiences a single temperature-pressure history, neglecting the possibility of mixing between air masses in the TTL having experienced different histories. It has been noted, however, that mixing of unlike air masses will necessarily lead to under-depletion because of the curvature of the Rayleigh fractionation vs. H_2O saturation vapor pressure relation (Eriksson, 1965; Gedzelman, 1988).

To see this, consider mixing between two parcels. The water vapor mixing ratio in the mixed parcel is the weighted average of the mixing ratios of the initial parcels, $f[\text{H}_2\text{O}]_1 + (1-f)[\text{H}_2\text{O}]_2$, where f is the mixing fraction. δD in the mixed parcel, however, is not a simple weighted average:

$$\delta D_{\text{mix}} = 1000 \left(\left(\frac{f[\text{HDO}]_1 + (1-f)[\text{HDO}]_2}{f[\text{H}_2\text{O}]_1 + (1-f)[\text{H}_2\text{O}]_2} \right) / \left(\frac{[\text{HDO}]}{[\text{H}_2\text{O}]} \right)_{\text{SMOW}} \right) - 1. \quad (3)$$

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This is shown graphically in Fig. 1: on this plot are the Rayleigh curve (gray line) and a mixing line between saturated parcels at 210 hPa and 100 hPa (dashed line). As suggested by Eq. (3), the mixing line is not straight in this coordinate system, and mixing between two parcels on the Rayleigh curve necessarily creates an under-depleted parcel. Also note that because the parcel at 210 hPa has much higher mixing ratios of H₂O and HDO than the 100-hPa parcel, the depletion of the mixed parcel is nearly equal to the depletion in the 210-hPa parcel until the mixture is almost entirely comprised of the 100-hPa parcel; e.g. on the mixing line in Fig. 1, the sharp bend occurs at a mixture of 95% 100-hPa parcel and 5% 210-hPa parcel. Using a simple two-level model of the TTL, Johnson et al. (2001a) argued that this process alone could account for HDO under-depletion in the stratosphere.

3. Model

In this paper, we adapt SD01's model of the TTL to investigate HDO, one of H₂O's stable isotopologues, in the TTL. SD01 showed that the model was capable of simulating average H₂O, O₃ gradients, and energy balance in the TTL. In a recent paper (Sherwood and Dessler, 2003), it is shown that the model can also produce realistic seasonal variations in CO₂ and H₂O in the lower stratosphere.

Briefly, the model divides the TTL into two regions, representing (1) a region of convective overshooting where convection penetrates into the TTL and (2) a "capped" region with no convection reaching the TTL. The two regions are each represented by a one-dimensional model, which are coupled together by a large-scale diabatic circulation and by parameterized isentropic mixing (a minor error in the isentropic mixing parameterization from SD01 was found and corrected for this study, which led to small (< 10%) changes in water vapor). The diabatic circulation includes a stratospheric "drain" (i.e. sinking) in the overshooting region in accord with the calculations of Sherwood (2000). The temperature profile in each region is specified from radiosonde observations during the season being simulated; the temperature profile in the over-

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shooting region is the mean of observations from the Indonesian convection region, while temperatures in the capped region are the mean from Natal, Brazil. For more details, see SD01.

At each time step, the model simulates convective penetration into the overshooting region of the TTL. Each convective event is assigned a random value of convective available potential energy (CAPE). The parcels are assumed to remain moist adiabatic and saturated during convection, with ice water content assumed to be four times the H₂O vapor content. The remainder of the ice is assumed to have fallen out of the TTL. The value of four for the ice/water ratio was set in SD01 in order to generate stratospheric humidity in agreement with measurements. There is a detailed discussion of the model, including sensitivity studies, in SD01.

Parcels are assumed to mix uniformly into the environment between the level of neutral buoyancy of the convection, which we assume is 140 hPa, and the level of maximum overshoot, which is determined from the value of CAPE assigned to that event. After mixing with ambient TTL air, the resulting mixed parcel is negatively buoyant and will descend to the level of neutral buoyancy. There the overshooting parcel mixes both vapor and ice into the environment; no insertion of air occurs above this level. It is important to remember that the altitude where the parcels mix and the altitude where they detraining are different, with the mixing altitude being higher.

The net result of the mixing and detrainment includes cooling between the mixing height and the detrainment height, and insertion of subsaturated air at the detrainment height whose tracer characteristics are a mix of TTL and boundary layer air. Though about 40% of the overshoots penetrate the tropopause in our model, the detrainment profile is concentrated mostly in the lower half TTL. In the capped region, constituent distributions are determined solely by advection and mixing with the overshooting region.

How much water a parcel delivers to the TTL in our model is determined by how high (and therefore, how cold) the parcel gets prior to mixing and detrainment – the lower the altitude that a parcel mixes and detrains in the TTL, the more water the parcel

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carries into the TTL. This can be seen in Fig. 2, which is a plot of the rate of water detrainment from convection. As one can see, injection of water into the TTL is indeed sharply peaked near the bottom of the TTL. The injection of mass is also weighted toward the bottom of the TTL, but much less strongly than water vapor (see Fig. 1 of 5 Sherwood and Dessler (2003) for the plot of mass detrainment rate).

Air near the bottom of the TTL is relatively moist compared to the stratosphere, with mixing ratios of \sim 10–20 ppmv (e.g. Fig. 6 of SD01). As TTL air rises towards the stratosphere, deeper convecting parcels, which have experienced colder temperatures and therefore carry lower water abundances, detrain into the air mass. This dehydrates the 10 TTL and keeps it below saturation, so no condensation occurs (outside convection) in the TTL. The water is in the form of both ice and vapor; this ice is assumed to have zero fall velocity and to sublime with a time constant of one day. Because one day \ll dynamic time constants, one can consider evaporation of ice to be essentially instantaneous, except for the transport of small cloud amounts to the other model region.

The SD01 model contained a number of tunable parameters. The amount of overshooting convection was tuned so that cooling in the TTL balanced the compressional heating diagnosed from the stratospheric “drain” calculations. The vertical profile of parcel mixing was specified arbitrarily as uniform in pressure, though an alternate case was considered in which the profile decreased with height. The mixing between up-drafts and their environment was prescribed following Emanuel (1991). The amount of 15 ice retained in overshoots (a factor of four in SD01) prior to mixing was tuned to produce the correct moisture at the top of the TTL. The isentropic horizontal mixing rate, a constant, was tuned to give appropriate humidity below the TTL. All of this tuning was done in a permanent December-January-February (DJF) run, the only condition 20 simulated in that study.

The SD01 model performed well in reproducing key details in the time-average DJF vertical profiles of water vapor, ozone, and cloud ice within the TTL. Importantly, SD01 confirmed that these features were not strongly sensitive to the parameter values or assumed circulation (for a thorough discussion, see SD01, Sect. 4.d).

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4. Modeling isotopes

The standard statement of Rayleigh distillation assumes that condensate is immediately removed after formation. If, however, the condensate is ice, then the slow rate of diffusion within ice suggests that as long as the ice doesn't evaporate, it will not affect the composition of the vapor even if it remains in the parcel. As a result, we will assume that the abundance of HDO vapor in a convecting parcel can be described by the Rayleigh curve. Our model uses standard HDO equilibrium fractionation factors (Merlivat and Nief, 1967; Majoube, 1971). We use the fractionation factor for liquid water for $T > 273\text{ K}$ and that for ice for lower temperatures.

One way to explain the observed HDO under-depletion is to suppose that condensation occurs in a supersaturated environment. In this situation, loss of HDO to the condensate is limited by diffusion of HDO to the particles' surface and less depletion occurs than is predicted by Rayleigh behavior. Both Keith (2000) and Johnson et al. (2001a) looked into this as a possible explanation, and both rejected it, because while supersaturation helped explain stratospheric HDO, it worsened agreement between measurements and model for H_2^{18}O . Because of this prior work, as well as a lack of knowledge of what the magnitude of supersaturation one might find in cumulus convection, we will not consider supersaturation further. Clearly, this is a subject for future work when more data become available.

Another key assumption in the model is the abundance of deuterium in the ice (hereafter, δD_{ice}). There are no measurements of δD_{ice} near the tropical tropopause, so it is not obvious how to describe this parameter in the model. We will test several ways of specifying of this quantity, and show that key results are insensitive to which one we select.

The SD01 model had a small source of water (5 ppbv/day) in the TTL to account for oxidation of methane (SD01, Sect. 3.d). A small fraction of atmospheric methane is deuterated and oxidation of these molecules would act as a minor source of HDO. The measurements of HDO that we will be comparing to have this source of HDO

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subtracted out. In order to make comparisons consistent between these data and our model, we will therefore not include in our model any production of H₂O or HDO from methane oxidation.

As in SD01, the simulations presented in this paper are permanent DJF conditions.

- 5 Permanent June-July-August results (not shown) are similar except for a small increase in δD associated with warmer temperatures. Finally, the discussion above emphasizes the isotopologue HDO. We have also simulated H₂¹⁸O, another stable isotopologue, but we find that it provides no information not found in the HDO simulations. For the sake of brevity, we will therefore not discuss any H₂¹⁸O results in this paper.

10 5. Data

There are few observations of δD in the upper troposphere and stratosphere to compare against our model, but those that exist paint a consistent picture. Mid-latitude in situ aircraft observations by Ehhalt (cited in Gedzelman, 1988) and Smith (1992) showed under-depletion in the upper troposphere. Measurements of H₂O, CH₄, and their isotopologues in the middle stratosphere have also been used to infer the global and annual average δD of air entering the stratosphere. These measurements also show under-depletion, with values of $-670 \pm 70\text{‰}$ (Moyer et al., 1996) and $-679 \pm 20\text{‰}$ (Johnson et al., 2001b) for δD entering the stratosphere.

15 Spectra measured by the Atmospheric Trace Molecule Spectroscopy (ATMOS) experiment (Gunson et al., 1996) have been reprocessed to retrieve δD throughout the TTL (Kuang et al., 2003). These data, which are plotted in Fig. 1, show a nearly constant δD profile (after removal of methane oxidation) near -650‰ , close to the stratospheric values inferred by others, from the stratosphere all the way down to $\sim 12\text{ km}$, below which δD increases.

20 Recent in situ measurements obtained during NASA's CRYSTAL-FACE mission show a large variability of isotopic depletion in the TTL (C. Webster, personal communication, 2003). It is unclear how to interpret comparisons between these point

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measurements, virtually all obtained near convection, and our model, which simulates the tropical average. More work on this issue will be undertaken when final versions of these data become available.

6. Model simulations

- 5 Simulations of HDO depletion δD are shown in Fig. 3. In these simulations, we assign a constant δD_{ice} of $-565\text{\textperthousand}$ to the retained ice and vary the amount of retained ice in the convecting parcels prior to mixing (sensitivities to δD_{ice} will be discussed below). The far left line has no retained ice, with lines lying progressively to the right retaining ice equal to one, two, four, six, and eight times vapor. Also shown are the ATMOS data
10 (Kuang et al., 2003).

Much like the ATMOS data, our model shows near-constant values of δD throughout the TTL for all values of retained ice. As discussed above, most of the water enters the TTL near the bottom of the TTL. δD in the bottom of the TTL is therefore determined by the combination of the Rayleigh curve (which determined the depletion of the
15 vapor), δD_{ice} , and amount of ice retained. As the air rises toward the stratosphere, dehydration in our model occurs through injection of dry air. From Eq. (1), we see that isotopic depletion is really a measure of the ratio of $[\text{HDO}]/[\text{H}_2\text{O}]$, so this injection of dry air, which reduces HDO and H_2O in equal proportions, does not change the ratio of $[\text{HDO}]/[\text{H}_2\text{O}]$, and therefore does not change δD . Thus, the isotopic composition of
20 air at the bottom of the TTL determines the isotopic composition throughout the TTL in our model. As we will show shortly, using different values of δD_{ice} does shift the lines to the left and right, but still produces near-zero gradients in δD . As a result, we conclude that this near-zero gradient is a general property of our model, and is in good qualitative agreement with the available TTL data.

In contrast with our model runs, we plot on Fig. 1 a line that simulates the vertical profile of depletion that would be produced by in situ condensation, as might be caused, for example, by dehydration through formation and sedimentation of thin cirrus
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clouds (e.g. Jensen et al., 1996; Jensen et al., 2001). We start with saturated air at the base of the TTL and then, as the air mass rises, condense sufficient water to maintain 100% relative humidity. We assign a value of $-65\text{\textperthousand}$ to air at the base of the TTL, around 140 hPa, in agreement with ATMOS observations there. As one can see, the in situ condensation leads to strong fractionation between the base of the TTL and about 100 hPa. Above the temperature minimum at 100 hPa, no further condensation or fractionation occurs. Kuang et al. (2003) showed a similar comparison and concluded that their ATMOS data did not support such in situ condensation. It should be noted, however, that mixing in the TTL or condensation under supersaturated conditions will reduce the vertical gradient of this in situ condensation line. Clearly, our analysis cannot definitively exclude in situ condensation; more model studies and observations will be needed to do that.

As discussed in SD01, some ice must be retained in detraining parcels. With no ice, the injection of dry air into the TTL dehydrates too effectively, so that air entering the stratosphere is far too dry. SD01 determined that a retained ice/vapor ratio of four in air prior to mixing into the TTL produced a water distribution in the TTL and entering the stratosphere in agreement with observations.

Adopting this value of four for the ice/vapor ratio, we present in Fig. 4 four simulations from our model where different values of δD_{ice} are. The simulations all produce under-depletion and a near-zero gradient of δD throughout the TTL. This emphasizes the conclusion from the previous runs that this near-zero gradient is a fundamental property of our model. It is also clear that our model is sensitive to the choice of δD_{ice} . The choice of δD_{ice} changes the depletion of water injected into the lower TTL, and this δD is then transported upward to the stratosphere, while the air is dehydrated through injection of dry air.

Setting δD_{ice} to $-565\text{\textperthousand}$, produces δD in the TTL and lower stratosphere in close quantitative agreement to the ATMOS measurements. While reasonable, without measurements of the δD_{ice} in the TTL, we cannot know for certain how realistic this choice of δD_{ice} is. This emphasizes the importance of a measurement of the isotopic compo-

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sition of the ice detraining into the TTL.

It should be noted that we are not specifying the isotopic content of the ice, just the isotopic content of that portion of the ice crystal that sublimates in the TTL. Because of the slow rate of diffusion of molecules in ice, the outer coating of the ice, which was deposited last, is expected to be isotopically more depleted than the core. For a sufficiently large ice crystal, it is reasonable that only the outer (and isotopically more depleted) portion of the crystal might evaporate in the TTL, with the remainder evaporating at lower altitudes. Thus, it is possible that the bulk composition of ice lofted into the TTL is potentially different from the composition of ice that evaporates there.

One obvious question is whether our results are unduly influenced by the specification of the depletion of retained ice δD_{ice} as a constant value. To investigate this, we performed runs where δD_{ice} is not constant, but is chosen randomly from a normal distribution prior to every call of the convective subroutine of our model (this subroutine simulates one convective event and is called thousands of times during each run of the model). We varied both the average value and standard deviation of the δD_{ice} distributions. In these runs (not shown), only changes in the average value of δD_{ice} mattered; varying the standard deviation from zero (i.e. constant δD_{ice}) through large values resulted in virtually identical δD profiles in the TTL.

As a further check, we also performed runs where the ice becomes more depleted (e.g. δD_{ice} becomes more negative) with increasing mixing height of the overshooting convection. For each convective event, we set δD_{ice} equal to the depletion predicted by the Rayleigh curve at a pre-determined distance below altitude where the parcel mixes into the environment. In Fig. 4, we plot a run (dashed line) where this distance is 3.5 km; e.g. if the parcel mixes with the environment at 16.5 km, we set δD_{ice} equal to the Rayleigh curve value at 13 km. This run again shows only a small vertical gradient in predicted δD . Other offset distances show a similarly shaped curve, with the predicted curve shifting as the altitude offset changes. The reason for this is that most of the water in the TTL entered over a narrow altitude range – narrow enough that δD_{ice} is effectively constant in this run. It seems unlikely that δD_{ice} would vary strongly enough

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with altitude to produce a significantly non-uniform vertical distribution of isotopic composition in the model TTL.

Figure 5 shows the budgets of HDO and H_2O vapor averaged over both the overshooting and capped regions for the run where the ice/vapor ratio is set to four and δD_{ice} is set to a constant value of -565% . The convective tendency term, Q_2 , is the time rate of change of vapor due to convection alone (see also Fig. 8 of SD01 and the accompanying discussion in that paper for more discussion of the water budget in the model). Detraining air also carries ice (which is not counted in Q_2), which in our model sublimates with a time scale of one day, releasing HDO and H_2O vapor. Advection, comprised of both vertical and horizontal components, and isentropic (i.e. horizontal) mixing both transport water within and between the regions.

In the lower half of the TTL (i.e. below the tropopause), the primary balance for both HDO and H_2O vapor is between moistening by ice sublimation and drying by convective injection of dry air. Advection and isentropic mixing are both small. Because ice sublimation is the dominant source of water vapor in the lower half of the TTL, δD there is strongly affected by the assumed value of δD_{ice} , which is obvious from the runs in Fig. 4.

Air parcels detraining in the upper half of the TTL have experienced such cold temperatures that their H_2O and HDO (both vapor and ice) content is small (compared to the ambient TTL). This drying tendency is balanced by upward advection of high H_2O and HDO mixing ratio air (remember that our model has no methane oxidation in it, so the mixing ratios of H_2O and HDO decrease with increasing altitude throughout the TTL). The insertion of dry air into the upper TTL dehydrates the TTL, but without changing the value of δD . This conclusion holds for all runs, regardless of what is assumed for the retained ice/vapor ratio or δD_{ice} .

One might ask whether there's anyway we can generate a strong gradient in δD in the TTL in our model. The only way we can think of is if air detraining in the upper half of the TTL had significant amounts of water (ice or vapor) and if this water had a depletion significantly different from that of the lower TTL. This would probably require

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strong vertical gradients in both the retained ice/vapor ratio and in δD_{ice} . It seems likely, however, that such gradients would produce water vapor distributions in the TTL and lower stratosphere that were unrealistic.

Previous studies (Moyer et al., 1996; Keith, 2000; Johnson et al., 2001a; Kuang et al., 2003) have concluded that parcel-based models of air ascent produce air that is much more depleted (i.e. follows the Rayleigh curve) than the air observed in the TTL and lower stratosphere. This shows that either ice lofting or irreversible mixing between parcels with different histories must play a significant role. At least one numerical model currently under study (J. Smith, personal communication) appears to depart successfully from the parcel-based calculations, and it is likely that others will as well, indicating that stratospheric “under-depletion” may be consistent with a number of dehydration paradigms. However, it can be difficult to disentangle the effects of mixing and ice lofting in complex numerical models, and it is also difficult to know whether or not irreversible mixing is significantly exaggerated by their numerics. Examination of the details of vertical variation of isotopologues, rather than just the final value in the stratosphere, may be the key to discriminating among models.

7. Conclusions

Two explanations for “under-depletion” in the lower tropical stratosphere (HDO abundances larger than predicted by the Rayleigh fractionation) have been suggested in the literature before: lofting of ice formed in the upper troposphere into the TTL, where it sublimates, and mixing between air masses with different thermodynamic histories. In this paper, we consider for the first time both processes and model them quantitatively, using a model that explicitly represents convective transport in a dynamically and energetically consistent way.

In our model’s TTL, most of the water vapor detrained near the bottom of the TTL. As this air slowly ascends towards the stratosphere, it is dehydrated by injection of dry air from convection. Because isotopic depletion is really a measure of the ratio

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of $[HDO]/[H_2O]$, dehydration by dilution does not change the depletion. As a result, our model predicts near-constant profiles of δD in the TTL, in agreement with the recent ATMOS observations. This conclusion is not dependent on our assumptions for retained ice/vapor ratio or the isotopic composition of the retained ice.

Quantitatively accurate simulations in the TTL can be achieved by adjusting the retained ice/vapor ratio and/or the isotopic composition of the retained ice δD_{ice} . If we assume that retained ice is four times the vapor, which SD01 found was required to reproduce the water vapor distribution in the TTL accurately, then setting δD_{ice} to -565‰ reproduces the δD distribution in the TTL. Measurement of this quantity will provide an important new constraint on models, including the present model.

One underappreciated aspect of ice lofting into the TTL is that it must be accompanied by a persistent source of dry air, without which the TTL would rapidly saturate and the lofted ice would sediment out of the TTL without evaporating. In our model, overshooting convection is the source of dry air that prevents the TTL from saturating (SD01).

The work described in this paper is in fundamental agreement with the work of Johnson et al. (2001a). Using a calculation that included two detrainment levels and no lofted ice, they suggested that most of the water vapor but a small fraction of the mass entering the stratosphere was dehydrated to the temperatures found at 11 km. Most of the mass entering the stratosphere, on the other hand, experienced very cold temperatures and therefore had small water vapor abundances (relative to the stratosphere). Johnson et al. emphasized the composition of air entering the stratosphere; our work shows that this explanation also explains the distributions within the TTL, too.

Johnson et al. also did not explore vertical variations in depletion or ice lofting. Recent observations (Sherwood, 2002) indicate that reevaporated ice plays a dominant role in attaining stratospheric moisture. It is not clear how such lofting/reevaporation would affect the dilution arguments, nor is it clear from existing studies whether the necessary convective insertion process (with or without ice) is consistent with other physical constraints on the system.

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Our model shows that dehydration from dilution is consistent with the physical processes of the TTL. In agreement with Johnson et al., most of the water vapor in the TTL and entering the stratosphere was injected near the bottom of the TTL around 14 km, with subsequent dehydration as the air slowly ascends provided by injection of dry air from overshooting convection. In addition, our analysis shows that the same ideas explain the near-zero gradient seen in the TTL in the ATMOS data qualitatively, and with some tuning, quantitatively. Our model also emphasizes the important role lofted ice plays in the TTL.

In conclusion, the model used here was previously shown in SD01 to simulate the average H_2O , O_3 gradients, and energy balance in the TTL successfully. Here, we have shown that the same model is also capable of simulating the near-constant value of δD seen in the ATMOS data throughout the TTL. While the final values of H_2O and δD are sensitive to the amount and isotopic composition of lofted ice, respectively, we find that the near-constant δD profiles simulated in the TTL result from the underlying physics of the model, not from arbitrary model parameters. The near-constancy of simulated δD results from the fact that dehydration in SD01 occurred through a dilution process, in which ascending air is mixed with very dry air, reducing the concentrations of all isotopes at about the same rate; lofted ice was important only in the lower TTL, where the initial values of H_2O and its isotopologues were established.

We anticipate that vertical variation of δD in the TTL, rather than the final value, will be the most powerful discriminator of dehydration theories. We suspect that models in which dehydration within the TTL occurs primarily outside convection will have difficulty in reproducing δD in the TTL as uniform as that simulated here (even if other mixing mechanisms enable them to get to the observed end-point in the lower stratosphere). Further corroboration of the Kuang et al. (2003) observations, which support the near-uniformity we have simulated, are of great interest.

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knowledge productive discussions with A. Andrews, D. Johnson, D. Keith, Z. Kuang, E. Moyer, and R. Smith. This work was supported by NASA EOS/IDS grant NAG-59632 and NASA grants NAG-11849 and NAG-11897 to the University of Maryland and Yale University, and by a NASA New Investigator Program in Earth Science grant NAG-58936 to the University of Maryland.

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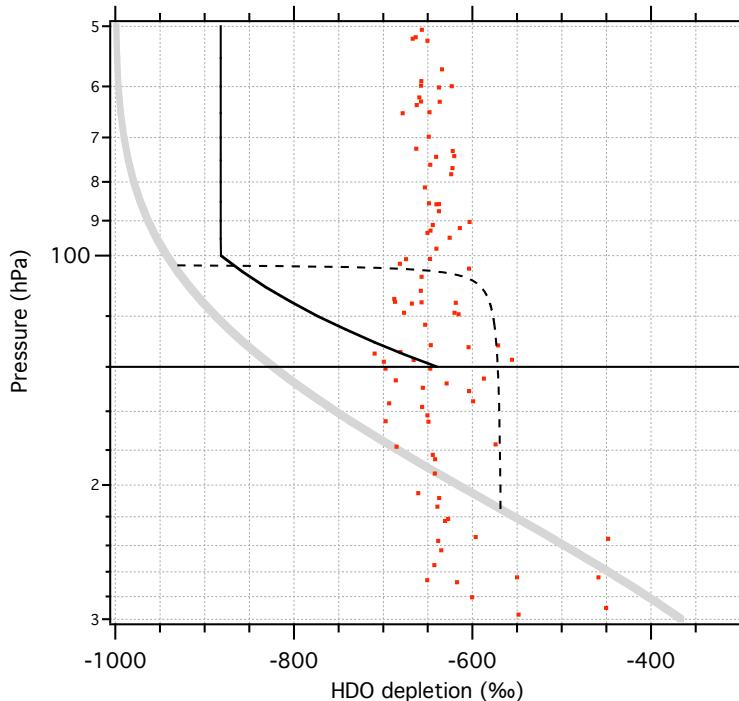


Fig. 1. HDO depletion δD (\textperthousand) in the vapor vs. altitude (hPa). The thick gray line is the Rayleigh curve, calculated for a parcel ascending from the surface pseudoadiabatically (see text). The dots are the ATMOS data in the TTL (Kuang et al., 2003), with the effects of methane oxidation removed. The dashed line is a mixing line between saturated parcels at 210 and 100 hPa, with H_2O and HDO abundances set by the Rayleigh curve at these altitudes. The thin solid line is a calculation of TTL depletion as a result of in situ condensation, for air beginning with the observed depletion at the base of the TTL.

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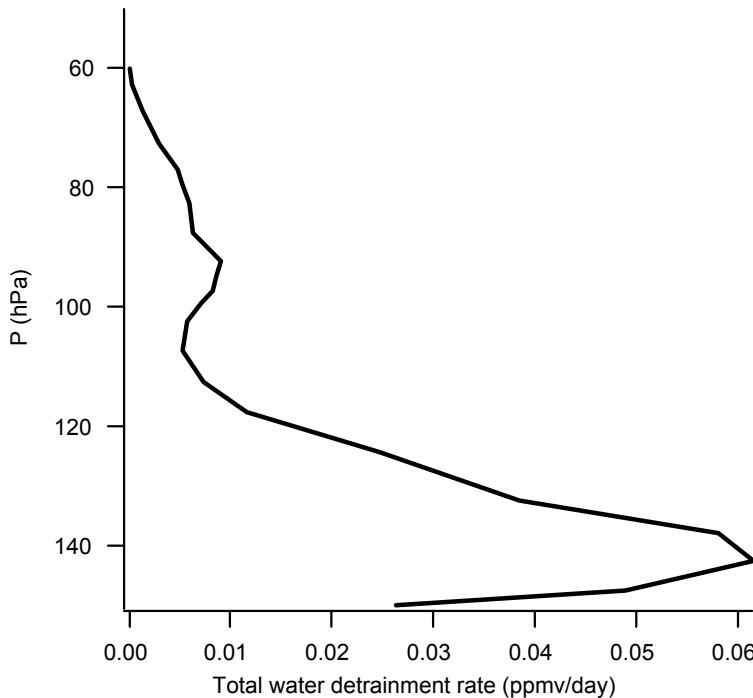


Fig. 2. Total water (vapor + ice) detrainment rate (ppmv/day) as a function of pressure (hPa). This quantity is detrainment mass flux (kg/hPa/day) times water mixing ratio (ppmv) divided by mass on that level (kg/hPa).

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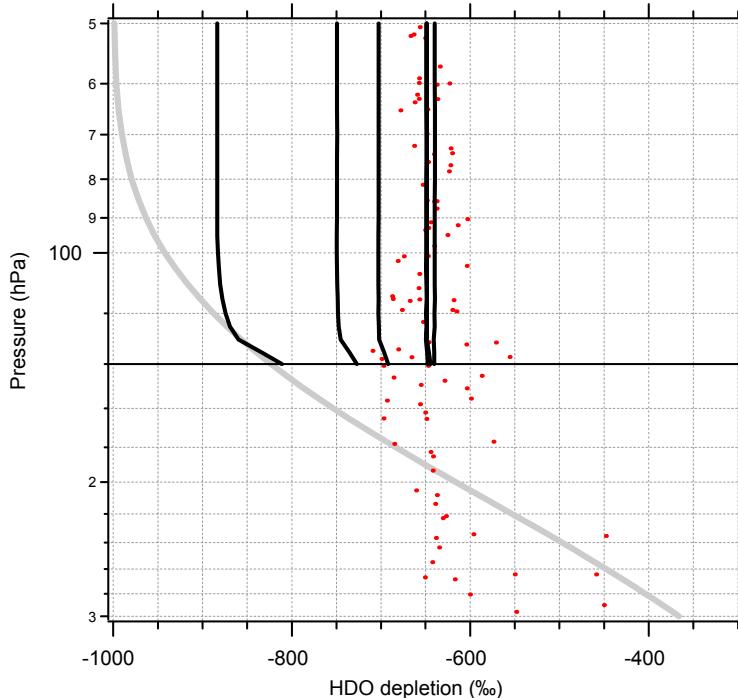


Fig. 3. HDO depletion δD (%) in the vapor vs. altitude (hPa) for several simulations; each result is an average of δD in the two regions of the model. The thick black curves are runs with retained ice equal to (from left to right) zero, one, two, four, six, and eight times vapor. For all runs, δD_{ice} was set to -565‰ . These runs have no methane oxidation in them. The dots are the ATMOS data in the TTL (Kuang et al., 2003), also with the effects of methane oxidation removed. The thick gray line is the Rayleigh curve, calculated for a parcel ascending from the surface pseudoadiabatically (see text).

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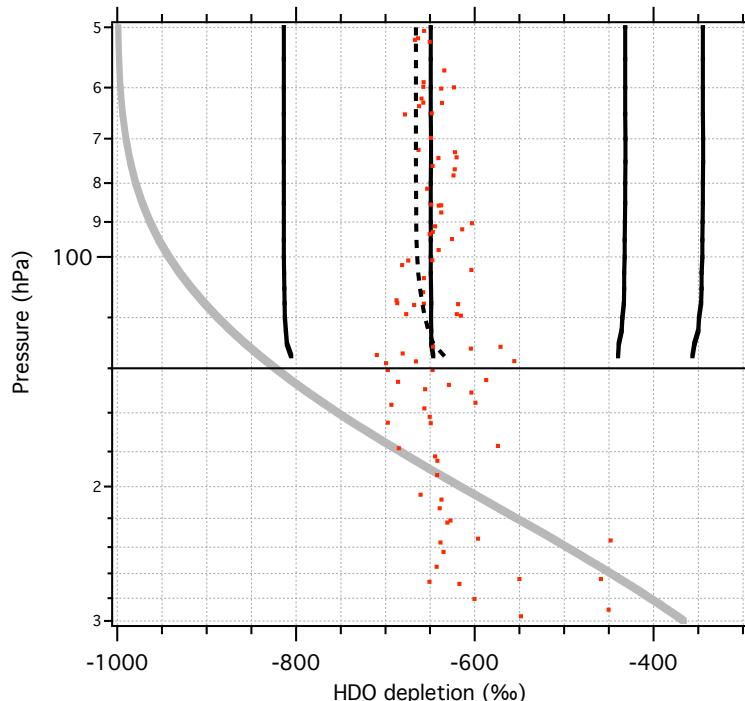


Fig. 4. HDO depletion δD (%) in the vapor vs. altitude (hPa) for several simulations; each result is an average of δD in the two regions of the model. The thick black curves are runs with δD_{ice} set to -750% , -565% , -314% , and -160% . The dashed line is a run with δD_{ice} equal to the Rayleigh curve at an altitude 3.5 km below the mixing altitude of the parcel. These runs have no methane oxidation in them. The dots are the ATMOS data in the TTL (Kuang et al., 2003), also with the effects of methane oxidation removed. The thick gray line is the Rayleigh curve, calculated for a parcel ascending from the surface pseudoadiabatically (see text).

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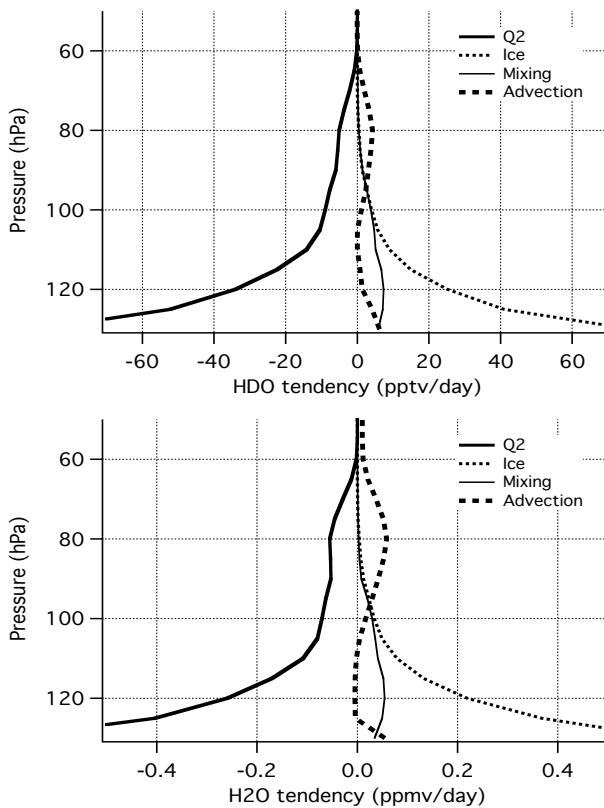


Fig. 5. Budget for HDO (top) and H₂O (bottom) in the TTL for δD_{ice} from the -565% run, averaged over the model's two regions. See the text for the definitions of the lines.

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