Tellus (1984), *36B*, 67–72

LETTERS TO THE EDITOR

Vertical distribution of deuterium in atmospheric water vapour: problems in application to assess atmospheric condensation models

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(Manuscript received February 24; in final form April 6, 1983)

ABSTRACT

The paper assesses the use of the author's data by Rozanski and Sonntag to support a multi-box model of the vertical distribution of deuterium in atmospheric water vapour, in which exchange between vapour and falling precipitation produces a steeper deuterium concentration profile than simpler condensation models. The mean deuterium/altitude profile adopted by Rozanski and Sonntag for this purpose is only one of several very different mean profiles obtainable from the data by arbitrary selection and weighting procedures; although it can be made to match the specified multi-box model calculations for deuterium, there is a wide discrepancy between the actual and model mean mixing ratio profiles which cannot be ignored. Taken together, the mixing ratio and deuterium profiles indicate that mean vapour of the middle troposphere has been subjected to condensation at greater heights and lower temperatures than those considered in the model calculations. When this is taken into account, the data actually fit much better to the simpler condensation models. But the vapour samples represent meteorological situations too remote in time from primary precipitation events to permit definite conclusions on cloud system mechanisms.

1. Introduction

Rozanski and Sonntag (1982) have applied the present author's data (Taylor, 1968, 1972) to support a multi-box cloud model of the vertical distribution of deuterium in tropospheric water vapour; they claim that the negative vertical gradient of deuterium concentration in the lowest 5 km is much steeper than the predictions of Rayleigh-type condensation models, and compare an average deuterium profile with a multi-box model which incorporates isotopic exchange between gaseous and liquid water in the cloud system. The claim regarding the steepness of the vertical gradient directly contradicts the evidence of the individual profiles. The purpose of the present note is to draw attention to this discrepancy and to present an alternative assessment of the applicability of the data to such condensation models.

2. Data used to assess multi-box model

Twenty vertical profiles of deuterium concentration were obtained over a full year (June 1967–June 1968) near Heidelberg; 10 sampling heights were spaced equally between 0.5 km and 5.0 km. The measured quantities for each vapour sample included δ-value relative to SMOW (difference per mille between D/H ratio of sample and that of Standard Mean Ocean Water), mixing ratio and temperature; these were supplemented by information on the prevailing meteorological situation and air masses. Rozanski and Sonntag

have now calculated mean δ-values for each sampling height, using all the results; these comprise 12 complete and 6 incomplete profiles from mainly anticyclonic situations with a marked temperature inversion at varying heights between 0.5 km and 4.0 km, and from 2 complete profiles taken within frontal situations.

3. Discussion

The models used to predict isotopic concentration gradients in cloud vapour compute changes of mixing ratio (w = g kg^{-1} dry air) and subsequently deuterium concentration (more readily calculated using $R = D/H$ rather than δ-values) as the system undergoes chosen meteorological condensation processes from specified initial conditions. Since R derives from w, the two parameters are closely related. It is therefore essential to give proper consideration to the profiles of both w and R when attempting to link them to any such cloud models. Moreover, if mean deuterium concentration profiles are to be evaluated for this purpose, the individual δ-values would have to be weighted by mixing ratio. The mean profile presented by Rozanski and Sonntag is unweighted, and therefore does not meet this criterion.

The effect of absence of weighting is shown in Fig. 1, which is also intended to demonstrate the extent of mean profile variation resulting from weighting and pre-selection of results. Profile A of Fig. 1 shows the mean unweighted profile determined by Rozanski and Sonntag. Weighting by mixing ratio leads to profile B, with mean δ-values up to 55‰ less negative than the unweighted values. Restriction of the data to only complete profiles produces weighted profile C, which shifts the top of the profile by about +10‰ relative to profile B. Profile D is the mean profile for the two frontal situations; here the vertical gradient is substantially weakened by the extension of turbulence to greater heights than usual. Exclusion of the two frontal situations from profile C leads to profile E, which tends again towards the unweighted mean profile A. Such large changes of mean profile resulting from arbitrary weighting and data selection indicate that mean δ-value profiles calculated from this particular data set could hardly be used with any confidence to test cloud

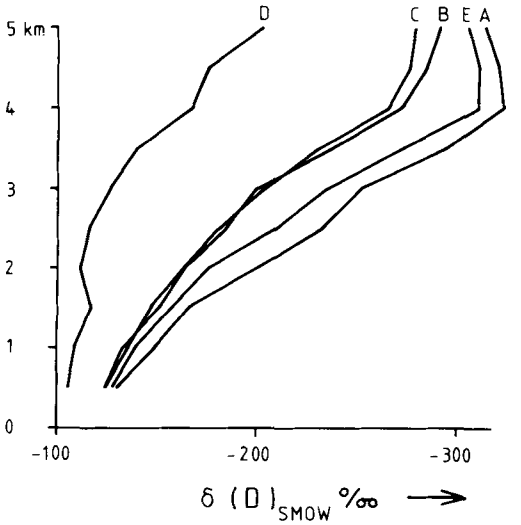

Fig. 1. Variability of mean deuterium/altitude profiles estimated from data of Taylor (1972). A: all results unweighted. B: all results weighted by mixing ratio. C: complete profiles only, weighted. D: two frontal region profiles, weighted. E: complete profiles, weighted, excluding two frontal profiles.

system models. Moreover, the model predictions can also vary widely, depending on initial parameter selection and postulated in-cloud mechanisms.

The question arises of whether even a reliable mean δ-value profile could be related at all to the mechanisms in precipitating clouds. It was already evident from the individual Heidelberg profiles (Taylor, 1972) that the deuterium gradient between the ground and the lowest prevailing temperature inversion is substantially weakened by turbulence and convection, and by admixture of newly evaporated vapour from the ground or ocean surface: this quickly removes evidence of the situation during and immediately following precipitation events. Above the temperature inversion, essentially all vapour has taken part in condensation processes, and vertical gradients of deuterium concentration are much steeper: the δ-values and their relationship to the mixing ratio w are both closely related to air mass origin and condensation history, but not to specific condensation mechanisms. Individual profiles demonstrate the strong interdependence of R and w in the form:

$$\Delta(\log R) = (\alpha - 1)\,\Delta(\log w),$$

where α can be considered as an effective deuterium separation factor characterizing the outcome of all the condensation and mixing processes which influence the deuterium distribution within an air mass up to the time of sampling. Linearity of the above relationship (constant α) is found in individual air masses of the middle troposphere over wide ranges of temperature and mixing ratio; the α-values are usually of magnitude close to the equilibrium separation factors pertaining at the adiabatic condensation temperature of the levels with highest w. The variation of R with w in individual profiles is thus, in general, less steep than that predicted by the simple Rayleigh condensation model (i.e. a model in which isotopic equilibrium exists between vapour and condensate, the latter being immediately removed without subsequent isotopic exchange with residual vapour in the system). Fig. 2 shows the (log R, log w) relationship for the mean profile E of Fig. 1, together with the data from two individual profiles; in both individual cases, the linearity is better than that of the mean profile. The α-values encountered in individual profiles ranged from 1.087 (oceanic air masses of tropical origin) to 1.200 (polar air masses).

Despite this evidence, Rozanski and Sonntag have accepted a profile (profile A of Fig. 1) which they consider to be far steeper than those predicted by Rayleigh and two-phase model calculations. The reason for the discrepancy appears to lie in their failure to recognize that the profiles of mixing ratio must also be considered when comparing actual and model profiles. Their calculations follow the usual procedure by varying temperature in small steps (2 °C) and calculating the resulting changes of w and R, using empirical deuterium separation factors: the starting conditions for comparison with the Heidelberg data assume an initial condensation height of 900 m at 4 °C, any liquid water in excess of 0.5 g m^{-3} being removed as precipitation. Fig. 3 compares the w-profile recalculated by the author for the Rozanski–Sonntag model with the w-profile actually pertaining to profile E of Fig. 1. Clearly some explanation has to be found for the difference between the actual and calculated mixing ratio profiles before moving to compare the deuterium profiles. There are other inconsistencies too. There is no correspondence between the temperature/height scale recalculated by the author, and that appended by Rozanski and Sonntag to their

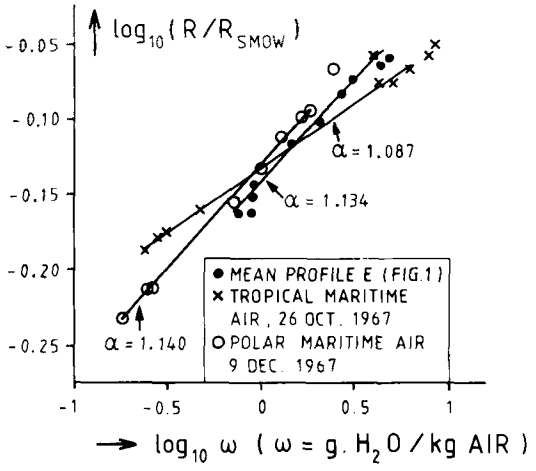

Fig. 2. Vertical variation of deuteriun concentration expressed as a linear (log w, log R) relationship. Shown are the mean profile E of Fig. 1, and individual profiles from 26 October and 9 December 1967.

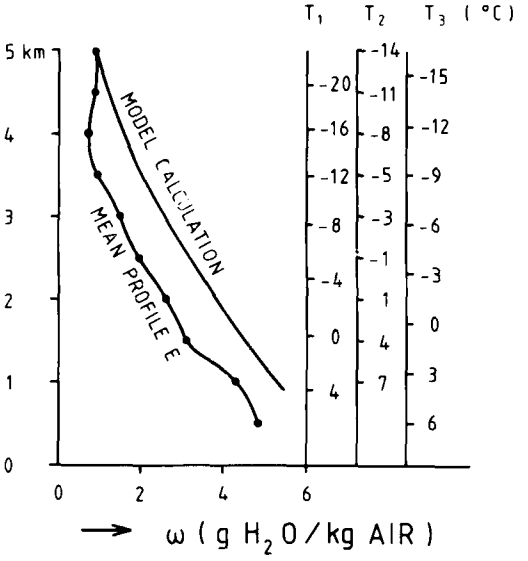

Fig. 3. Comparison between actual mean profile of mixing ratio for vapour sampling (profile E of Fig. 1) and that predicted by the Rozanski–Sonntag multi-box model calculation. Actual vapour has undergone condensation at greater heights (by 1–2 km on average). T_1 = moist-adiabatic temperature scale calculated for the multi-box model (non-linear with height). T_2 = mean temperature of vapour samples in profile E. T_3 = temperature scale appended to model calculation by Rozanski and Sonntag (linear with height).

calculated profile; both differ substantially from the temperatures of the average profile E. Rozanski and Sonntag have assumed an initial condensation temperature of 4 °C, compared to the 8 °C actual average temperature at that level: their temperature then drops by the average atmospheric lapse rate of 0.5 °C/100 m, rather than the steeper moist-adiabatic lapse rate imposed by their model. A further problem concerns the subsequent status of the 0.5 g m^{-3} liquid cloud water remaining at the cut-off point of the Rozanski–Sonntag calculations, which may substantially affect the deuterium concentration of the vapour if it again turns to vapour after the precipitation event.

The major problem of the foregoing is the wide discrepancy between calculated and measured mean mixing ratio profiles (Fig. 3). The difference arises because of the sinking motion of the middle and upper tropospheric air within the mainly anticyclonic sampling situations. The collected vapour has usually participated in condensation processes at greater heights, and therefore lower temperatures, than those pertaining to the sampling, and those applied by Rozanski and Sonntag in their model calculations. When this is taken into account, it becomes possible to match the mean mixing ratio profile more closely to moist adiabatic condensation models. Proceeding then to model estimates of deuterium profiles, Rayleigh-type and simple two-phase models can be fitted quite realistically to the measurements, whereas the multi-box model with appreciable isotope exchange between vapour and falling precipitation now predicts much greater deuterium depletions than the measurements indicate.

Nevertheless, it would be incorrect to say that this constitutes support for the simpler models. In the actual atmospheric situation, numerous processes intervene to redistribute water vapour and heavy isotopes in the intervals of days, or even weeks, between primary precipitation events and the time of sampling of the middle tropospheric vapour. These may include advection, sinking, turbulence, convection, and cirrus or other types of cloud formation with fall and re-evaporation of ice crystals at lower levels. Therefore, it is inappropriate to use deuterium profiles from such sampling situations to assess condensation models. For these reasons, no such attempt was made in the original paper (Taylor, 1972), which concentrated instead on the features of the individual profiles, viz. the smoothing of deuterium concentration gradients by turbulence and convection below the subsidence inversion, and the characteristic linear (log w, log R) relationships encountered in air masses of varying origin in the middle troposphere.

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