Response to Anonymous Referee #2

In this manuscript the authors present an implementation of the heavy stable water isotope HDO in the National Taiwan University (NTU) microphysical scheme of the Weather Research and Forecasting (WRF) model. Isotopic fractionation during both liquid and ice cloud formation is computed according to kinetic mass transfer principles, which is possible because the NTU scheme does not apply the saturation adjustment technique. The model was subsequently used to study a cold front event in northern Taiwan, and disentangle different mechanisms that led to a decrease of δD in vapor and precipitation. The results show that cloud microphysical processes, the initial vertical distribution of δD, and lower boundary conditions are all similarly important for reproducing the observed evolution of δD during the cold front passage. This is a well-written and interesting manuscript that takes advantage of the kinetic mass transfer formulation in the NTU scheme to physically simulate isotopic fractionation during cloud formation, avoiding equilibrium assumptions or uncertain parameterizations of supersaturation. The study nicely demonstrates how numerical models can help interpret isotope measurements and improve our understanding of the water cycle also on short time scales. I recommend that the manuscript be published after minor revisions.

General comments

1) The bibliography could be updated with some newer references. For example there are now isotope models that no longer apply saturation adjustment during ice cloud formation, e.g., IsoSAM (Blossey et al. 2010), COSMOiso (Pfahl et al., 2012).

Reply: Agree. We have revised the introduction section and added some newer references as suggested: "In conventional AGCMs, isotope exchange between liquid or ice and gas phases is usually assumed to be in a partial or full equilibrium state [*Hoffmann et al.*, 1998, *Risi et al.*, 2010, *Nusbaumer et al.*, 2017, *Werner et al.*, 2011, *Yoshimura et al.*, 2010]. In a synoptic weather system such as a front or typhoon, thermal equilibrium fractionation may not be appropriate for describing fractionation during phase change since the clouds are usually not in vapor equilibrium [*Laskar et al.*, 2014]. Therefore, in recent years, several regional models start to consider kinetic fractionation during evaporation from open water, condensation from vapor to ice, or isotope exchange from raindrops to unsaturated air [*Hoffmann et al.*, 1998, *Yoshimura et al.*, 2010; *Blossey et al.* 2010; *Pfahl et al.*, 2012; *Dütsch et al.*, 2016]. However, the microphysics in these global or regional models are usually described with single moment schemes.". We also add more description about the concept of "saturation adjustment" in section 2.1 in the revised manuscript: "Another advantages of the NTU scheme is that it does not apply the "saturation adjustment" strategy, as done in most global and regional models. This saturation adjustment treatment assumes that water vapor and liquid (or ice) water are in thermodynamic equilibrium once water (or ice) saturation is reached in non-mixed-phase clouds (i.e., all hydrometeors are either liquid or ice). Therefore, for models applying the saturation adjustment strategy, condensation is not calculated explicitly but rather by converting all excess water vapor into condensate regardless of the cloud drop size and number concentration or the time needed for condensing out all supersaturated water. So, under the saturation adjustment assumption, kinetic effect as described in Eq. (5) cannot be solved fully and explicitly. In mixed-phase clouds (i.e., water and ice coexist), the equilibrium is maintained by assuming either water saturation or ice saturation (e.g., *Sundqvist*, 1978), or by varying linearly from water saturation to ice saturation between two specified temperature thresholds (e.g., *Tiedtke*, 1993). Then, condensation on ice can be calculated following the kinetic approach, but the condensation on cloud drops still follows the saturation adjustment in most models. If the air is subsaturated but with the presence of cloud drops (or cloud ice), the cloud drops (or cloud ice) are forced to evaporate to maintain the equilibrium until they are all evaporated. As the saturation adjustment strategy conventionally is not applied in subsaturated conditions for precipitation particles (e.g., raindrops, snow, etc.), it should be denoted as a partial equilibrium assumption.”.

2) As per comment (1) it would be interesting to see an additional experiment (maybe in the supplement) that assumes thermodynamic equilibrium only for the liquid phase, but not for ice. The nonequilibrium effect is generally assumed to be much stronger during ice than during liquid cloud formation due to higher supersaturations. Such an experiment would show how accurate this assumption is, and whether applying saturation adjustment and isotopic equilibrium during liquid cloud formation (as done in the models mentioned above) is reasonable or not.

Reply: Thanks for the nice suggestion. We added an extra simulation assuming thermodynamic equilibrium for the liquid phase only (LiqEQ). As shown in Fig. R1. the decrease in δDv was reduced by 20-50‰ in the LiqEQ run before the frontal passage in the early morning of 12 June, but enhanced by 10‰ at one point of frontal passage (06:00-12:00 LST on June 12). As shown in Figure R1b, the decrease in δD_l was enhanced when the saturation adjustment treatment was applied. But we would expect that this “partial equilibrium” simulation would produce results sitting somewhere between those of the CTRL and EQ runs. So, we are not very confident of including these new results. Due to the interest of time, we would like to leave it out of this revision. In the mean time, we will re-check/re-run the simulations and see if something more concrete can be obtained for the next revision (if allowed).

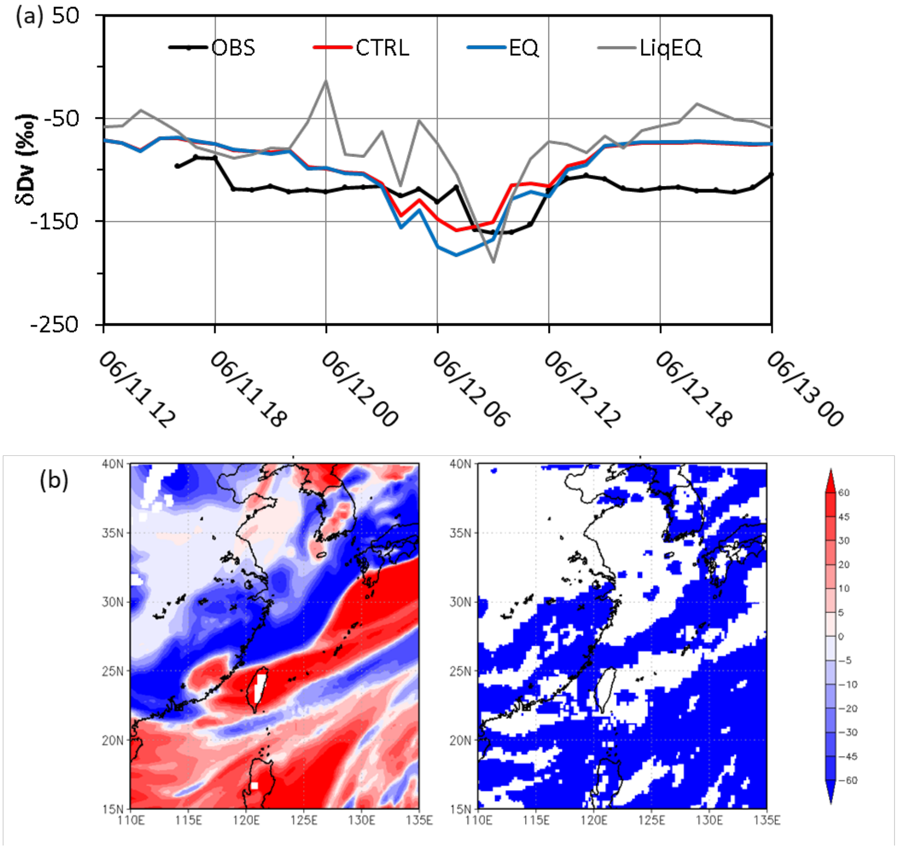

Figure R1. (a) Same as Fig. 11a. Simulated (CTRL: red line) and observed (OBS: black line) water vapor δDv (in ‰) at "AS" on 11-13 June 2012; and (b) Difference in simulated δD (in ‰) of water vapor (left) and liquid-phase condensates (right) at 850 hPa between CTRL and LiqEQ on 12 June, 2012.

Due to the low diffusivities of water molecules in ice there is no homogenization of isotopes in ice crystals and snow flakes. During deposition, the vapor only "sees" the outermost layer of the ice crystal / snow flake. Could you add a few sentences on how you define the isotopic composition of this outermost layer in the NTU scheme? For example, is it equal to the bulk composition of ice / snow, or the composition of the deposition flux? What is your rationale for one or the other?

Reply: Thanks for the suggestion. We added the following descriptions: "In Eq. (6), the activity of water stable isotope depends on the composition of the particle. For ice particles, the model cannot trace the history of water stable isotope deposition and thus cannot distinguish between the surface layer from the inner core of the ice particles. Therefore, the water stable isotope activity of ice-phase hydrometeor is assumed to depend on its bulk composition (i.e. assuming well-mixed). In reality, however, there is no homogenization of isotopes in ice particles due to the low diffusivities of molecules in ice. *Blossey et al.* [2010], *Pfahl et al.* [2012] and *Dütsch et al.* [2016] dealt with this problem by setting the ice particle's isotope ratio equal to that produced by vapor deposition. This is an effective approach as only the most recently deposited ice is exposed to the vapor. However, during evaporation the mass exchange depends heavily on the residual composition, making the treatment rather tricky. Before a better solution is devised, this study adopted the bulk composition approach for both condensation and evaporation processes." We also mentioned a suggested approach in the conclusion section as the following: "The problem in determining the activity of water stable isotope in ice particles without knowing the inhomogeneity of chemical composition in the bulk ice, as mentioned at the end of section 2.1 is another issue worthy of further study. To accommodate the different conditions between condensation and evaporation, it might be feasible to assume that the water stable isotope activity is determined by the vapor phase during condensation following the approach of *Blossey et al.* [2010], *Pfahl et al.* [2012] and *Dütsch et al.* [2016]; whereas for the evaporation process, one may assume a well-mixed bulk composition for determining the isotope activity as done in this study."

3) Supersaturation and associated nonequilibrium effects are especially important for the second-order isotope parameter deuterium excess ($d=\delta D - 8*\delta^{18}O$). It would be interesting to see the results of this case study also for deuterium excess, which would, however, require having $H_2{}^{18}O$ in the model. This could be something for a future study.

Reply: Deuterium was selected because we expect that the fractionation effect for $H_2{}^{18}O$ would be less significant according to Fig. 2. But, we do agree that it will be interesting to test it out in the future. This is mentioned in the discussion section as "Finally, whether the nonequilibrium effects are important for the second-order isotope parameter, deuterium excess, is an interesting subject worthy of further investigation by including the description of $\delta^{18}O$ isotope in the model."

4) There are some minor English mistakes / typos, which I won't correct here. I suggest to ask someone for proofreading before resubmission.

Reply: Thanks for the suggestion. We have asked help for proofreading in the revision.

Specific comments

Line 63: Better "liquid or ice and gas phases". For ice, supersaturation is (presumably) even more important than for liquid.

Reply: Agree. Revised accordingly.

Line 195: I personally don't like the factor 1000 in the definition of δ, because it is not part of the definition but part of how the definition is expressed. A similar thing would be to define pressure as $p = F/A * 0.01$ because it is often expressed in hPa. But this is a detail.

Reply: Agree. We have remove the factor "1000" from Eq. (9).

Line 207: "the ratio between the HDO concentration (QIV) and QV changes rather linearly with height." I am confused about this sentence. It does not seem linear in Fig. 4b.

Reply: The absolute values of QIV and QV vary exponentially with height, but the ratio QIV:QV is quite linearly varying (which cannot be seen in Fig. 4b). We have modified the sentence as: "Although the concentrations of water vapor (QV) and HDO (QIV) usually decrease exponentially with height, their ratios (i.e., QV:QIV) vary rather linearly with height." Hope it is less confusing now.

Line 260-262: This sentence is not very clear. Do you mean depletion due to rainout of heavy isotopes? Strong fractionation would otherwise lead to higher δD in the hydrometeors.
Reply: Yes, the low δD around the frontal system is due to precipitation. We modified the sentence as: "Secondly, precipitation inside the frontal system caused a strong reduction (fractionation) in δD of hydrometeors as can be seen in Figs. 8e and 8f"

Line 283: Better "low δDV" instead of "significant δDV".
Reply: Revised accordingly.

Line 304: Do you mean δDI (instead of δDV)?
Reply: Thanks for the correction. It is revised accordingly.

Line 322: Better "the decrease in δDV was overestimated", because δDV itself is underestimated.
Reply: Revised accordingly.

Line 328: For consistency I would also discuss the NoFrac simulation already in the results section.
Reply: Agree. We have added a brief description of the NoFrac result in the results section.

Line 370: Reference for precipitation measurements?
Reply: For precipitation measurements from the Central Weather Bureau of Taiwan, we added the website of CWB Taiwan in section 2.3 in the revised draft.

Line 380: Also evaporation from the ocean can have a large influence on the isotope values close to the surface. As far as I understood, this is not explicitly considered in the model. Is that correct?
Reply: This is correct. We added a few sentence in the last paragraph of discussion to clarify this: "In addition, the evaporation from the ocean is assumed as in equilibrium between liquid and vapor phases. This assumption may also affect the simulation of δD in the model, and the process needs to be explicitly considered in the future.

Line 403: "overestimate the decrease of δD", or "underestimate δDV".
Reply: Revised accordingly.

Line 410: Same as line 403 but the other way round.
Reply: Revised accordingly.

Fig. 1: Write out the terms CCN, GCCN, IN in the figure caption.
Reply: Thanks for the suggestion. We modified the caption of Fig. 1 as: "Schematics of modified NTU scheme. The blue boxes are the hydrometeors considered in the model and the H/D indicated that both H_2O and HDO are included. The arrows are the microphysical processes considered and the light blue boxes are cloud condensation

nuclei (CCN), giant CCN (GCCN) and ice nuclei (IN)."

Fig. 4b: Labels for the axes would be helpful.
Reply: Revised accordingly

Fig. 6: Is this a daily average? Please specify.
Reply: It is at 08:00 LST on 11 and 12 June. We added the information in the caption.

Fig. 7: For consistency "observed (OBS: black line)". Rotate time axis labels so they are more readable.
Reply: Revised accordingly

Fig. 10: Figure resolution is bad, labels are small and not readable. Where do you calculate the vertical distribution? Is the x axis in space or time?
Reply: Figure quality is probably degraded during the conversion to pdf file, but we have revised the figures with larger labels. The area over northern Taiwan is defined as (121-123˚E, 25-27˚N), and the x axis is time. We have modified the figure caption to include these information.

Fig. 12: Add units.
Reply: Revised accordingly

References

Blossey, P. N., Z. Kuang, and D. M. Romps (2010). Isotopic composition of water in the tropical tropopause layer in cloud-resolving simulations of an idealized tropical circulation. J. Geophys. Res., 115, D24309, doi:10.1029/2010JD014554

Pfahl, S., H. Wernli, and K. Yoshimura (2012). The isotopic composition of precipitationfrom a winter storm – a case study with the limited-area model COSMOiso. Atmos. Chem. Phys., 12, 1629–1648, doi:10.5194/acp-12-1629-2012

Reply: Thanks for providing these useful references.