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Alexandre Armengaud, Randal Koster, Jean Jouzel, Philippe Ciais. Deuterium excess in Greenland snow: Analysis with simple and complex models. *Journal of Geophysical Research: Atmospheres*, 1998, 103 (D8), pp.8947-8953. 10.1029/98JD00274 . hal-02923817

**HAL Id: hal-02923817**

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Submitted on 28 Oct 2020

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## Deuterium excess in Greenland snow: Analysis with simple and complex models

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**Abstract.** A simple Rayleigh-type isotope model, typical of those used to develop algorithms for extracting climatic information from stable water isotope paleodata, is evaluated against the more complex and presumably more reliable calculations of a general circulation model (GCM) fitted with isotope tracer diagnostics. The evaluation centers on an analysis of how the temperature  $T_e$  of an oceanic moisture source affects the deuterium excess  $d$  of Greenland precipitation. The annual  $T_e$ - $d$  relationship derived from the GCM diagnostics is largely reproduced by the simple isotope model when the latter is properly initialized. This, coupled with the fact that the GCM itself reproduces observed isotope behavior, suggests that the simpler model's atmospheric calculations are indeed adequate for isotope studies. Furthermore, the GCM results support the idea, originally developed with the simpler models, that polar deuterium excess values contain information on meteorological conditions at distant evaporative sources.

### 1. Introduction

The stable isotopes of water, HDO and H<sub>2</sub><sup>18</sup>O, have been measured in ice cores and other paleowaters in varying concentrations. Through a detailed analysis of current isotope concentration fields, isotope/climate relationships have been derived which allow the extraction of paleoclimatic temperatures from paleowater measurements (see Jouzel *et al.* [1997] for a recent review). A related isotopic quantity, deuterium excess, is now being used to infer additional paleoclimatic information. Deuterium excess  $d$  was defined by Daansgaard [1964] as  $d = \delta D - 8\delta^{18}O$ , where  $\delta$  indicates a permil deviation from the corresponding isotope ratio in standard mean ocean water (SMOW). The factor 8 comes from the meteoric water line, which defines the locus of modern precipitation in a  $\delta D/\delta^{18}O$  plot [Craig, 1961].

Using a simple evaporation model and a Rayleigh-type precipitation model, Merlivat and Jouzel [1979] inferred that the deuterium excess of precipitation primarily depends on the mean relative humidity above the evaporative (oceanic) source for the moisture. Jouzel *et al.* [1982] then interpreted the reduced glacial  $d$  values (relative to modern values) in an East Antarctic core as a reflection of higher relative humidity over the oceanic areas providing moisture for Antarctic precipitation. Johnsen *et al.* [1989] pointed out that  $d$  is also significantly affected by the temperature of the moisture source and examined  $d$  variations with respect to absolute (rather than relative)

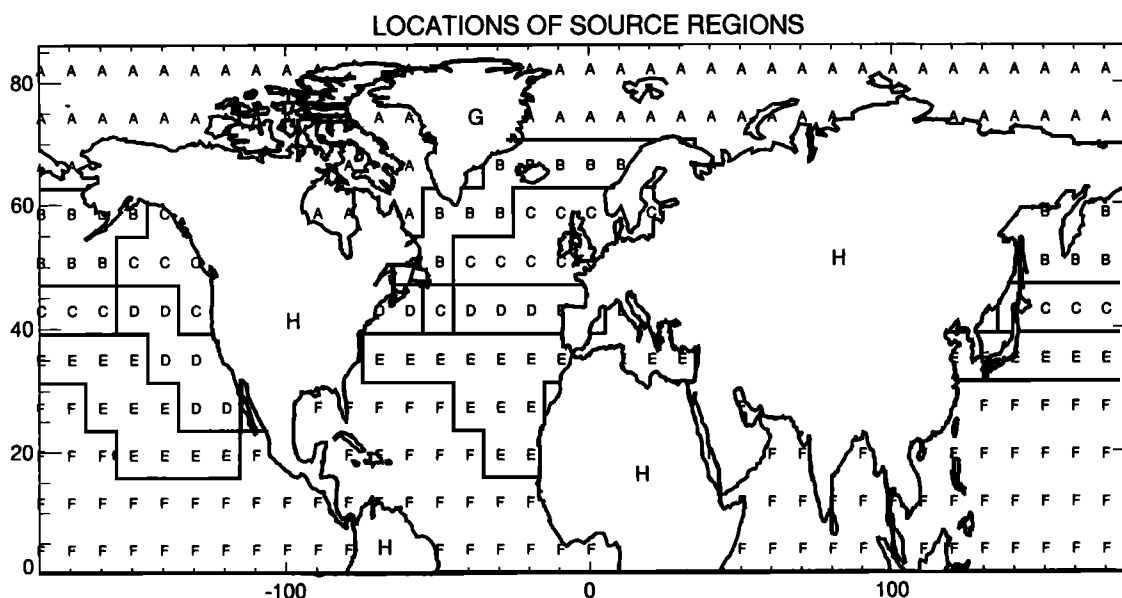
humidity, leading Daansgaard *et al.* [1989] to interpret the abrupt  $d$  change at the termination of the Younger-Dryas in Greenland's Dye 3 core in terms of a rapid retreat of sea-ice cover. The dual importance of humidity and temperature at the evaporative source has also been recognized for Antarctica [Petit *et al.*, 1991; Ciais and Jouzel, 1994; Ciais *et al.*, 1995; Fisher, 1991].

The Rayleigh or Rayleigh-type distillation models (hereinafter often referred to as "simple isotope models") usually applied in these studies essentially model isotope behavior within isolated air masses transported poleward from an ocean source. The idealized paths traversed by these air masses are determined by prescribed initial and final states for temperature and pressure. The simple isotope models account for the interplay between cloud microphysics and the fractionation processes occurring at each phase change of the water. They cannot, however, account for the complexity of dynamical processes that lead to the formation of precipitation. Furthermore, Jouzel and Koster [1996] recently showed that the standard approach used in these models for specifying the initial isotope contents within the air parcels introduces a systematic bias that can significantly affect the simulated relationships between deuterium excess and evaporative source conditions.

An alternative approach to studying global water isotope behavior is to incorporate the isotopic cycles into an atmospheric general circulation model (GCM), which does simulate the dynamical complexity of the atmosphere and which avoids the noted initial conditions bias in the simple Rayleigh-type models. Isotope tracer diagnostics have been incorporated into at least four different GCMs [Joussaume *et al.*, 1984; Jouzel *et*

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Paper number 98JD00274.  
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**Figure 1.** Locations of the tracer source regions. The tracer water for an oceanic source region (regions A–F) is not allowed to evaporate from the continental or sea-ice portion of any grid cell.

*al.*, 1987; Hoffmann and Heimann, 1993; R. Matthieu, personal communication, 1997], and simulations of isotope behavior have been performed by each modeling group for both present-day and glacial conditions [Joussaume and Jouzel, 1993; Jouzel *et al.*, 1994; Hoffmann and Heimann, 1995; Andersen, 1997]. The isotope GCMs can also “tag” moisture evaporating from selected source regions and then trace it through the GCM’s atmosphere until it precipitates. Joussaume *et al.* [1986], Koster *et al.* [1986], and Druyan and Koster [1989] used this tagging approach to study the contributions of various evaporative sources to continental precipitation, and Koster *et al.* [1992] and Charles *et al.* [1994, 1995] used it to examine how the sources of polar precipitation affect its isotope content.

In the present study, we use the  $8^\circ \times 10^\circ$  Goddard Institute for Space Studies (GISS) isotope GCM [Jouzel *et al.*, 1987] to determine how sea surface temperature (SST) conditions at an evaporative source affect the deuterium excess of Greenland precipitation. We then repeat the study using a simple Rayleigh-type distillation model to establish the extent to which the simpler model can reproduce the results from the more complex GCM. This particular evaluation is important given the numerous recent applications of simple models to the  $d/SST$  problem [Johnsen *et al.*, 1989; Dansgaard *et al.*, 1989; Barlow *et al.*, 1993; White *et al.*, 1995, 1997; Fisher, 1990, 1991, 1992]. Clarifying the relationships between deuterium excess over Greenland and evaporative source conditions also paves the way toward improved utilization of the extensive  $\delta D$  and  $\delta^{18}O$  data, covering at least 100 ka, now available from the Greenland Ice Core Project (GRIP) and Greenland Ice Sheet Project 2 (GISP2) ice cores [e.g., Dansgaard *et al.*, 1993].

## 2. Experimental Design

The GISS isotope GCM is chosen for this study because it is quite effective in reproducing the observed patterns of  $\delta D$  and  $\delta^{18}O$  across the globe [Jouzel *et al.*, 1987]. In central Greenland, the focus of this study, the GCM generates annual  $\delta D$ ,

$\delta^{18}O$ , and  $d$  values of  $-237\text{‰}$ ,  $-31\text{‰}$ , and  $12\text{‰}$ , respectively, which are reasonably similar to the observed values of about  $-270\text{‰}$ ,  $-35\text{‰}$ , and  $9\text{‰}$ , respectively, at the GRIP and GISP2 sites [Johnsen *et al.*, 1992; Grootes *et al.*, 1993; White *et al.*, 1997]. (An exact match with the point observations is not, in any case, expected or necessarily desired given the spatial scale represented by the GCM’s grid cells.) The GISS isotope GCM, however, when run at an  $8^\circ \times 10^\circ$  resolution, does fail to reproduce the observed seasonal cycles of isotope concentration over Greenland [Jouzel *et al.*, 1987]. This deficiency should be kept in mind when interpreting the results presented below.

Although both temperature and humidity at an evaporative source have an influence on the deuterium excess of Greenland snow, we use temperature alone to define oceanic evaporative source regions in the present experiment. For each ocean grid cell in the northern hemisphere we determine the July 1 temperature  $T_e$  from the SST climatology standardly used in the GISS GCM [Hansen *et al.*, 1983]. The ocean grid cells are then collected into six source regions using the following criteria:

Source region A	$T_e < 5$
Source region B	$5 < T_e < 10$
Source region C	$10 < T_e < 15$
Source region D	$15 < T_e < 20$
Source region E	$20 < T_e < 25$
Source region F	$25 < T_e$

Figure 1 presents the geographical locations of these regions and of two additional land regions, one representing Greenland (region G) and the other representing all other land surfaces (region H). Note that the locations of the ocean sources remain constant despite the fact that the SSTs vary seasonally in the experiment according to the prescribed climatology.

For each of the six oceanic source regions, we also define two additional tracers, one representing the HDO molecules

and the other representing the  $\text{H}_2^{18}\text{O}$  molecules that evaporate from the source. This allows us to track the isotopic evolution of the source's water as it is transported to other regions through the atmosphere [Koster *et al.*, 1992].

The design of this experiment is similar to that of Charles *et al.* [1994] in that it isolates the sources of Greenland precipitation in an attempt to explain isotopic variations there. The present experiment, however, is unique in that it separates the evaporative sources according to temperature rather than geography and follows independent isotopic tracers from each source. In the present experiment, the isotope analyses focus on water traveling directly from the oceanic sources to the Greenland site. Although the water evaporating from the continental regions G and H must, at some earlier time, have been derived from oceanic sources, this continental water is not considered in the isotope analyses.

The experimental design allows a direct comparison of GCM results, including those for deuterium excess, with output from simple isotopic models (see section 4). The tracer model is integrated for 27 months, with tracer precipitation rates continuously monitored to produce monthly totals. To account for model spin-up, data from the first three months are neglected.

### 3. GCM Results

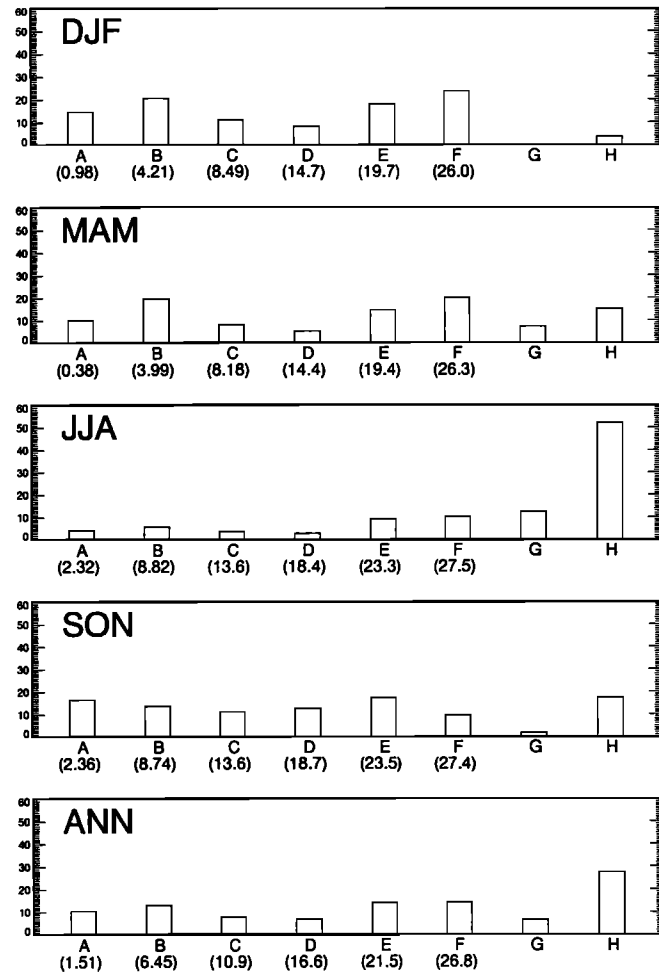
Our analysis focuses on the GCM grid cell in central Greenland ( $40^\circ\text{W}$ ,  $74^\circ\text{N}$ ) which contains the site of the GRIP and GISP2 ice cores. Following the procedure outlined by Koster *et al.* [1986, 1992], we use the  $\text{H}_2\text{O}$  tracer diagnostics in the GCM to separate the precipitation in central Greenland into relative contributions from each of the eight source regions in Figure 1. These simulated relative contributions are presented for each season and for the annual mean in Figure 2. All oceanic sources contribute to Greenland precipitation, though in varying amounts. The contribution of the distant tropical ocean (regions E and F) is indeed quite significant, particularly in winter and spring. The extratropical source regions C and D contribute less, perhaps because of their smaller areas in the Atlantic. Simulated summer precipitation in central Greenland is dominated by contributions from northern hemisphere continents.

Of more relevance to the present study is the isotopic nature of the precipitated water from each source. Following Koster *et al.* [1992], the Greenland deuterium excess associated with a specific evaporative source (say, source region A) is defined in a unique way. Consider a precipitation volume in Greenland. A certain mole fraction of that volume,  $X_{\text{HA}}$ , consists of  $\text{H}_2\text{O}$  molecules that most recently evaporated from region A. Similarly,  $X_{\text{DA}}$  and  $X_{\text{OA}}$  represent the corresponding mole fractions of HDO and  $\text{H}_2^{18}\text{O}$  derived from region A. Thus within region A's contribution to the total precipitation water in the Greenland cell, the tracer isotope concentrations are

$$\delta_{\text{DA}} = 1000\text{‰} \frac{\frac{X_{\text{DA}}}{X_{\text{HA}}} - R_{\text{SMOW,HDO}}}{R_{\text{SMOW,HDO}}} \quad (1)$$

$$\delta_{\text{OA}} = 1000\text{‰} \frac{\frac{X_{\text{OA}}}{X_{\text{HA}}} - R_{\text{SMOW,H}_2^{18}\text{O}}}{R_{\text{SMOW,H}_2^{18}\text{O}}}, \quad (2)$$

where  $R_{\text{SMOW,HDO}}$  and  $R_{\text{SMOW,H}_2^{18}\text{O}}$  represent the isotope ratios within standard mean ocean water. This leads to a deuterium excess in the water originating from region A of

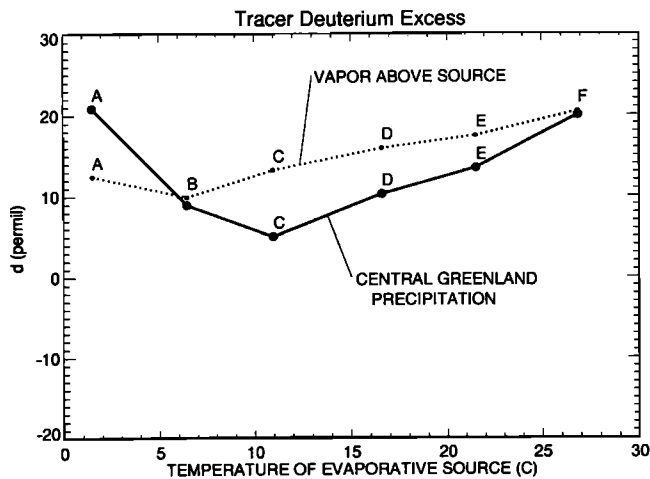


**Figure 2.** Contributions (percent) of the source regions to precipitation in central Greenland. The average temperatures of the oceanic source regions (in degrees Celsius) are shown in parentheses.

$$d_A = \delta_{\text{DA}} - 8\delta_{\text{OA}}. \quad (3)$$

Equations (1)–(3) allow us to determine the annually averaged deuterium excess in central Greenland associated with each of the source regions in Figure 1. The thick solid line in Figure 3 shows the variation of this deuterium excess with the average annual temperature of the source region. Curiously,  $d$  does not vary monotonically with source region temperature, showing instead a clear minimum at a source temperature of about  $11^\circ\text{C}$ . The mechanism behind the curve's shape (which is also seen in corresponding plots for the other Greenland squares) is not clear. Given, however, the definition of deuterium excess as the difference between two similar isotope concentrations ( $\delta\text{D}$  and  $8\delta^{18}\text{O}$ ), each of which varies nonlinearly with a host of atmospheric and surface properties, the shape is not unreasonable.

Any tracer water precipitating in Greenland clearly began its journey as water vapor overlying its evaporative source. The deuterium excess in this "initial" tracer water vapor can be calculated using equations analogous to (1)–(3). The dotted line in Figure 3 shows, for each source region, the deuterium excess for its corresponding tracer vapor averaged throughout the region's near-surface atmospheric grid boxes. Only a slight hint of a minimum at  $6^\circ\text{C}$  is seen in these excess values, which



**Figure 3.** GCM-simulated variation of deuterium excess with evaporative source temperature, for central Greenland precipitation (thick solid line) and tracer water vapor overlying the evaporative source (dotted line).

are controlled by such factors as temperature, relative humidity, and local wind speed. Thus the effective initial conditions for the isotopes prior to their transport through the GCM's atmosphere are apparently not fully responsible for the "U shape" of the deuterium excess in the precipitation curve.

#### 4. Test of the Simple Isotope Model

Again, a simple isotope model typically transports an air parcel from an oceanic source to a polar site via a highly idealized atmospheric path. The myriad and complex atmospheric transports present in the GCM (which itself is a highly idealized model of the real atmosphere) are ignored. We now test the simple isotope model of *Ciais and Jouzel* [1994] against a GCM by comparing how each transforms the isotope content of water vapor as it is transported into Greenland. More specifically, we determine the extent to which the simple model can reproduce the precipitation curve in Figure 3.

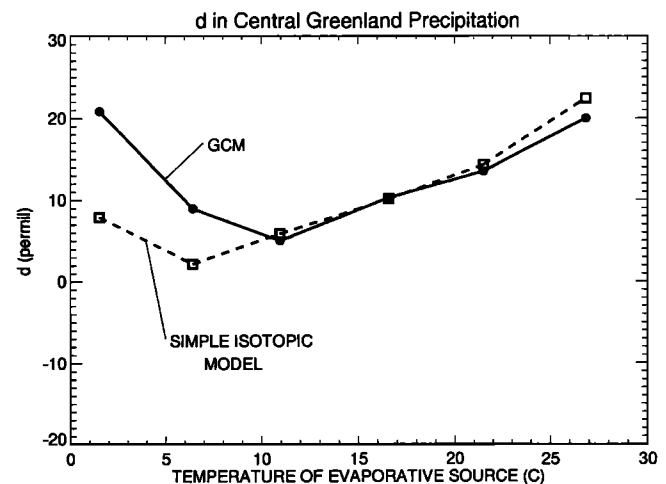
Each oceanic source in Figure 1 is considered separately in independent integrations of the simple isotope model. The specified initial conditions for the model are the sea surface temperature below the air parcel, the relative humidity within the parcel, the surface pressure, and the "tracer" values of  $\delta D$  and  $\delta^{18}O$  within the parcel's water vapor, all of which are obtained from the monthly diagnostics of the GCM simulation described above, taking areal averages over the oceanic source being considered. The tracer values of  $\delta D$  and  $\delta^{18}O$  for source region A, for example, are those computed with (1) and (2), using the mole fractions of  $H_2O$ ,  $HDO$ , and  $H_2^{18}O$  in water vapor rather than in precipitation; these are, in fact, the tracer  $\delta D$  and  $\delta^{18}O$  values that in the annual mean generated the dotted curve in Figure 3. The prescribed final state for the air parcel consists solely of the pressure and temperature over central Greenland, again determined from monthly GCM diagnostics. (The GCM-generated monthly mean temperature over central Greenland is used as the prescribed final temperature for the air parcel. Although it may have been more appropriate to use a precipitation-weighted temperature to account for the fact that in the real world, precipitation events in Greenland tend to be correlated with higher temperatures,

the information needed for this weighting was not available from the GCM diagnostics.) We do not, of course, prescribe the final states for the isotopic quantities, since these are products of the simple model's calculations.

The critical temperatures describing the phase change of water in the simple model are, for the most part, consistent with the critical temperatures used in the GCM. Perfect consistency, however, is unattainable because the GCM treats convective and nonconvective processes separately and assumes a slightly different phase change behavior in each. Ice crystals begin to appear as the temperature falls below  $-5^\circ C$ , and liquid and solid water coexist down to  $-40^\circ C$ . The supersaturation  $S$  of vapor over ice is computed as  $S = 1 - 0.003T$ , where  $T$  is the temperature in degrees Celsius.

For a given oceanic source region, we run the simple isotope model once for each month of the year and then average the monthly results (weighted by the corresponding monthly precipitation from the GCM) to produce annual means. The resulting annual deuterium excess values in central Greenland precipitation are compared to those produced by the GCM in Figure 4. The salient feature of the plot is the strong agreement between the simple model and the GCM results for temperatures above  $10^\circ C$ . For these temperatures this test indicates that the idealized air mass transport assumed by the simple isotope model is indeed a valid approximation to the more complex transports simulated by the GCM. Similar agreement is generally seen when data for individual months are compared (not shown); the agreement is indeed better for spring months, though significantly worse for winter months.

For lower source temperatures the simple model underestimates deuterium excess. As might be expected, the simple model also has trouble reproducing the individual  $\delta D$  and  $\delta^{18}O$  values in central Greenland precipitation for these source temperatures, as indicated in Figure 5. We can only speculate on the reasons for this disagreement. A limitation of the GCM itself may be responsible: in the GCM, the coexistence of sea ice and open ocean in a grid cell leads to a severe temperature discontinuity between the open-ocean portion of the cell and the overlying air, which is assumed well mixed over the cell and thus strongly influenced by the very cold sea ice. This discon-

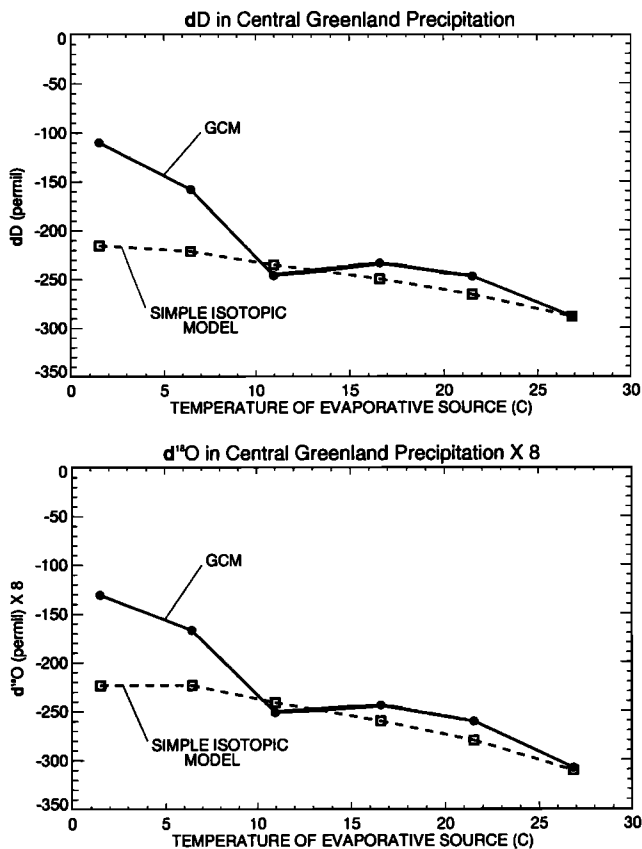


**Figure 4.** Variation of deuterium excess in central Greenland precipitation with evaporative source temperature, for the GCM (thick solid line) and the simple Rayleigh-type model (dashed line).

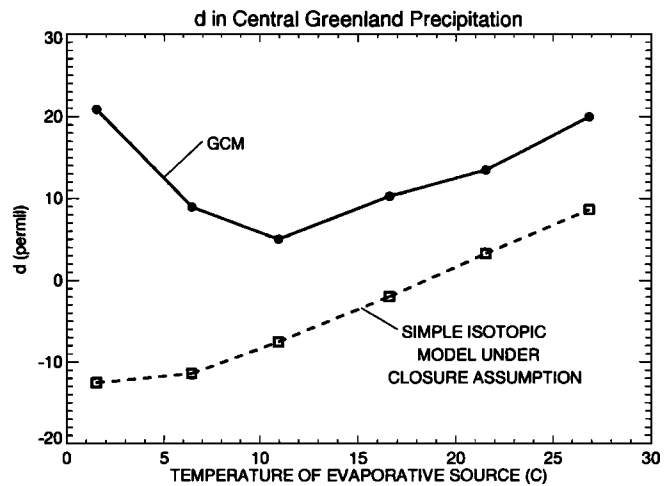
tinuity may compromise the gradual cooling of an ocean-derived vapor mass that occurs both in nature and in the simple Rayleigh-type model. This explanation is supported by the fact that the disagreement between the GCM and the simple model disappears for source region B during the warm season, i.e., during the period for which this region contains absolutely no sea ice. Given this potential GCM limitation, the testing of the simple isotope model against the GCM should perhaps focus only on the warmer source regions (C through F).

## 5. Closure Assumption

In standard practice, the HDO and H<sub>2</sub><sup>18</sup>O concentrations in a simple isotope model's air parcel are not initialized at the assumed vapor source with observed or GCM-generated data. Limitations in data availability instead often necessitate the use of the so-called "closure assumption" [Merlivat and Jouzel, 1979] to relate  $\delta D$  and  $\delta^{18}O$  in near-surface vapor to ambient relative humidity, wind speed, and surface temperature. In essence, the closure assumption equates the mean isotopic content of evaporation to that of precipitation. While this may be valid for a global-scale calculation, it is certainly invalid at the local scale and even the regional scale. Jouzel and Koster [1996], using GCM-generated data, provide strong evidence that this deficiency at the regional scale does, in fact, seem to translate into unreasonable initial values of near-surface  $\delta D$  and  $\delta^{18}O$ . They speculate that the resulting poor initialization



**Figure 5.** Variation of (top)  $\delta D$  and (bottom)  $\delta^{18}O$  in central Greenland precipitation with evaporative source temperature, for the GCM (thick solid line) and the simple Rayleigh-type model (dashed line).



**Figure 6.** Variation of deuterium excess in central Greenland precipitation with evaporative source temperature, for the GCM (thick solid line) and the simple Rayleigh-type model using the closure assumption (dashed line).

of a simple isotope model could result in unrealistic model integrations.

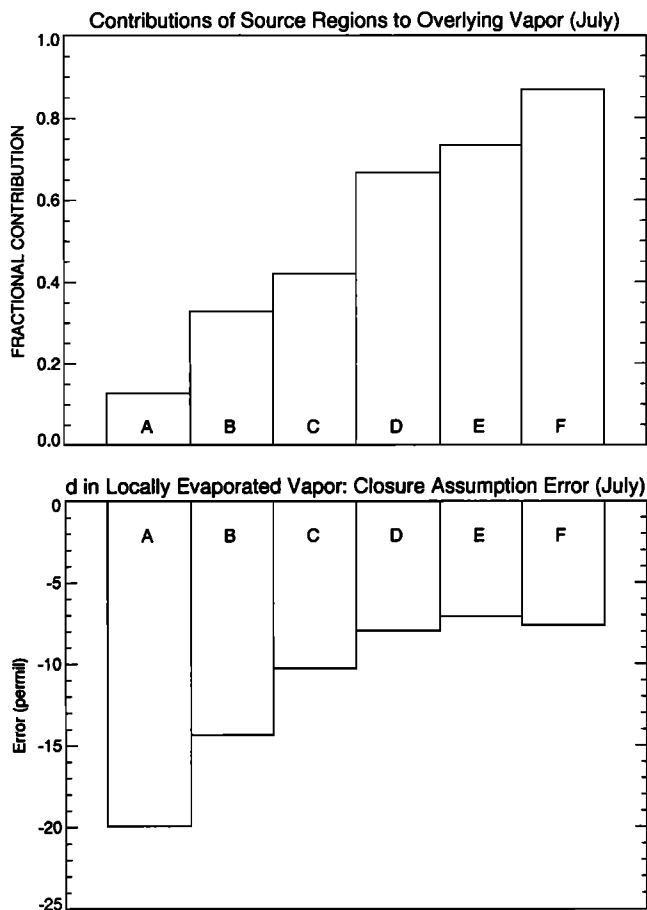
We now investigate this. We repeat the simple isotope model simulations that produced the dashed curves in Figures 4 and 5, but instead of initializing  $\delta D$  and  $\delta^{18}O$  with GCM data, we compute the initial values with the closure equation. For example, for  $\delta D$  we use

$$1 + \frac{\delta D}{1000} = \left( \frac{1}{\alpha} \frac{1-k}{1-kh} \right) \left( 1 + \frac{\delta D_{\text{ocean}}}{1000} \right), \quad (4)$$

where  $\alpha$  is the equilibrium fractionation factor,  $k$  is the kinetic fractionation coefficient (a function of wind speed),  $h$  is relative humidity, and  $\delta D_{\text{ocean}}$  is the HDO concentration in surface water [Merlivat and Jouzel, 1979]. Results are shown in Figure 6. As predicted by Jouzel and Koster [1996], the use of the closure assumption clearly results in strongly underestimated  $d$  values.

Jouzel and Koster [1996] argue that the validity of the closure assumption decreases as a region obtains a greater fraction of its overlying moisture from external sources. Their assertion is supported by the present experiment. Figure 7 (top) shows, for each oceanic source region in July, the fraction of the vapor in the lowest atmospheric layer above the region that is derived from the source region itself. Note, for example, that almost all of the vapor above source region F was evaporated locally, whereas the vapor above source region A is largely of external origin. Figure 7 (bottom) shows, for the locally derived vapor above each source region, the difference between the deuterium excess calculated with the closure assumption and that simulated by the GCM. The error is clearly larger for those source regions with large external vapor contributions.

We must point out, however, that while the underestimation of  $d$  in vapor through application of the closure equation has been confirmed by other GCMs, the magnitude of this underestimation appears model dependent [Delmotte, 1997; Andersen, 1997], giving larger values. The few available measurements of deuterium excess in oceanic vapor suggest that the closure equation underestimates  $d$  in vapor only by 3 to 4‰, i.e., by only



**Figure 7.** (top) Fractional contribution of locally evaporated water to the total vapor in the lowest atmospheric layer above each oceanic source region. (bottom) Error induced by the closure assumption in the calculation of the deuterium excess of locally evaporated water vapor.

half of what is predicted by the GISS GCM for the corresponding latitudes (S. Johnsen, personal communication, 1997).

## 6. Summary and Discussion

The Rayleigh-type distillation model, referred to here as a "simple isotope model," generally reproduces the GISS GCM's predictions of  $\delta D$ ,  $\delta^{18}O$ , and deuterium excess in Greenland precipitation when initialized with temperatures, relative humidities, and isotope concentrations from the GCM. Disagreement occurs only for the lower source temperatures, possibly due to the formulation of sea ice in the GCM. The general agreement with the more complex (and presumably more accurate) GCM supports the use of such simple models to infer remote meteorological quantities from polar isotope concentrations and, in general, to help understand and explain the global processes behind observed isotope variability. At the very least, we can say from the experiment that our simple isotope model captures the relevant isotope distillation characteristics of the GCM.

Figure 2 implies one word of caution. A significant fraction of Greenland precipitation might be derived from northern hemisphere continents, whereas simple models implicitly assume a well-behaved ocean source. The continental contribution may compromise the interpretation of real-world deute-

rium excess measurements in terms of simple model results. A separate word of caution about the comparison between the GCM and the simple model results involves their potential sensitivity to the parameter values chosen to describe fractionation processes. The robustness of our conclusions can best be established through sensitivity studies with the models used (e.g., the analyses of supersaturation functions performed by Jouzel *et al.* [1991] and Ciais and Jouzel [1994]) and through similar experiments conducted with independent models.

The simple isotope model does not reproduce the GCM results in nonpolar regions (not shown). This is not surprising, since the simple model considers only nonconvective processes and thus cannot account for the complexities of isotopic exchange during moist convection. In any case, the simple model is not designed to follow an air parcel from a cold ocean source to a warmer continental site. GCMs are preferred for such problems.

Various simple isotope models have already been used to infer relationships between polar deuterium excess and meteorological conditions (temperature and humidity) at the evaporative source [e.g., Johnsen *et al.*, 1989]. The GCM confirms the strong influence of source temperature on deuterium excess in Greenland, at least for source temperatures of 10° or more. An impact of relative humidity (not shown) is also seen in the GCM results, implying that absolute humidity at the evaporative source also affects  $d$ . (Note, however, that temperature and humidity over the GCM's oceans are correlated, so a proper separation of temperature and humidity effects is difficult.) Overall, this analysis provides strong support for the use of polar deuterium excess to extract information on remote conditions and for the use of simple models to determine the relevant relationships, provided that these models employ GCM-derived isotope concentrations rather than the closure assumption in their initialization.

**Acknowledgments.** Our colleagues at the Goddard Institute for Space Studies (GISS), particularly Reto Ruedy, Gary Russell, Rick Healy, and David Rind, provided significant scientific, computational, and logistical support for this project. The lead author, in fact, spent a year at GISS, with financial support from GISS (and thus the NASA Climate Program). Financial support also came from the Programme National d'Etudes de la Dynamique du Climat of France.

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(Received March 19, 1997; revised January 5, 1998; accepted January 7, 1998.)