Does the isotopic signal in cloud droplets only depend on condensation fractionation?

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Introduction

Stable water isotopes (D and ^{18}O) are used to trace the hydrological cycle:

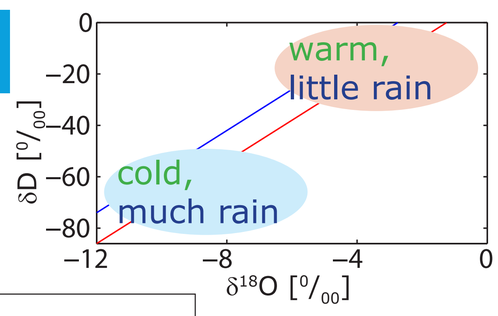
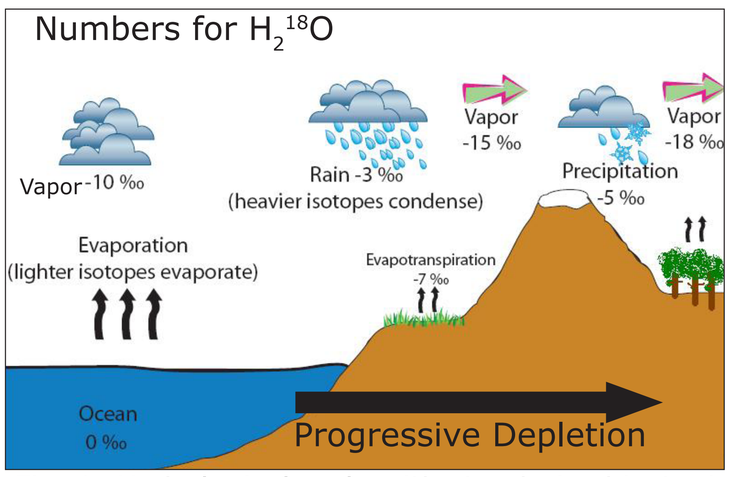


Fig. 2: Illustration of the isotopic effects in the δD-δO diagramm



The airmass is more depleted:
a) the colder the airmass is (Temperature effect [1])
b) the more the airmass rained along the trajectory (Rain-out effect [2])

Fig. 1: Hydrological cycle with standarized isotopic ratios for $H_2{}^{18}O$
Adapted from: http://serc.carleton.edu/microbelife/research_methods/environ_sampling/stableisotopes.html

Sampling procedure

Cloud water samples were collected using a 3 stage fog collector during the Hill Cap Cloud Thuringa 2010 (14thSep. 2010 - 24thOct. 2010) campaign on the Schmuecke (50° 39' N/ 10° 46' E, 937 m a.s.l).

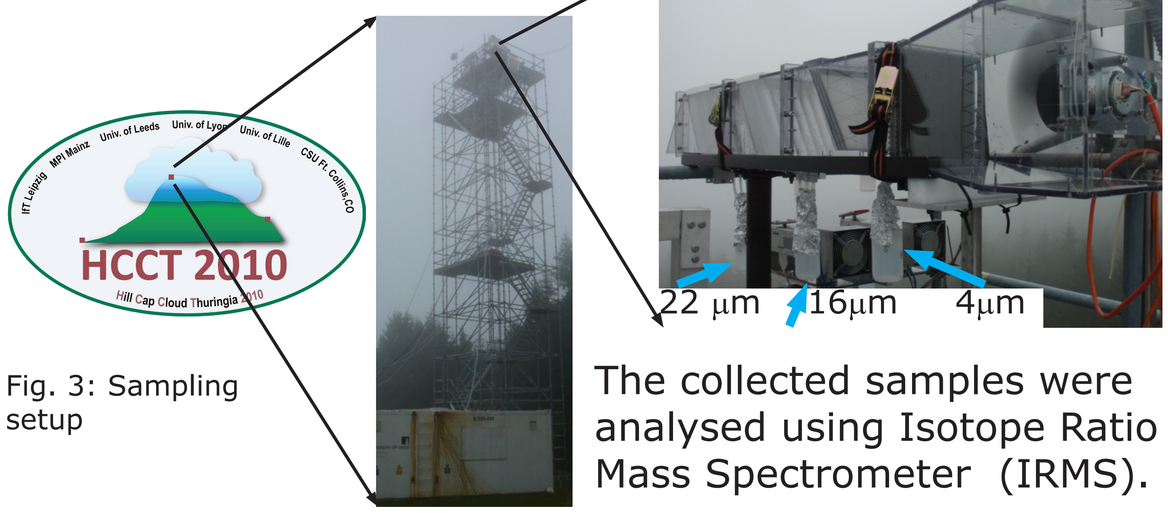


Fig. 3: Sampling setup

The collected samples were analysed using Isotope Ratio Mass Spectrometer (IRMS).

Results & Disscusion

1) Temporal evolution of the signal > differences between stages

Explanation:
Equilibration time of droplets at that size < 10s [3]

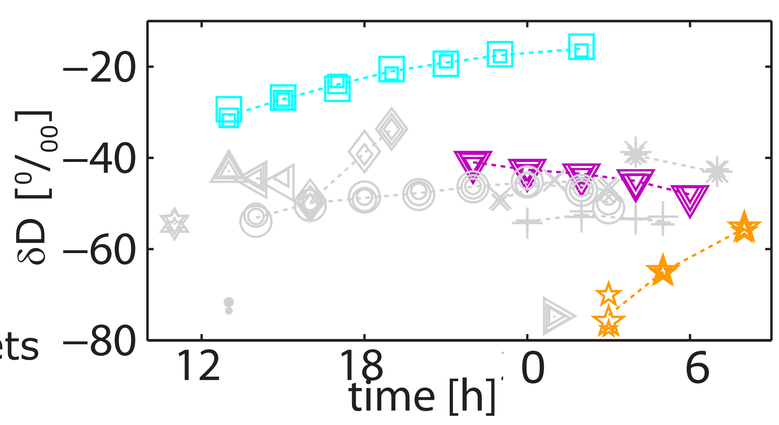


Fig. 4: Diurnal evolution of δD in the cloud droplets (HDO) for all measured events (symbols) and all stages (smallest symbol 4 µm etc.). The colored events are discussed further.

2) Classification of the events using a thermodynamic box model:

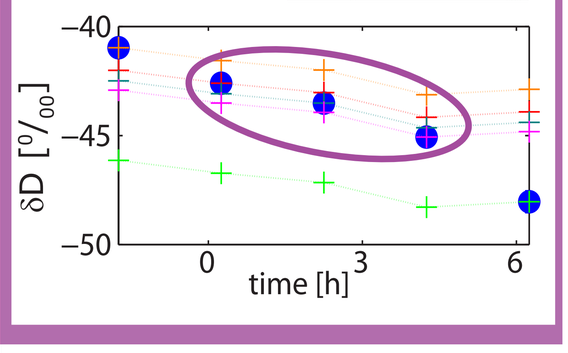
The temporal evolution of the istotopic signal in the droplets is...

the modeled points for one starting value fit to several measurement points (= starting values are very similar or even equal)

YES → ...basically condensationally driven

Fig. 6: For 3 different starting values the curves aline well with the measured data (blue points)

Starting values:
δD (T_d) = -124.7 ‰
δD (T_d) = -125.6 ‰
δD (T_d) = -126.0 ‰
δD (T_d) = -126.4 ‰
δD (T_d) = -129.4 ‰



NO → *expected rainout or temperature patern in the isotopic data*

YES → ...basically explained by airmass history: rainout and temperature

Starting values:
δD (T_d) = -111 ‰
δD (T_d) = -106 ‰
δD (T_d) = -103 ‰
δD (T_d) = -116 ‰
δD (T_d) = -108 ‰
δD (T_d) = -104 ‰
δD (T_d) = -102 ‰

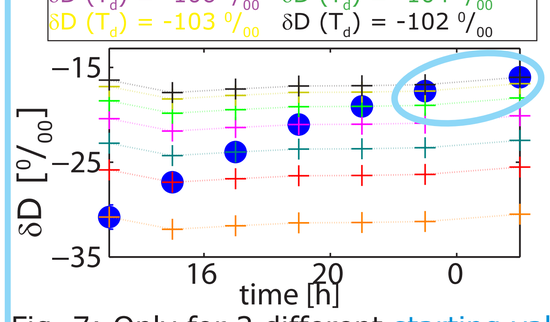


Fig. 7: Only for 2 different starting values the curves overlap with the measured data (blue points), so fractionation due to condensation only plays a minor role.

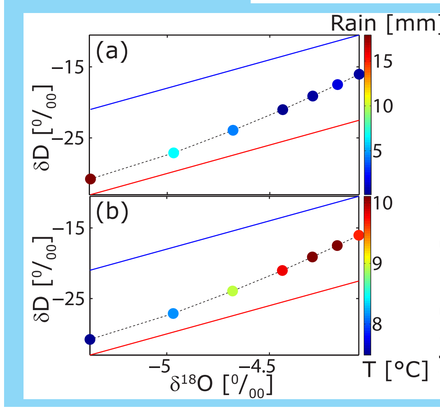


Fig. 9: The measured isotopic values **follow** the expected (a) rainout effect (summed rain along a 96h calculated backward trajectory) and (b) temperature effect.

NO → ...neither explained by the airmass history nor by condensation fractionation

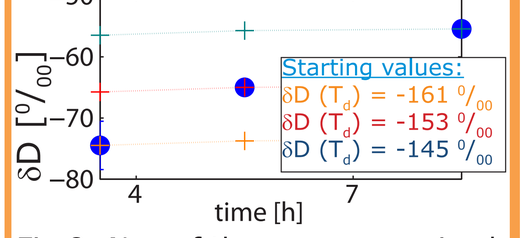


Fig 8: Non of the curves received for purely condensational fractionation describes the event properly

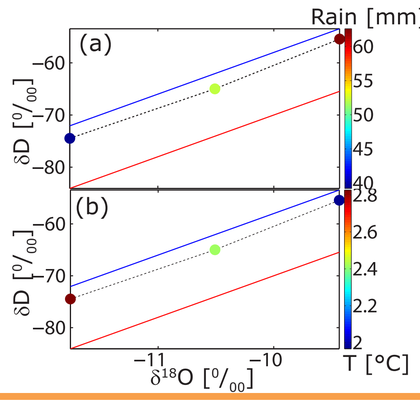
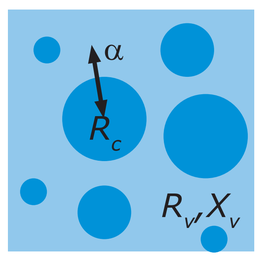


Fig. 10: The measured isotopic values **do not follow** the expected (a) rainout effect (summed rain along a 96h calculated backward trajectory) and (b) temperature effect.

Thermodynamic box model

1) Model description [4]:

Fig 5: box model scheme:



R_c: Isotopic ratio of the condensed phase
R_v: Isotopic ratio of the vapor phase
X_v: mole fraction of the vapor phase
$\alpha(T)$: fractionation coefficient

$$\frac{dR_v}{R_v} = \frac{(X_v - 1)\,d\alpha + (\alpha - 1)\,dX_v}{X_v + \alpha - \alpha X_v}$$

2) Modelling principle:

- For one measurement point i of a event, we initiate the model with the measured data: $R_{ci}(T_i, X_{vi}) \rightarrow R_{vi}(T_i, X_{vi})$
- We then use the model to calculate R_v at the dewpoint $R_{vi}(T_{di}, X_v=1)$ = starting value
- Now we use this starting value to calculate $R_v(T, X_v)$ and $R_c(T, X_v)$ for the other measurement points (this corresponds to one line in Fig. 6, 7 & 8) and compare it to the measured data (blue points in Fig. 6, 7 & 8).
- We repeat this procedure for all points of the event (various lines).

Conclusion

No, the isotopic signal in cloud droplets **does not only depend on condensation fractionation**, but also on **airmass history**!

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[3] Lee J & Fung I (2007), "Amount effect" of water isotopes and quantitative analysis of post-condensation processes, *Hydrol. Process.* (2007), DOI: 10.1002/hyp.6637

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