Assessing raindrop evolution over northern Western Ghat from stable isotope signature of rain and vapour

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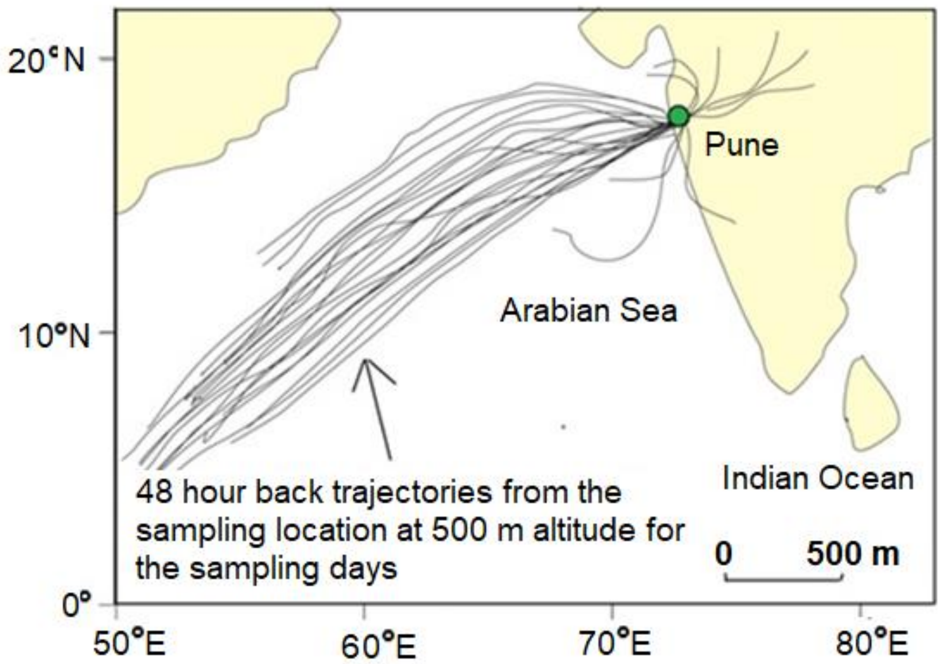

Figure S1. 48-hour air mass back trajectories calculated using the HYSPLIT model for Pune at 500 m for the sampling period 2019. Each black line represents the trajectory for one sampling day.

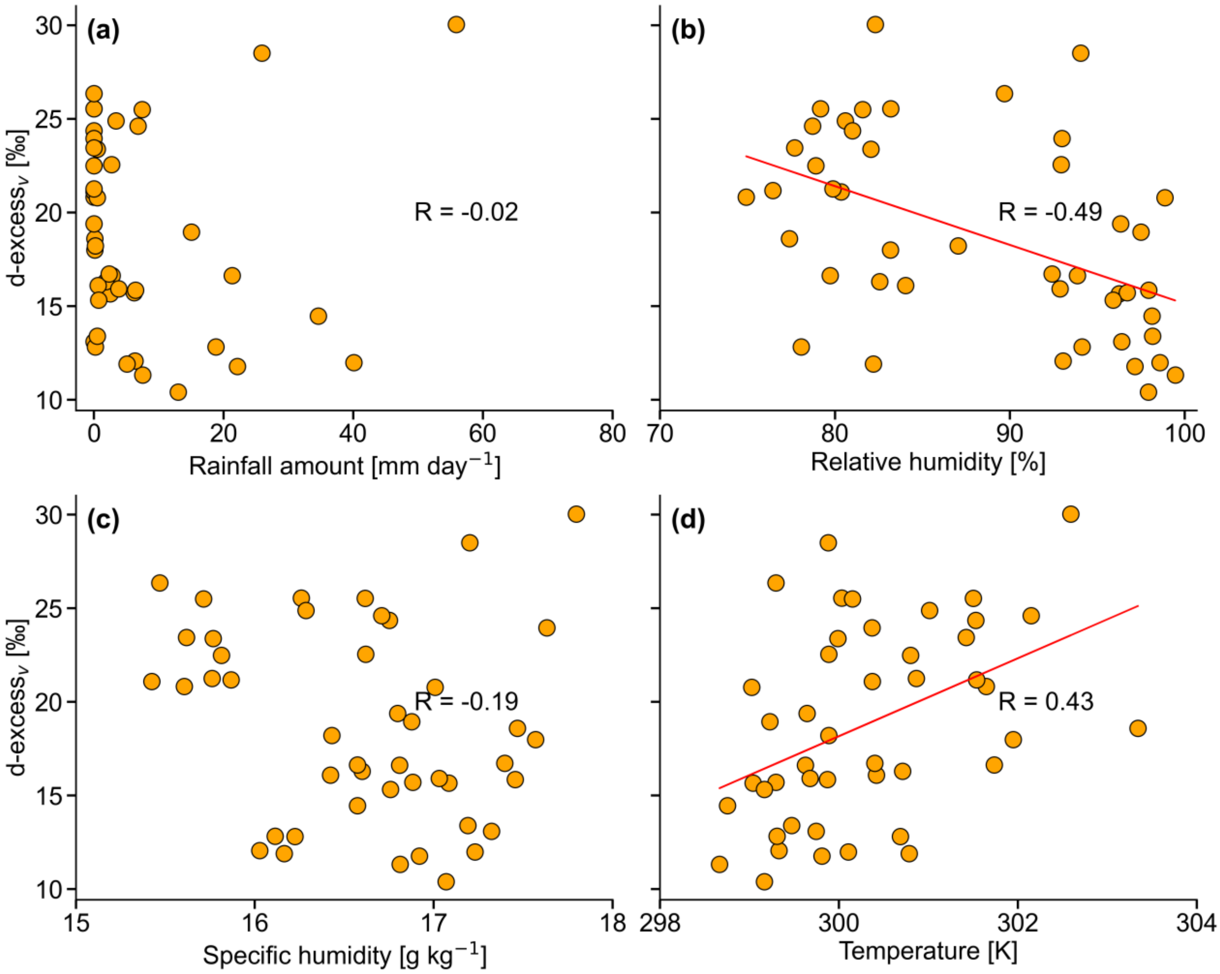

Figure S2. Water vapour d-excess values (d-excess$_v$) measured at Pune plotted against the corresponding **(a)** rainfall amount, **(b)** relative humidity, **(c)** specific humidity, and **(d)** air temperature on the corresponding days.

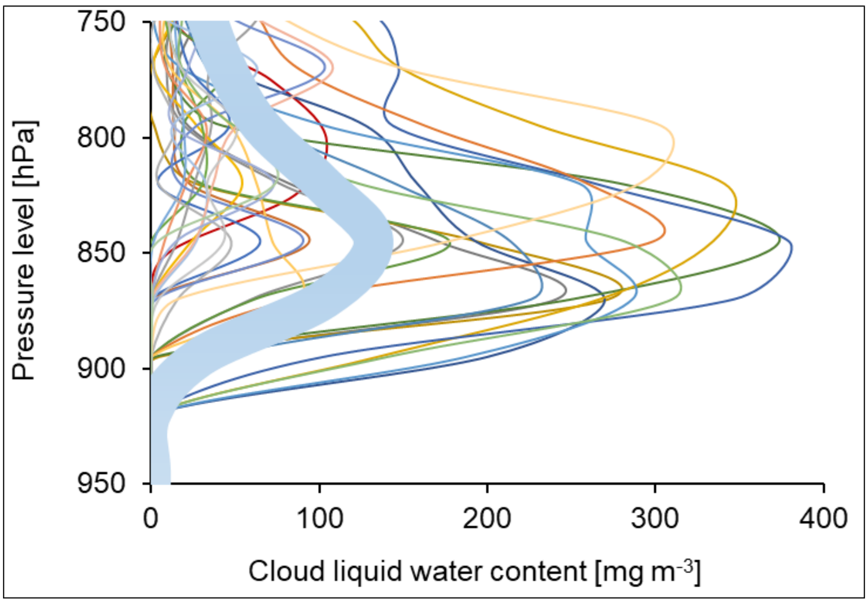

Figure S3. Cloud liquid water content (CLWC; mg m^{-3}) variation at different pressure levels over Pune during the study period. Each coloured line indicates the daily average CLWC obtained from the ERA5 dataset. The thick grey line shows the mean CLWC for the study period.

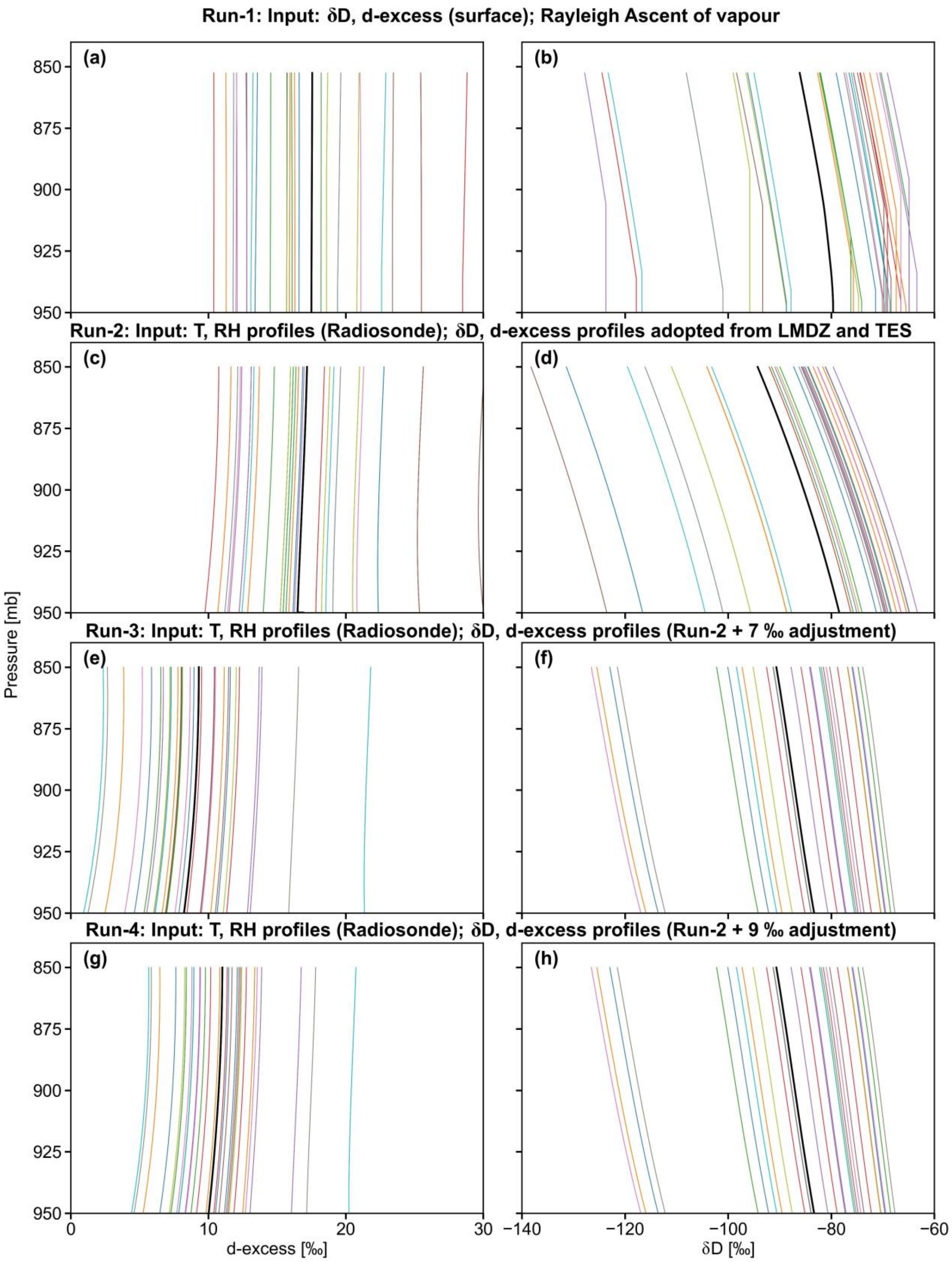

Figure S4. Input d-excess (**a, c, e, and g**) and δD (**b, d, f, and h**) profiles of atmospheric vapour for various runs used in the below-cloud interaction model (BCIM). The bold black line indicates the average profiles. Details of the runs are discussed in Section 4.3; the run numbers are given above (in bold red letters).

Supplementary Information-SI 1: Uncertainty estimate for δD (rain) and d-excess (rain) model values

Using the model output values for the days when rain and vapour were analysed simultaneously, we made a multi-parameter fit for the δD values of rain. The obtained regression equation for δD (rain) as function of the four variables, δDvapor, RH, Temp, and Diameter, is

$$\delta D_r = 143.16+1.068*\delta Dv-0.433*RH-0.782*T-3.269*diameter \quad (1)$$

Using a similar method for the model-predicted d-excess (rain) values, we obtained the following multi-parameter fit equation:

$$d\text{-}excess_r = -10.5557+0.60164*d\text{-}excess_v+0.169599*RH-0.31632*T+2.2921*dia \quad (2)$$

Plots of the predicted values against the observed values show the goodness of these equations for prediction (Fig. S5 and Fig. S6).

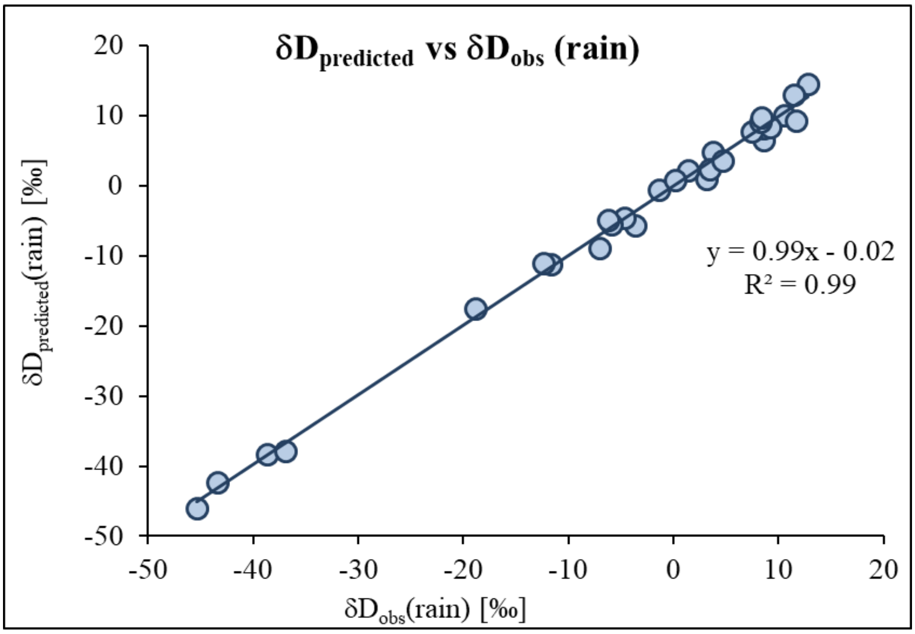

Figure S5. δD predicted by BCIM Run-4 plotted against the δD observed values for rain.

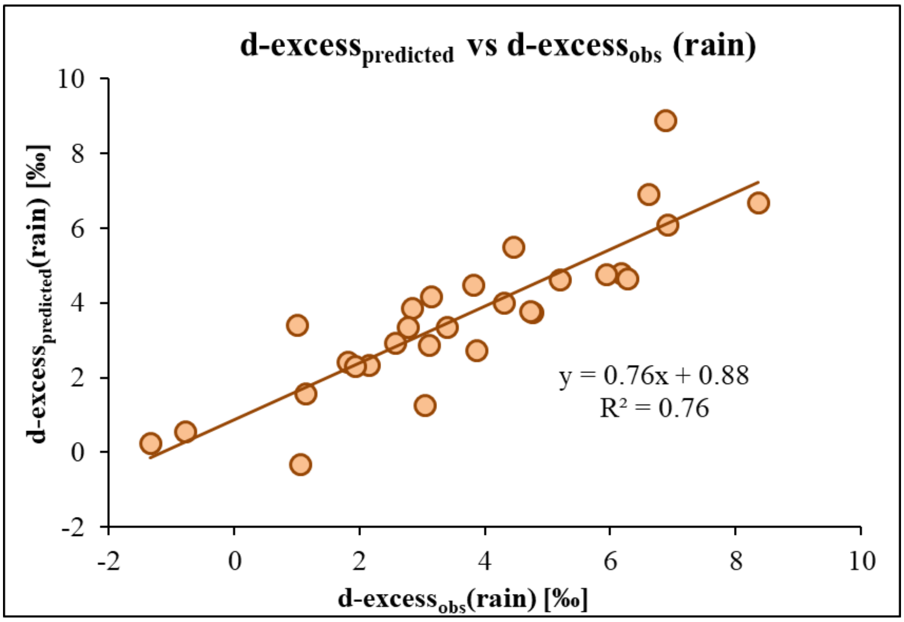

Figure S6. d-excess rain predicted from run-4 of the BCIM simulation plotted against the observed d-excess rain values.

Using the above two linear equations, we can estimate the uncertainty in the model's predicted values using the quadrature method. The uncertainty values for δD_{rain}=3.5 ‰ and d-excess$_{rain}$=2 ‰. The assumptions are the input uncertainties of δD_{vap}=4 ‰, d-excess$_{vap}$=2 ‰, RH=5 %, Temperature =0.5° C, and Diameter =0.5 mm.

It is to be noted that we do not have any concrete measures or guidelines for assuming the input vapour isotope uncertainties. Therefore, to get an order-of-magnitude estimate, we used the input data variances in δD_{vap} and d-excess$_{vap}$ (excluding some outliers) to obtain the errors associated with the model inputs. Since the variance is an extreme measure, we divided the variance by 2 and used that value as the 1-sigma uncertainty.

Supplementary Information-SI 2: Sensitivity Analysis

Sensitivity of the isotopic composition of rain to different input parameters for BCIM is discussed in this section. A change in the input parameters (from the values adopted for the reference case) can change the position of the data points in the Δδ–Δd diagram. We assess the change in the slope value Δδ/Δd due to a change in one input parameter at a time. The results are shown in Fig. S7. The input parameters include relative humidity (RH), temperature, the background isotope profiles in terms of δD and d-excess, and the drop diameter. Each simulation was done for three drop diameters (0.6 mm, 1 mm, 1.3 mm), and result points for the same input parameter are connected with a line (dotted/solid). RH low and RH high correspond to values with a 10 % decrease and a 10 % increase in the relative humidity, respectively. T low and T high denote simulations with 5 % decrease and 5% increase in average temperature profiles. The ranges of variation of these two parameters (air temperature and relative humidity) are decided by the uncertainty of the same for the radiosonde measurements. Simulations with altered background profiles of stable water isotopes are denoted as δD low and δD high (δD profile changed by ±20 %) and d-excess low and d-excess high (d-excess profile changed by ±10 %). A significant difference was observed in the simulation results when RH was altered.

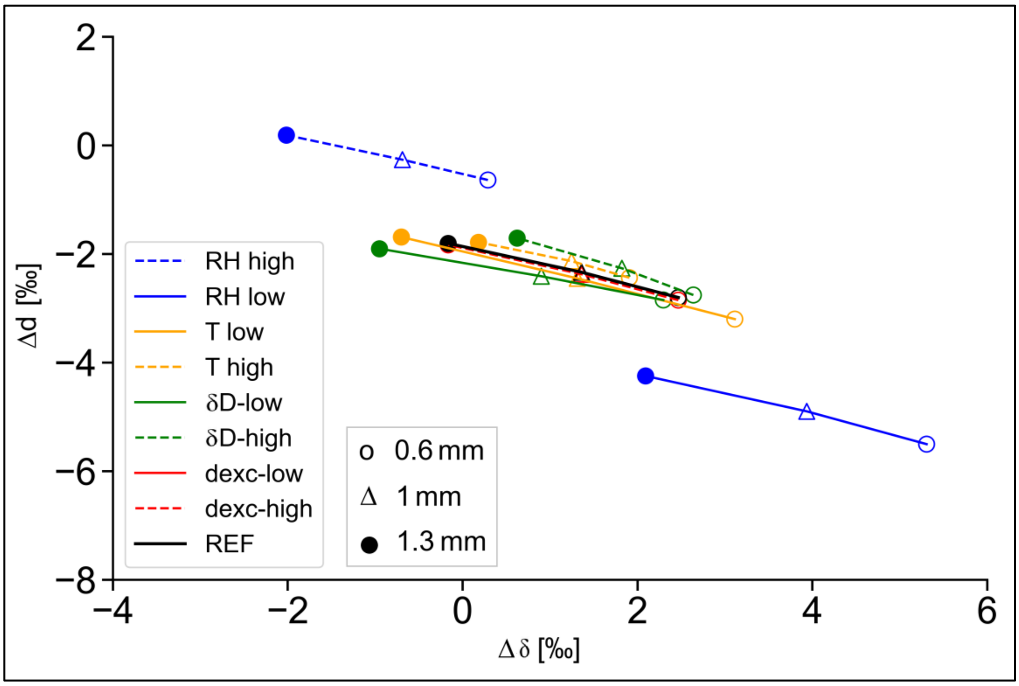

Figure S7. $\Delta\delta$-Δd diagram summarising the results of sensitivity experiments using the BCIM. The black line shows the result of the reference setup, and the coloured lines show simulations with various input parameters. Three points denote three sizes: Solid circle (●) 1.3 mm, triangle (Δ) 1 mm, and open circle (O) 0.6 mm.

It is important to note that all data points for small diameters (and for all input parameters except RH) converge in the $\Delta\delta$-Δd diagram for different simulations. Because below-cloud processes have a larger influence for small diameters, which overwrite initial differences. High evaporation in case of RHlow shifts low diameter samples to high $\Delta\delta$ and low Δd, whereas less evaporation in RHhigh leads to near complete equilibration with the ambient vapour (the points are closer to zero).