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

Summary:
The investigators aim to assess the degree of evaporation of rain between cloud base and ground level by linking ground based water isotope measurements with a below cloud isotopic model.

Ground based samples of rainwater and ambient vapor, located at the Indian Institute of Tropical Meteorology in Pune, are collected throughout the 2019 summer monsoon season. It appears that rainwater was collected on a 24 hr basis. Vapor was collected by pumping air through a chilled vessel to condense moisture out of the air, with a time resolution of 3-4 hours. Vapor and rainwater were sometimes collected on the same day, and are collocated.

The water isotopic trend throughout the season is analyzed in the context of large scale meteorology. Using outgoing longwave radiation anomaly (from satellite), a link between mesoscale convection and depletion of rainwater isotopes is demonstrated. Analysis of observed rainwater and vapor isotopes in $\Delta\delta$-Δd space indicates the prevalence of below-cloud evaporation taking place to various extents throughout the season. Additionally, there are signatures in the vapor isotope data which lead the authors to believe that depleted vapor is being downdrafted potentially as a result of raindrop evaporation.

The authors then attempt to use the BCIM model to compare modeled raindrop isotopic evolution with the ground based measurements. Several estimates must be provided as inputs into the model: droplet formation height, droplet size, atmospheric temperature, humidity, and vapor isotopic profiles. The authors describe three approaches:

1. Using ground based temperature, relative humidity, and vapor isotopes, assume a moist adiabatic atmospheric lapse rate. The droplet begins at a fixed height of 850 mb. Droplet sizes are calculated from daily rainfall rates.
2. Radiosondes are used for thermodynamic profiles. Vapor dD is obtained from 2005-2009 satellite retrieval, while dO18 is obtained using d-excess profiles from a GCM and extrapolating from the observed vapor d-excess. Droplet sizes from daily rainfall rates. 850 mb droplet formation height.
3. Vapor isotopic profiles from method 2 are tuned to produce a d-excess at ground-level value in agreement with the observations.

The authors use the final tuned model input to conclude that on average 23% of rainfall during the season evaporated during descent.

Overall assessment:

This study presents a new set of collocated rain and vapor isotope observations, a measurement approach which holds great potential for addressing the topic of interest here. The simultaneous observations of vapour and precipitation (as opposed to only one or the other) is particularly advantageous. However, with the exploration of the BCIM that is presented here, there is too much going on in this study, making the results muddy. This reviewer believes that the impact of and insights from the in-situ measurements is detracted by the troubled attempt to reproduce the observations with the BCIM.

This reviewer does not find the methods used to initialize the BCIM model to be convincing, and therefore holds little confidence in the quantitative values for evaporated fraction obtained from the model. While acknowledging and commending the work that the authors have devoted to the model analysis, this reviewer feels that in its current state it can be omitted entirely. It is therefore not ideal that the focus of the abstract is based on the BCIM results. A more impactful study might focus thoroughly on the observational dataset, strengthening the analysis in that area, and removing or heavily revising the modeling component.

Observational measurements are less common and highly valuable, and thus the sampling of rain and vapor isotopes is compelling. The authors have produced an observational dataset of high value, and the measurements are likely to provide a valuable and novel contribution to this research field. The authors can strengthen their findings from the observational data by further substantiating points #1-4 and #6 of the conclusions. More care should be put into describing the data collection methods and ascertaining firm conclusions on the observed trends within the context of other literature.

Research Methodology:

Below are specific comments on the research methodology, first for the experimental component, and then for the modeling.

<u>Observational measurements:</u>

The measurements in this study that are compelling:

- Rainwater isotope fraction and d-excess: I'm convinced that these measurements are highly reliable, although the authors should clarify if the rainwater is collected every 24 hours, and how this is done (e.g. what time of day is the sample collected?). Also, the link to the IAEA document is not easily accessible (I was not able to access it on a number of different web browsers and devices). So, it would be good to briefly summarize the collection method in case the reader can't access the link.
- Ground based rainfall rate, temperature, humidity from IMD: Although not precisely collocated with the rainwater isotope samples, the daily averages are temporally consistent with the rainwater collection and are likely valid.
- Radiosondes: Although not precisely collocated with the rainwater samples, and the mismatch in temporal resolution is severe (24 hr integration for isotopes, vs averaging of two point measurements for sondes), atmospheric profiles in the same region and date-range as the rainwater isotopes provide extremely valuable atmospheric context. I think the authors could be more careful with dealing with the temporal discrepancy. For example, on days with precipitation, it would be wise to verify that radiosondes were launched during the time of precipitation. As an extreme

example, if the day's precipitation accumulated between 04-09Z, but the sondes are launched at 00Z and 12Z, the atmospheric soundings for that day might be very misrepresentative, and may not even contain clouds. In general, I think these radiosondes could be utilized to a greater extent in the observational analysis.

Vapor water isotope collection:

- It's possible that these vapor isotope measurements are highly reliable, but more details need to be provided by the authors to allow the reader to make this assessment. It seems that a detailed description of the vapor collection apparatus has not been published prior, and no literature references were provided for the design. As such, including a picture or a diagram of the apparatus would be helpful. More importantly, were there any calibrations or validation of the apparatus performed prior to collecting the measurements?
- A question I have is about the level of certainty that the flow rate, temperature, and surface area within the chilled flask was sufficient to condense out all of the water vapor from the air sample as it's pumped through? If the water vapor is only partially removed from the sample (with the remainder flowing out), then significant kinetic fractionation could occur during the process of condensing the water vapor inside the dewar. If this were to be the case, the water condensed in the dewar would adopt an enriched bias, and an elevated d-excess that could also have a dependence on humidity. This may possibly explain some of the d-excess trends in the water vapor that were unaccounted for. Can the authors comment on this?
- More details on the timing of the vapor sample collection is also needed - Were the samples collected once per day? Or during intensive observation periods?

The authors explain that the local advective vapor fluxes in this region are dominant, and any vapor from evaporating rain is likely negligible in the vapor isotopic signature. Could the authors assume a well-mixed condition below cloud base (assume that vapor isotope ratio is constant below cloud base)? In fact, this is the assumption that is made in the Rayleigh model: See Fig 4C of Graf et al 2019, where ambient vapor isotopes (black line) are constant up until the cloud base. A linear temperature profile from the radiosondes would also be indicative that a well-mixed boundary layer assumption is valid. The authors mention that the rainwater is not in equilibrium with the vapor at ground level. If the vapor isotope is constant below cloud base, and a temperature profile is known from radiosondes, the authors could identify (based on temperature) the height where the rainwater would be in equilibrium with the vapor. It would be informative to compare this to the lifting condensation level (aka cloud base height), and potentially some qualitative insights on isotopic exchange could be derived.

The interpretation of the slope in Line 402-403 and point #6 of the Conclusions section could be strengthened. Fig 7 of Graf et al 2019 shows different conditions that would lead to a steeper slope in the $\Delta\delta$-Δd space. The authors have discussed the effects of RH and temperature on the expected slope: What about the ambient vapor isotope values? For example, if the ambient vapor d-excess in Pune is larger than the ambient vapor d-excess in Switzerland, then one would indeed expect the Pune rainfall data to fall along a steeper slope than that in Switzerland. From the observed data presented, it appears that Pune vapor d-excess values range from 10-30 while the data in Graf et al 2019 is in the 5-20 range, so this could be a potential source of the steeper slope. Likewise, higher vapor δD values would produce a steeper slope, which again the Pune values appear to be larger than those observed in Switzerland. For completeness, all the different factors that might contribute to a steeper slope should be considered and discussed to strengthen this analysis.

Line 357: Is the ambient temperature used for the $\Delta\delta$-Δd plots obtained from the IMD observatory?

Line 389: Can the authors comment on if the <5mm rain rate of the data in the lower right quadrant is relatively lower than those in the lower left quadrant?

Line 390: The text says nine samples, but the scatter plot shows 11 points in the lower left quadrant.

Line 391-392: The statement on size of raindrops and precip intensity - Is this a finding from your own observations? If so, please demonstrate how this was assessed.

<u>BCIM model:</u>

In general the authors should clarify if the radiosondes are being used for the thermodynamic profile in all of these cases (other than the Rayleigh case), or if the GCM, or satellite profiles are being used. Perhaps the authors could provide a table for each run, specifying the BCIM inputs and where they came from: droplet size, introduction height, Temperature and humidity profile, Deuterium, and O18. Or just be very clear in the description.

Showing plots of the atmospheric temperature and RH profiles that are being input into the model would be an enormous help to the reader for understanding/assessing the atmospheric structure being modeled. In addition to the isotope profiles shown in Figure S4.

If I understood correctly, run-2 uses radiosonde for the thermodynamic profile, dD comes from satellite data from 2005-2009, and d18 comes from the d-excess of the GCM extrapolated to the observed ground-based vapor measurement. There are major concerns with these inputs coming from different data products that all have different spatial/temporal scales and measurement principles.

For example, if the thermodynamic profile and the dH profile were both taken from satellite retrievals, then that dataset would at least be internally consistent. But, taking the different atmospheric properties from different sources, there is little physical basis to believe that they would be internally related to each other.

In my view, there are large sources of error being introduced with each model input assumption, starting with the droplet generation height, continuing with the droplet sizes, and compounding with each additional data product incorporated. I feel these uncertainties are too severe to enable a useful comparison with the in-situ rainwater measurements. Likewise, the measurements are unable to offer an advantage in assessing the model performance. For these reasons, this reviewer does not see added value in presenting the BCIM in its current form alongside the in-situ rainwater measurements.

L 426-427

Clarification on the choice of droplet formation height: I would agree that raindrops are most likely to be generated in regions where CLWC is maximum, but Section 2.4 indicates that the BCIM releases the droplets from cloud base, and it seems implied that this height is what is input to the model. Is the droplet released from the cloud base, or from a height inside the cloud? Showing the input profiles for temperature and RH, as mentioned above would clarify this for the reader.

If the droplet is released from cloud base:

The moist adiabatic lapse rate in the Rayleigh method should provide a cloud base height based on the height where the moist parcel reaches saturation, would it not? Could that be used in place of this fixed 850 mb level?

If radiosondes from the study period are provided, why not use the lifting condensation level (LCL) from the soundings to define the cloud base height?

If the droplet is released from a height inside the cloud:

The authors acknowledge that assuming the same droplet formation height for each day is going to be erroneous. How does this fixed 850 mb height compare with the cloud base height from the moist adiabatic assumption and the cloud base height from the radiosondes on each day? Can the authors confirm that it is above the cloud base in all cases? The ERA5 data in Figure S3 indicates that the cloud base is not often above 850 mb, which is encouraging, but it does vary. Perhaps the authors could instead calculate the average height above cloud base where CLWC peaks, allowing the droplet formation height to be adjusted based on the cloud base height on each day.

L 431-437

A citation is needed for the Marshall-Palmer relationship. Looking at the Marshall & Palmer 1948 paper, it may not be an ideal choice for the droplet diameter estimate. Have the authors considered using droplet size distributions from Murali Krishna et. al. 2021, which is included in the references? It appears that Murali Krishna et. al. 2021 has DSD's for a range of rain rates in the Western Ghats which would seem more appropriate than those collected in Ottawa in the Marshall & Palmer paper, and of higher quality using modern measurement techniques.

L 443-448

Can the authors please describe or reference the equations used to calculate these profiles.

Line 470-471

The authors should provide a brief description and references for the isotope enabled LMDZ GCM model.

Referencing a personal communication to obtain the isotopic inputs to the BCIM is a bit peculiar. If this particular GCM output has not been published prior, and was provided for use and publication in this analysis, this reviewer wonders if it would be more appropriate to include the contributor of the GCM isotope profiles as a co-author.

Line 522, 556-559

I find the assertion that "the main source of error in Run 1 and Run 2 could be improper vapor isotope values", to be invalid. Considering the uncertainties around the choice of droplet introduction height, droplet size, and thermodynamic profiles from the radiosondes, there are many possible sources for the lack of agreement with the observations. It is very likely a combination of many sources. The conclusions of L 556-559 are not justified, for these same reasons.

Figure S4

The vapor δD profiles in panel (b) of Figure S4 show indications of cloud base heights where the profile transitions from being constant, to undergoing rayleigh distillation within the cloud. This further begs the question of whether or not the droplet is intended to be released at cloud base or not. In many of these profiles it appears that the droplet formation height of 850 mb is quite far above the cloud base.

This also raises questions about the isotopic profiles used in Runs 2-4 that needs to be discussed: Why are the ambient vapor isotopes below the cloud base decreasing with height (and where is the cloud base)? I don't see a physical basis to justify this given that there are strong/dominant surface moisture fluxes, and the expectation of a well-mixed boundary layer.

Figure S7
I'm not seeing where the dexc-low line is in the plot?

Overall organization:

The manuscript needs significant reorganization for readability and to meet expected standards. Reorganizing the sections could also result in a much more concise paper. The line of logic leading to each conclusion is rather circuitous (not direct). In my opinion the lack of organization makes for a laborious reading experience.

A few opportunities for re-organization are:

- Lines 263 – 275 is more appropriate for the discussion section, rather than results.
- All of Section 4.3 belongs within the methods and results section.
- Lines 667 – 676 belong in the discussion section, not in conclusions.

Line by line comments:

Check with editor: Should “micro-physics” be hyphenated?

L 78: capitalization of “Estimate”

L 81-82: I think the assessment of previous studies being “often innacurate” is too strong. Suggest for example: “However, it remains a challenge to account for all cloud microphysical processes and their associated isotopic fractionations.” The sentence could simply be removed as well.

L 97: “riming” mispelled

L 109: As the referenced study has four years of 1-min disdrometer data, I think the sentence stating “limited to scanty observations” is too strong. Furthermore in the following sentence, I think it is again too strong to claim the current study as “accurate and simpler”. I would suggest avoiding these types of qualifier words. For example, the authors could revise to: “The question arises of whether one can determine the raindrop evaporation and its variation using measurements of isotope ratios in rain and vapor”.

L 147: The link to this document does not appear to be working. Perhaps the authors can briefly summarize the design and requirements of the rain collection in case the reader is unable to access the document.

L 150: "indigenously” is not the correct word here. Did the authors mean “in-house”?

L 152: units “min-1” needs a superscript.

L 351: punctuation.

L 566: What is “point #4”?