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# A reconsideration of the initial conditions used for stable water isotope models

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**Abstract.** Many of the “simple models” used to study and explain the global distributions of stable water isotopes assume that the initial isotope content of an air mass can be estimated from local conditions at the oceanic source through the application of a global-scale closure equation. Using a general circulation model fitted with isotope tracer diagnostics, we show that the closure equation is generally invalid at the local scale, leading to a systematic bias in the modeled isotope contents. The quantitative results obtained in earlier simple model studies may need some adjustment in light of this bias, particularly if deuterium excess was used to characterize conditions prevailing at the evaporative source.

## 1. Background

The two stable isotopes of water, HDO (deuterium) and H<sub>2</sub><sup>18</sup>O (oxygen 18), have long been used to characterize paleoclimate through established relationships between isotope concentrations and climatic variables. The most important relationship, derived from present-day measurements, is the decrease in isotope concentration with decreasing temperature in middle to high latitudes [Dansgaard, 1964]. Paleoclimatologists assume that this present-day isotope-temperature relationship also describes how isotope concentrations in a region vary in time in response to changes in climate. This allows them to estimate paleotemperatures from measurements of isotope concentration in ice cores [Jouzel et al., 1987a; Dansgaard et al., 1993].

Such interpretations of paleodata rely in part on analyses with models of isotope behavior. These models, which range from simple Rayleigh-type models to isotopic general circulation models (GCMs), account for the fractionation processes in the atmospheric water cycle that govern the isotope content of precipitation. In this report, we address an important, systematic bias inherent in the initial conditions used for the most common simpler models. The report recommends that the bias be corrected with GCM isotope data.

## 2. Simple Isotope Models

Simple isotope models typically transport an idealized parcel of air poleward from an oceanic source. As the parcel cools, water vapor condenses along with some of the water isotope. Parameterizations of isotope fractionation physics give the condensate a higher isotope concentration than the remaining vapor, and thus each successive condensation event in the parcel depletes the vapor's isotope concentration further. As a

result, isotope concentrations in the vapor, and thus in precipitation, decrease with the temperature of the parcel in agreement with the measured isotope-temperature relationship. The precise relationship generated by the model can be examined in the context of the air parcel's initial state, the model's assumptions regarding isotopic fractionation, and the model's precipitation physics.

The earliest simple isotope models assumed equilibrium fractionation during changes of phase [Dansgaard, 1964; Friedman et al., 1964; Craig and Gordon, 1965]. The standard Rayleigh model, in differential form, can be written

$$\frac{d\delta v}{1 + \delta v} = (\alpha - 1) \frac{dm_v}{m_v} \quad (1)$$

$$1 + \delta P = \alpha(1 + \delta v), \quad (2)$$

where  $\alpha$  is the equilibrium fractionation factor (a function of isotope species, temperature, and phase change),  $m_v$  is the amount of vapor in the air parcel,  $\delta v$  is the isotope content of the vapor, and  $\delta P$  is the isotope content of the condensate. (The value  $\delta v$  is thus the  $\delta D$  or  $\delta^{18}O$  of the vapor, where  $\delta D$  and  $\delta^{18}O$  are the concentrations of deuterium and oxygen 18 relative to those in standard mean ocean water or SMOW.) Equation (1) can easily be extended to allow a fraction of the condensate to remain in the air parcel as it travels further poleward [Craig and Gordon, 1965]. Though simple, these early models successfully explain the observed variations of isotope concentration in precipitation with temperature. They also successfully reproduce the observed linear relationship between  $\delta D$  and  $\delta^{18}O$  in precipitation. The values  $\delta D$  and  $\delta^{18}O$  in modern precipitation generally fall on the “meteoric water line,” which can be written as  $\delta D = 8\delta^{18}O + 10$  [Craig, 1961]; deviations from that line can be determined from the deuterium excess  $d$ , defined as  $\delta D - 8\delta^{18}O$  [Dansgaard, 1964].

These models, however, have a major limitation. The initial isotope concentration in the air parcel  $\delta_{v0}$  is not easily estimated because the vapor and the ocean surface waters are not

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in isotopic equilibrium and because the overlying air is subsaturated, so that  $\delta_{vo}$  differs from  $\delta_E$ , the isotope content of the evaporating water [Craig and Gordon, 1965]. To overcome this difficulty, Craig and Gordon [1965] suggested two approaches: (1) treating exchange processes at the ocean surface in detail and (2) assuming a material balance with the mean precipitation. The first approach has generally proven impractical because of the paucity of adequate observations and the conceptual difficulty of separating the advective contributions to  $\delta_{vo}$  from the evaporative contributions. Merlivat and Jouzel [1979] therefore followed the second approach. They started with a relationship between  $\delta_{vo}$  and  $\delta_E$  based on kinetic fractionation considerations:

$$1 + \delta_E = (1 - k) \frac{\frac{1 + \delta_{ocean}}{\alpha} - h(1 + \delta_{vo})}{1 - h}, \quad (3)$$

where  $k$  is a kinetic fractionation parameter,  $h$  is the relative humidity, and  $\delta_{ocean}$  is the isotope content of seawater. Then, by invoking global-scale closure, they argued that  $\delta_E$  can indeed be equated to  $\delta_{vo}$ , leading to the following formula:

$$1 + \delta_{vo} = \left( \frac{1}{\alpha} \frac{1 - k}{1 - kh} \right) (1 + \delta_{ocean}). \quad (4)$$

Merlivat and Jouzel [1979] were somewhat justified in applying the closure assumption because they were addressing global-scale isotope behavior and were not applying (4) to a regional study.

Numerous later studies with simple isotope models, however, have used (4) in order to relate initial isotope concentrations in the air parcel to conditions (temperature, humidity, and wind speed) at the evaporative source [e.g., Jouzel and Merlivat, 1984; Aristarain et al., 1986; LeTreut et al., 1988; Johnsen et al., 1989; Petit et al., 1991; Fisher, 1992; Ciais et al., 1995]. These studies typically interpret polar isotope concentrations in terms of polar temperatures and evaporative source meteorology, often through the analysis of simulated deuterium excess variations. Because the closure arguments used to derive (4) are not valid at the regional scale, these studies may suffer from an important inaccuracy. In this report, we address the extent of this inaccuracy through the analysis of GCM-generated data.

### 3. GCM Evaluation of the Closure Assumption

A GCM fitted with isotopic tracer diagnostics can provide a clean, direct test of the closure assumption. "Isotope GCMs" [Joussame et al., 1984; Jouzel et al., 1987b; Hoffmann and Heimann, 1993] model the global geochemical cycles of water isotopes by transporting isotopic tracers across the Earth's surface and between atmospheric grid boxes during a full simulation of climate. These models have successfully reproduced the main features of the observed global distributions of  $\delta D$  and  $\delta^{18}O$  in precipitation on both an annual and a seasonal basis. Recent GCM studies of isotope behavior during the last glacial maximum (LGM) [Joussame and Jouzel, 1993; Jouzel et al., 1994] have focused on evaluating a critical assumption in paleoclimatology, the assumption that the isotope-temperature relationship measured for present-day conditions (essentially a "spatial" relationship, relying on geographically distinct measurements) is equivalent to the "temporal" relationship valid for a single point experiencing a sequence of different climates.

This assumption, which allows the estimation of paleotemperatures from isotope concentrations in ice cores, groundwaters, and speleothems, is now being challenged by paleothermometry measurements in central Greenland [Cuffey et al., 1995; Johnsen et al., 1995].

Because the flux  $E_i$  of an isotopic tracer across the ocean surface in the GCM is computed using detailed information on isotope concentrations in near-surface vapor, the GCM effectively calculates  $\delta_{vo}$  using the first approach of Craig and Gordon [1965]; that is, the GCM does not rely on the closure assumption to compute  $\delta_{vo}$ . To examine the closure assumption then, the simulated isotope concentration in vapor within each near-surface grid box can be compared to that computed with (4), using the values of  $\alpha$ ,  $k$ , and  $h$  obtained for that box. Of course, the GCM has its own limitations; the simulated values of  $\delta_E$  and  $\delta_{vo}$  depend on numerous parameterizations of physical processes in the GCM, and some of these parameterizations may be overly simplistic. The simulated  $\delta_{vo}$  values, for example, could be influenced by errors in the estimation of planetary boundary layer stability. Nevertheless, the GCM fitted with isotope tracer diagnostics is perhaps the best environment available for evaluating the closure assumption given the extreme paucity of  $\delta_E$  and  $\delta_{vo}$  observations over the ocean.

The final 2 years of a 5-year simulation of the LGM [Jouzel et al., 1994] provided the special diagnostics needed for this comparison. (Although the corresponding diagnostics for a simulation of present-day climate are not currently available, the main results of an analysis with such data would undoubtedly be the same.) At every time step and for every near-surface grid box, the GCM computed the diagnostic

$$m_{vi}^* = \frac{m_v R_{SMOW} (1 + \delta_{ocean})}{\alpha} \frac{1 - k}{1 - kh} \quad (5)$$

for both deuterium and oxygen 18, where  $m_v$  is the mass of water vapor in the grid box and  $R_{SMOW}(1 + \delta_{ocean})$  is the concentration of isotope in near-surface water, which is slightly higher than that in SMOW. The isotope concentration consistent with (4)  $\delta_{vo}^*$  is afterward computed with

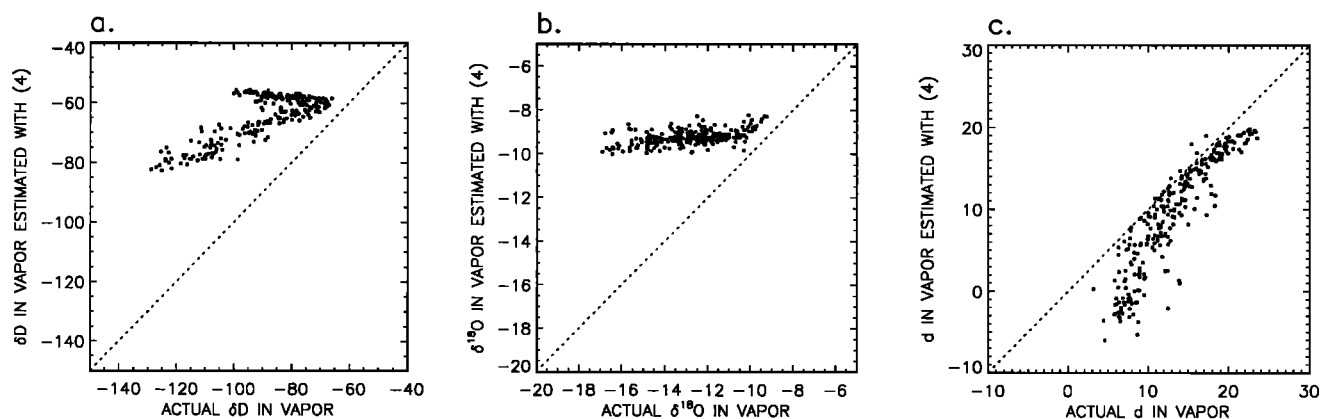
$$\delta_{vo}^* = \frac{1}{R_{SMOW}} \frac{\int_{\Delta T} m_{vi}^* dt}{\int_{\Delta T} m_v dt} - 1, \quad (6)$$

where  $\Delta T$  is the time period under consideration. Similarly, the actual isotope concentration in vapor simulated during the same time period is computed with

$$\delta_{vo} = \frac{1}{R_{SMOW}} \frac{\int_{\Delta t} m_{vi} dt}{\int_{\Delta t} m_v dt} - 1, \quad (7)$$

where  $m_{vi}$  is the actual mass of water isotope vapor in the grid box.

A scatterplot of  $\delta_{vo}^*$  versus  $\delta_{vo}$  is provided in Figure 1 for both deuterium and oxygen 18 in northern hemisphere summer (June, July, and August (JJA)). Each point in the plot represents a near-surface grid box lying over a pure open ocean



**Figure 1.** Scatterplots (June, July, and August (JJA)) comparing the actual isotope concentrations in water vapor over the ocean ( $x$  axis) against the concentrations obtained when applying the closure assumption of Merlivat and Jouzel [1979] ( $y$  axis): (a)  $\delta D$ , (b)  $\delta^{18}O$ , and (c) deuterium excess.

grid square. Squares containing some land (i.e., coastal squares) are not included; squares containing sea ice are also not included, so that the latitudes considered lie between  $50^{\circ}S$  and  $50^{\circ}N$ .

For both deuterium and oxygen 18,  $\delta_{vo}^*$  clearly overestimates  $\delta_{vo}$ . For oxygen 18, for example, the  $\delta_{vo}^*$  values of  $-10\text{‰}$  or higher lie far above the simulated  $\delta_{vo}$  values of  $-18$  to  $-10\text{‰}$ , which are, in fact, consistent with the observed values in the northern oceans, which range from  $-14$  to  $-10\text{‰}$  [Craig and Gordon, 1965]. Notice that the bifurcation seen for deuterium in Figure 1a is much more obvious than that for oxygen 18 in Figure 1b. The upper branch of the scatterplot for  $\delta D$  contains points for tropical squares, which feature weak temperature variations, whereas the lower branch contains points for middle- to high-latitude grid squares, which feature strong temperature variations. For deuterium,  $\delta_{vo}^*$  varies with temperature because it is mostly influenced by temperature-dependent equilibrium fractionation processes; the temperature-independent kinetic fractionation parameter  $k$  in (4) is small. For oxygen 18, on the other hand, the effects of equilibrium fractionation are about 10 times smaller than they are for deuterium, and thus kinetic fractionation is relatively more important. As a result,  $\delta_{vo}^*$  does not vary as much with temperature and shows little variation in Figure 1b, so that a bifurcation is not as clear.

We hereafter refer to the difference  $\delta_{vo}^* - \delta_{vo}$  as the “error” or “bias” associated with using the closure assumption of Merlivat and Jouzel [1979]. Figures 2a and 2b show the zonal mean values (over ocean) of the  $\delta D$  and  $\delta^{18}O$  biases obtained using (4). The biases show a clear latitudinal structure, with the errors increasing with latitude. This characteristic, shown here for JJA results, is also seen in the data for the other seasons.

The latitudinal variation in the bias is consistent with some recent calculations performed with the Goddard Institute for Space Studies (GISS) isotope GCM by the authors with Alexandre Armengaud of the Centre National de la Recherche Scientifique, France. These calculations, to be documented at a later date, show that the relative contribution of an oceanic evaporation source to the water vapor overlying it decreases with increasing latitude in the northern hemisphere, because of, in part, the reevaporation in high latitudes of falling rainwater derived from tropical sources, for example. A smaller

relative contribution of an oceanic source to the overlying  $\delta_{vo}$  implies a greater violation of the closure assumption.

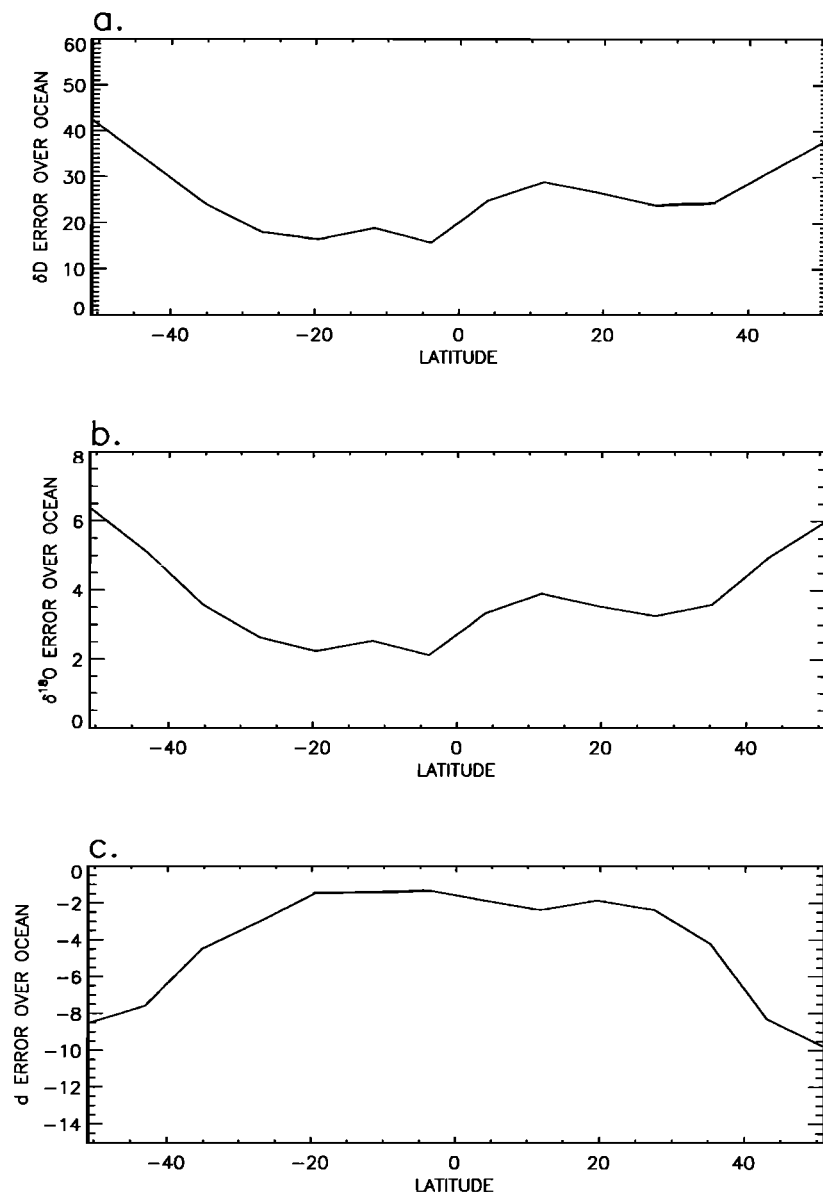
Figures 1c and 2c show the corresponding plots for deuterium excess. The biases obtained from the use of (4) can be large and negative; again, the largest errors occur at higher latitudes.

#### 4. Implications

Direct confirmation of the GCM-generated  $\delta_{vo}$  values is currently limited to comparisons with extremely rare measurements, such as those taken by Craig and Gordon [1965]. Thus our evaluation of the closure assumption with GCM data should be regarded as preliminary. With this caveat we now describe some implications of the biases in Figures 1 and 2 for the interpretation of results from simple isotope models.

Simple Rayleigh-type isotope models are used to check the consistency between observed and predicted isotope-temperature relationships, to examine in detail the various microphysical processes affecting isotope concentrations in the atmosphere and to identify the key parameters, aside from local temperature, that affect the isotope content of precipitation (e.g., the temperature and relative humidity at the evaporative source). As seen in Figure 2 and in corresponding plots for the other seasons, initial conditions in these simple models require adjustments of  $15\text{--}40\text{‰}$  for  $\delta D$  and  $2\text{--}6\text{‰}$  for  $\delta^{18}O$  to correct the biases associated with the use of the closure assumption, under LGM conditions and presumably under other conditions as well.

Although these biases are important, being of the same order as the glacial-interglacial isotope shifts observed in polar ice cores, they are not critical given that neither  $\delta D$  nor  $\delta^{18}O$  is used by itself in simple isotope models to estimate paleotemperatures, the paleothermometer is mainly based on the observed present-day isotope-temperature relationship. Still, the quantitative results of many past studies using simple isotope models should be adjusted. As an example, because the bias increases with decreasing sea surface temperature, previous estimates of the influence of this temperature  $T_e$  on the isotope content of precipitation should be reduced. Aristarain *et al.* [1986] estimated  $\partial\delta^{18}O/\partial T_e$  to be  $-0.55\text{‰}/^{\circ}C$ ; the magnitude of this slope is apparently too high.



**Figure 2.** Latitudinally averaged errors (over the ocean) obtained when the closure assumption of *Merlivat and Jouzel* [1979] is applied: (a)  $\delta D$ , (b)  $\delta^{18}O$ , and (c) deuterium excess.

The bias becomes much more important for simple model analyses of deuterium excess. Deuterium excess above an oceanic source should increase with increasing temperature and decrease with increasing relative humidity in accordance with well-understood equilibrium and kinetic fractionation principles [Merlivat and Jouzel, 1979; Johnsen *et al.*, 1989]. The scatterplots in Figure 3 show that this is indeed the case for the GCM-generated deuterium excess in water vapor. The LGM JJA points can be fitted through multiple regression to the line

$$d = 0.380T_e - 23.8h + 23.1, \quad (8)$$

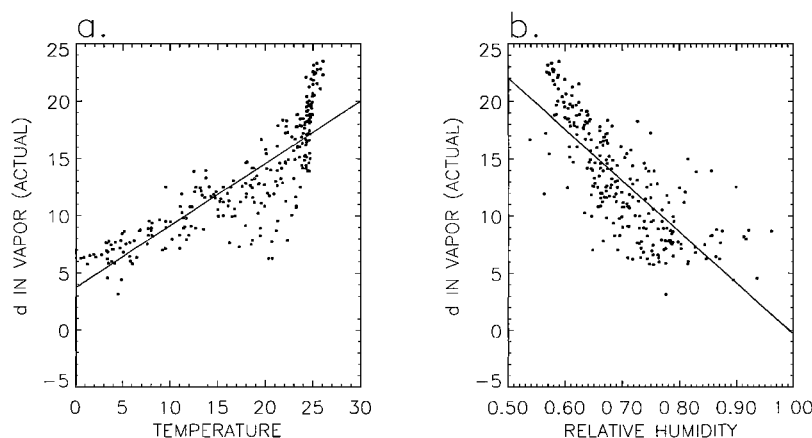
which is associated with a correlation coefficient of 0.89. (The slopes in the equation look somewhat different from those in Figure 3 because relative humidity is itself negatively correlated with temperature.)

The closure assumption, however, exaggerates the sensitivity of deuterium excess to  $T_e$  and  $h$ . Multiple regression on the

LGM JJA data from the GCM leads to the following relationship:

$$d^* \equiv \delta D^* - 8\delta^{18}O^* = 0.598T_e - 38.1h + 25.5, \quad (9)$$

which is associated with a very high correlation coefficient ( $r = 0.99$ ) because the temperature and humidity effects are built directly into (4). Simple isotope models have shown that information regarding the temperature and relative humidity at the evaporative source is partially preserved in an air parcel as it is transported poleward, so that evaporative source conditions can be retrieved from polar deuterium excess values [Petit *et al.*, 1991; Ciais and Jouzel, 1994]. In relying on (4), however, these models effectively extract evaporative source information using an equation similar to (9) rather than the more appropriate (8). Also, note that the biases indicated in Figures 1c and 2c are as large as, if not larger than, the deuterium excess signal



**Figure 3.** Scatterplots (JJA) comparing (a) deuterium excess in water vapor against sea surface temperature and (b) deuterium excess in water vapor against relative humidity.

observed in paleodata. Glacial-interglacial changes in  $d$  are typically only 5‰ [Jouzel *et al.*, 1982; Johnsen *et al.*, 1989].

White [1995] and Peel *et al.* [1996] have proposed the use of polar deuterium excess paleodata to isolate that part of the  $\delta D$  or  $\delta^{18}O$  signal related to a change in evaporative source temperature. The source temperature change is estimated by assuming that  $d$  depends only on source temperature (an overly simple assumption, even for simple isotope models, as indicated in Figure 3). A correction is then applied to the inferred local temperature, again using information derived from the simple model. The validity of such a correction, entirely based on simple models, does not seem warranted given the biases described above, which are particularly large for deuterium excess. The fact, however, that a relationship between  $d$  and sea surface temperature is seen in Figure 3a suggests that this approach should still be explored.

## 5. Recommendation

We are still convinced of the usefulness of simple isotope models. They are invaluable tools for understanding isotope processes occurring at various stages of the water cycle, both in the real world and in the GCM environment. Still, the difficulty of accurately defining their initial conditions, which would allow isotope concentration in precipitation to be related to conditions at the evaporative source, must be kept in mind. To overcome this difficulty, we recommend that the initial conditions for these models be taken from GCM-generated estimates of  $\delta D$  and  $\delta^{18}O$  in grid boxes overlying ocean squares, since the GCM calculations are not limited by the closure assumption of Merlivat and Jouzel [1979]. The relevant monthly data generated with the GISS model, for both present-day and LGM conditions, are available from the authors.

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