Microphysical controls on the isotopic composition of wintertime orographic precipitation

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Key Points

- Distinct isotopic signatures of microphysical processes can be determined.
- Temperature and mountain height control precipitation isotopic composition more strongly
- 5 than CDNC.
- Microphysical changes with CDNC, temperature, and mountain height establish isotopic

7 composition.

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X - 2 MOORE ET AL.: ISOTOPIC COMPOSITION OF OROGRAPHIC PRECIPITATION Abstract. The sensitivity of mixed-phase orographic clouds, precipita-8 tion and their isotopic content to changes in dynamics, thermodynamics and 9 microphysics is explored in idealized two-dimensional flow over a mountain 10 barrier. These simulations use the Weather Research and Forecasting (WRF) 11 Model with stable water isotopologues (HDO and $H_2^{18}O$), which have been 12 integrated into the Thompson microphysics scheme within WRF as part of 13 the present project. In order to understand how the isotopic composition of 14

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precipitation ($\delta^{18}O_{precip}$) is fixed, the mountain height, temperature, and the 15 prescribed cloud droplet number concentration (CDNC) have been varied 16 in a series of simulations. For the given range of values explored in this work, 17 changes in mountain height and temperature induce stronger responses in 18 domain-averaged $\delta^{18}O_{precip}$ than do changes in CDNC by a factor of approx-19 imately 10. The strongest response to changing CDNC leads to local vari-20 ations of $\delta^{18}O_{precip}$ of about 3%, though those occur in regions of weak pre-21 cipitation (<0.1 mm hr⁻¹). Changes in $\delta^{18}O_{precip}$ can be understood through 22 the microphysical pathways by which precipitable hydrometeors are formed 23 and by the isotopic signature associated with each pathway. The decrease 24 in $\delta^{18}O_{precip}$ with increasing mountain height is not just a function of decreas-25 ing temperature, but also reflects the changing contributions and distinct isotopic signatures of riming of cloud liquid and vapor deposition onto snow, 27 the leading sources of precipitation in these simulations. The changes in $\delta^{18}O_{precip}$ 28 with mountain height, temperature and CDNC are governed in part by the 29 microphysical pathways through which precipitating hydrometeors are formed 30 and grow. 31

1. Introduction

Precipitation that forms due to interaction with mountain barriers, or orographic precipitation, is an important contributor to surface water resources. In particular, runoff from rainfall and melting of the mountain snowpack feed into river basins that provide water to a number of heavily populated regions. As the amount and location of precipitation on the mountain barrier will determine the volume of runoff and the watershed into which it flows, understanding all of the factors that influence and control orographic precipitation is essential for current and future forecasts of this necessary resource.

There have been extensive regional studies regarding the formation and behavior of 39 orographic precipitation [e.g. Hobbs, 1975; Smith et al., 2005; Smith and Evans, 2007; 40 Zubler et al., 2011]. The total precipitation and its spatial distribution have been found to 41 be dependent upon several variables, including the orientation and geometry of the terrain, 42 atmospheric stability, orographic flow dynamics and cloud microphysics [e.g. Colle, 2004; 43 Galewsky, 2008; Muhlbauer and Lohmann, 2008]. In terms of cloud microphysics, the 44 different pathways through which precipitating hydrometeors grow can be more or less 45 efficient and thus greatly influence the amount of precipitation. For example, in mixed-46 phase orographic clouds, the growth and fallout of snow and graupel may be enhanced by 47 the "seeder-feeder" mechanism [*Reinking et al.*, 2000], wherein ice crystals grow by vapor 48 deposition in an ice cloud aloft before sedimenting to lower levels in the cloud where 49 the ice continues to grow by collecting cloud droplets (riming). This enhanced low-level 50 riming increases the fallspeed of snow and also the overall precipitation efficiency of the 51 cloud [Mitchell et al., 1990; Borys et al., 2003], thereby augmenting precipitation on the 52

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⁵³ windward side of the mountain at the expense of the transport of hydrometeors to the ⁵⁴ leeward slope (and the resulting precipitation there, which is known as "spillover").

The stable isotopologues of water $(H_2^{16}O, HDO, H_2^{18}O)$ have been used in precipita-55 tion analysis dating back to the initial work of *Dansgaard* [1952]. In the mid-latitudes, 56 where westerlies impinging on north-south oriented mountain ranges form the motiva-57 tion for our idealized simulations, the isotopic composition of precipitation is primarily 58 temperature-dependent [e.g. Dansgaard, 1964; Noone and Simmonds, 2002; Jouzel, 2003; 59 Lee et al., 2007], such that the ratio of the heavy (e.g., $H_2^{18}O$) to light ($H_2^{16}O$) isotopes 60 correlates positively with temperature. In mountainous regions, the relationship between 61 the isotopic composition of precipitation and temperature is additionally linked with alti-62 tude [Dansgaard, 1964]. Air cools as it rises along the upslope on the windward side of a 63 mountain, and the progressive removal of precipitation produces a gradient in the isotopic 64 composition with altitude. This leads to precipitation enriched in heavy isotopes forming 65 at lower altitudes, and more depleted precipitation (i.e., with lower isotopic ratios) at 66 higher altitudes, as well as on the downslope in the lee of the mountain peak [Smith et 67 al., 2005]. This isotopic gradient was connected to the fractional removal of water by a 68 mountain barrier and its drying ratio by *Smith et al.* [2005]. The relationship between 69 isotopic composition and altitude has also been used to relate paleoclimate proxies for the 70 isotopic composition of precipitation to past mountain elevation [Poage and Chamberlain, 71 2001; Rowley et al., 2001]. However, as demonstrated by Galewsky [2009] and Lechler and 72 Galewsky [2013], in different dynamical regimes, the airflow over the mountain can com-73 plicate the relationship between the isotopic composition of precipitation and the altitude 74 of a mountain barrier. 75

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In addition to the dynamical influences on orographic precipitation and its isotopic 76 content, microphysical processes can also modify the isotopic signature of precipitation. 77 Coplen et al. [2015] connected variations in the isotopic content of precipitation in land-78 falling extratropical cyclones with changes in the storm structure and different pathways 79 of precipitation formation. Observations of snowfall from the Sierra Nevada [Demoz et al., 80 1991] and snowfall and cloud liquid in Colorado [Lowenthal et al., 2011] suggested that the 81 isotopic composition of snowfall is influenced by the degree of riming. By sampling both 82 the isotopic and chemical composition of both snowfall and cloud droplets at a mountain-83 top site in Colorado, Lowenthal et al. [2011] related the degree of riming of snowfall to the 84 chemical composition of the snow and concurrently sampled cloud droplets. They found 85 that snow mass formed mainly through riming was more enriched and had an isotopic 86 signature that was similar to the cloud droplets. This relationship was then employed to 87 make predictions about the altitude at which snow formed through vapor deposition.

The relative role of riming in mixed-phase orographic precipitation can be reduced by 89 decreasing temperature, through the glaciation of liquid clouds, and by increasing aerosol 90 concentrations, which tend to lead to more numerous and smaller cloud droplets that 91 are less likely to be collected by falling snow [Pruppacher and Klett, 1997; Wang and 92 Ji, 2000]. Increased aerosol concentrations can also suppress or delay the formation of 93 precipitation in liquid-only clouds by reducing the efficiency of collision and coalescence 94 processes [Albrecht, 1989; Ramanathan et al., 2001]. While aerosols have the potential 95 to impact individual microphysical processes that contribute to precipitation, their influ-96 ence on the amount and distribution of orographic precipitation has not been definitely 97 established and appears to depend strongly on the environmental conditions of the region 98

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⁹⁹ being considered [*Borys et al.*, 2000, 2003; *Khain and Pokrovsky*, 2004; *Lynn et al.*, 2007; ¹⁰⁰ *Muhlbauer and Lohmann*, 2008; *Saleeby et al.*, 2013]. In a study of warm (liquid-only) ¹⁰¹ orographic clouds and precipitation, *Miltenberger et al.* [2015] suggested that interactions ¹⁰² between dynamical and microphysical processes can lead to regimes where the precipita-¹⁰³ tion and the precipitation efficiency are insensitive to changes in cloud droplet number ¹⁰⁴ concentration (CDNC), which is used as a proxy for aerosol concentrations.

To explore how such changes in microphysical processes influence mixed-phase oro-105 graphic precipitation and its isotopic content, we perform a number of idealized two-106 dimensional simulations in which the mountain height, temperature and CDNC are var-107 ied. Particular attention is paid to changes in the microphysical processes that contribute 108 to the growth of precipitating hydrometeors and how those processes and their isotopic 109 signatures control the amount, distribution and isotopic content of precipitation in these 110 experiments. By tracking the isotopic ratio associated with precipitation growth processes 111 within these simulations, we determine if each microphysical process has a distinct isotopic 112 signature and how each process contributes to the overall isotopic signal of precipitation 113 in different regimes. 114

Although the results presented in this paper are based on idealized simulations, they represent a step towards constructing an isotope-enabled regional modeling capability for WRF. Previous work with isotope-enabled global models [e.g. *Noone and Simmonds*, 2002; *Vuille et al.*, 2003; *Lee et al.*, 2007; *Field*, 2010] has advanced our knowledge of how largescale processes affect isotopic composition. However, the limitations of global climate models (GCMs) in representing topography and cloud-scale processes leaves room for higher-resolution regional models that more faithfully represent such fine-scale phenomena

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¹²² [*Prein et al.*, 2015]. The work of *Pfahl et al.* [2012] provides an example of how a fine-scale ¹²³ regional model can improve the representation of isotopic signals over that of an isotope-¹²⁴ enabled GCM. By using a regional model with horizontal grid spacing of O(1 km), this ¹²⁵ work aims to understand orographic precipitation resolving scales much finer than those ¹²⁶ represented in a typical isotope-enabled GCM whose horizontal grid spacings vary from ¹²⁷ approximately 200 to 400 km [*Conroy et al.*, 2013].

2. Model and Experiments

2.1. Model Setup

To conduct the orographic precipitation experiments, we use the WRF model version 128 3.5.1 [Skamarock and Klemp, 2008] provided by the Mesoscale and Microscale Meteorol-129 ogy Division of the National Center for Atmospheric Research. The model is configured 130 to perform simulations of idealized 2D flow over a hill. The domain consists of 300 grid 131 points with 2 km spacing in the horizontal direction and 105 vertical levels whose spacing 132 varies from 25-200 m in the lower 5 km and is uniform above 5 km. The duration of each 133 simulation is twelve hours. The Thompson microphysics scheme [Thompson et al., 2008] 134 was chosen for this work because a study of wintertime precipitation in a mountainous re-135 gion of the western United States found that, along with one other scheme, the Thompson 136 microphysics scheme provided the best representation of cold season snowfall [Liu et al., 137 2011]. In addition, the Thompson scheme includes a detailed treatment of the riming of 138 cloud droplets by snow (as in *Saleeby and Cotton* [2008]), which has proved important for 139 realistic simulation of the effects of pollution on riming in mixed-phase orographic clouds 140 [Lohmann, 2004; Saleeby and Cotton, 2008; Saleeby et al., 2011]. 141

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As noted previously, different atmospheric regimes are simulated in order to study the 142 response of model microphysics. The Thompson microphysics scheme allows the user to 143 specify the CDNC value, which is utilized in this work as a proxy for aerosols. The chosen 144 CDNC values represent conditions that range from pristine to polluted. The working 145 assumption is that for high aerosol loading, there are more cloud condensation nuclei 146 (CCN) and thus a higher CDNC value, while a lower CDNC value indicates a scenario 147 with few aerosols and therefore fewer CCN. More specifics about the experimental setup 148 are given in section 2.3. 149

2.2. Isotopic implementation

In its default configuration, the Thompson scheme only treats microphysical transfers of 150 the standard isotopologue of water $(H_2^{16}O)$ among water vapor and the different hydrom-151 eteors included in the scheme: cloud liquid, rain, cloud ice, snow and graupel. As part of 152 the present project, we have extended the Thompson scheme so that the microphysical 153 transfers of the stable isotopologues of water (HDO and $H_2^{18}O$) are also included. The 154 isotopic composition of water vapor and each hydrometeor is tracked, and the exchanges 155 of the heavy isotopologues of water are accounted for during each microphysical process 156 represented in the Thompson scheme. Isotopic fractionation — the unequal exchange of 157 heavy and lighter isotopologues of water — is accounted for in processes that involve the 158 deposition of vapor onto liquid or ice hydrometeors and those involving the evaporation of 159 liquid phase hydrometeors (rain or cloud liquid). Other processes that involve the trans-160 fer of whole hydrometeors from one category to another (e.g., freezing, melting, riming), 161 occur without fractionation. As in Bony et al. [2008], Blossey et al. [2010] and Pfahl et 162 al. [2012], the sublimation of ice phase hydrometeors (snow, cloud ice, graupel) is also 163

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assumed to occur without fractionation, so that the vapor produced by sublimation of 164 snow, for example, has the same isotopic composition as the snow. While sublimation 165 is expected to produce vapor from the outer shell of an ice phase hydrometeor, and this 166 layer may not have the same isotopic composition as the particle as a whole, tracking the 167 composition of individual layers within the crystals is judged to be too complicated and 168 expensive to include in the present implementation. Note that the implementation, which 169 follows Blossey et al. [2010], includes few approximations in its representation of isotopic 170 exchanges beyond the assumption that the isotopic composition of each hydrometeor cat-171 egory in a given grid cell is uniform and that no fractionation occurs during sublimation. 172 The model uses time steps on the order of a few seconds, so that only cloud liquid and 173 vapor are assumed to equilibrate within a single time step. Other processes are integrated 174 in time explicitly by the model. A more detailed description of the water isotope physics 175 is given in Appendix A. 176

The quality of the isotopic simulation depends strongly on the representation of the 177 standard isotopologue of water. If the microphysics scheme and the broader model do a 178 poor job in representing the amount and distribution of precipitation of the standard iso-179 topologue of water, this will be reflected in the isotopic composition as well. Encouraged 180 by the performance of the Thompson scheme within WRF on wintertime orographic pre-181 cipitation [Liu et al., 2011] and by the representation of isotopic composition in tropical 182 convection in a similar implementation of water isotopologues in *Blossey et al.* [2010], we 183 proceed with the simulations here. 184

For the isotopic analysis, our results on $H_2^{18}O$ are presented in delta-notation such that $\delta^{18}O = 1000 \left(\frac{R}{R_o} - 1\right)$, where R is the isotopic ratio of $H_2^{18}O$ in a specified water

¹⁸⁷ species and R_o is the isotopic ratio of the standard. While HDO is also included in the ¹⁸⁸ microphysics scheme, the additional information that can be gained by considering both ¹⁸⁹ HDO and H₂¹⁸O will be left to future work.

2.3. Experimental Setup

Several experiments are conducted that alter the initial temperature profile, mountain 190 height (800, 1500 and 3000 m), and the CDNC (25, 100, 200, 400, and 800 cm⁻³). Two 191 initial temperature profiles are used here and are referenced as warm or cold based on the 192 surface temperature of the upstream sounding ($T_{sfc} = 7^{\circ}C$ and $0^{\circ}C$, respectively). Ex-193 periments are referenced by abbreviations (e.g., W800m), which indicate the temperature 194 sounding (W=warm or C=cold) and mountain height settings. The setup and initial con-195 ditions, including the temperature profiles, are similar to those in Muhlbauer et al. [2010], 196 with a mountain half-width of 20 km and a horizontal wind profile that is a constant 197 15 m s^{-1} below 10 km and linearly increases to 40 m s⁻¹ at the top model layer (30 km). 198 To generate the initial vapor conditions for $H_2^{18}O$ and HDO, a Rayleigh distillation 199 profile is generated assuming equilibrium with ocean water at 20° C, which represents the 200 average temperature of the ocean surface where the initial isotopic signature of the air 201 mass will be set. The model's initial conditions for the isotopic content of water vapor are 202 interpolated from this Rayleigh profile based on the water vapor mass mixing ratio. As 203 the cold sounding is drier than the warm sounding, it is also more depleted, such that the 204 δ^{18} O of vapor at the surface is 8% less than that of the warm sounding. Neither liquid 205 nor ice condensate exists initially, and therefore their isotopic compositions do not need 206 to be initialized. 207

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2.4. Model Validation

The model's ability to simulate orographic clouds and precipitation is on par with previ-208 ous studies. The results are very similar to the WRF simulations in Muhlbauer et al. [2010], 209 despite the use of a different microphysical scheme. There are some small differences in 210 the simulated orographic clouds, and our experiments produce more accumulated precip-211 itation. However, these deviations can be attributed to our implementation of a longer 212 simulation time and a larger range of CDNC values in addition to the choice of microphys-213 ical scheme. The changes in the liquid orographic cloud are also similar to results seen 214 by Xiao et al. [2014], who also used the same idealized WRF setup, but coupled with a 215 detailed bin microphysics scheme and a warmer initial temperature profile. The evolution 216 of cloud liquid and microphysical processes as CDNC increases in our cold experiments is 217 similar to that of Saleeby et al. [2006], who used the Colorado State University - Regional 218 Atmospheric Modeling System with a different microphysics scheme to simulate realistic 219 wintertime orographic clouds in northern Colorado. 220

This project represents the first use of this isotope-enabled version of the Thompson 221 scheme within WRF. As the present modeling study is idealized and the incorporation of 222 water isotopologues into the real-case forecasting capability of WRF is not complete, we 223 focus on the performance of the scheme within the present simulations. First, the isotopic 224 composition of precipitation along the upslope of the mountain approximately conforms to 225 that of a Rayleigh process and is slightly more depleted than the Rayleigh process due to 226 dynamical effects of the mountain [Galewsky, 2009] and the formation of precipitation from 227 more depleted vapor above the surface of the mountain (supplemental Fig. S1). Second, 228 in section 3.3, closed budgets for the surface precipitation are constructed that explain 229

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the precipitation itself and its isotopic composition in terms of the various microphysical processes that contribute to the formation and growth of precipitating hydrometeors. Last, the isotopic composition of water vapor and hydrometeors described in sections 3.1 and 3.4 shows the expected influence of microphysical processes on isotopic composition, such as isotopic equilibration of cloud liquid and water vapor, vapor deposition onto ice and the evaporation of rain in subsaturated conditions.

It should also be noted that the average δ^{18} O values of total precipitation compare well 236 with observations in conditions similar to those used here for initial conditions. Ander-237 son et al. [2015] calculated the average δ^{18} O of snowpack using the Isotopes in Rocky 238 Mountain Snowpack (IRMS) database, and found that values ranged between -10% and 239 -25‰, which compares well with the range of δ^{18} O in the average precipitation for our 240 experiments (approximately -10% to -16% in warm simulations and -19% to -26% in 241 cold experiments). Comparable $\delta^{18}O_{precip}$ values (-12\% to -24\%) were measured during 242 a 1985 March storm in Kingvale, CA, which is located upwind of the Sierra Nevada crest 243 at an elevation of 1859 m [Warburton et al., 1993]. Warburton and DeFelice [1986] ana-244 lyzed samples in the Central Sierra Nevada, and found that snow formed through vapor 245 deposition had a δ^{18} O signature that ranged from -18.4‰ to -22.9‰, which corresponds 246 well with our cold temperature profile experiments (see further discussion in section 3). 247 The snow samples from the same study that indicated growth by a combination of riming 248 and vapor deposition, were less depleted and ranged between -6.4‰ and -16.8‰, which 249 resembles results in our warm simulations (see section 3). Values similar to Warburton 250 and DeFelice [1986] were measured in Colorado by Lowenthal et al. [2011] for snow that 251 had undergone little riming. In the same study, snow that experienced more riming (as in-252

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dicated by higher concentrations of sulfate), was less depleted and ranged between -15.6‰
and -20.4‰.

3. Model Results

3.1. Reference Simulation

To outline the general characteristics of the cloud and precipitation in these simulations, 255 the simulation with the warmer sounding $(T_{sfc} = 7^{\circ}C)$, a 800 m high mountain, and a cloud 256 droplet number concentration (CDNC) of 200 cm^{-3} is chosen as the reference simulation. 257 Fig. 1b shows the average simulated mass of cloud liquid along with the combined mass 258 of cloud ice and snow for the reference simulation. (Figs. 1a and 1c will be discussed 259 in section 3.2.) The figure combines cloud ice and snow together, as the setup of the 260 Thompson scheme quickly leads to the conversion of cloud ice to snow, and as a result, 261 produces little cloud ice [Thompson et al., 2008]. Note that while a wave cloud exists 262 aloft and downstream of the mountain in these simulations, our focus is on the cloud and 263 precipitation over the mountain, where almost all precipitation is produced. 264

The orographic cloud in the reference simulation does not extend higher than 4 km and has a much higher mass of cloud liquid than a combined mass of snow and ice (Fig. 1b). The frozen hydrometeors occur predominately upstream of the mountain peak with only a little spillover (\sim 10 km) to the downstream side. For the most part, the liquid and ice/snow regions of the cloud overlap, except on the leeward slope, where the glaciated cloud is located above the liquid one.

The isotopic values of vapor, cloud liquid, rain and ice/snow for the reference simulation are presented in Fig. 2. Cloud liquid isotopic values range from approximately -7% near the mountain surface to -22% at cloud top (Fig. 2c). Isotopic equilibrium is

enforced between cloud liquid and vapor, so that decreasing δ^{18} O of cloud liquid with 274 height is expected given that the vapor δ^{18} O shows the same trend (Figs. 2a and 2c). The 275 cloud liquid that extends further leeward has roughly the same δ^{18} O value as the cloud 276 liquid on the corresponding windward side, so there is no obvious δ^{18} O difference between 277 the windward and leeward cloud liquid. As expected from rainout (i.e., the progressive 278 removal of heavy isotopologues by precipitation across the mountain barrier [Clark and 279 Fritz, 1997; Smith et al., 2005]), the δ^{18} O of water vapor does show asymmetry about 280 the mountain and is more depleted at low levels further downstream of the mountain. 281 The cloud ice/snow δ^{18} O values range from -10% near cloud base to -35% at cloud top 282 (Fig. 2d). 283

Fig. 3a shows the profile of accumulated precipitation across the mountain for the refer-284 ence simulation along with a number of different CDNC concentrations. (The sensitivity 285 to CDNC will be discussed in the following section.) The precipitation for the reference 286 simulation (CDNC = 200 cm^{-3} , red line) peaks over the mountain top and is nearly sym-287 metric, with slightly more precipitation falling downwind of the peak and a spillover ratio 288 of 0.56 (Tab. 1). (The spillover ratio is the ratio of the accumulated leeward precipitation 289 to total precipitation.) Most of the precipitation falls as rain at the surface, with similar, 290 smaller amounts of snow and graupel (Tab. 1). The isotopic composition of the accumu-291 lated precipitation $\delta^{18}O_{precip}$ in the reference simulation (Fig. 3b, red line) becomes more 292 enriched as one ascends the lower slope on the upwind side of the mountain. This is also 293 seen in the isotopic composition of rain in Fig. 2b, and is associated with a shift from rain 294 resulting from the melting of snow that was formed aloft through vapor deposition, to rain 295 and snow that grew through the conversion and accretion of cloud liquid. Such changes 296

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in the microphysical pathways through which precipitating hydrometeors are formed and 297 their impact on $\delta^{18}O_{precip}$ will be discussed in greater detail in section 3.3. Following this 298 peak in $\delta^{18}O_{precip}$ at x=280 km, the isotopic composition of precipitation falls off across 299 the mountain as the heavier isotopes are removed preferentially through fallout. As noted 300 in the introduction, this may be modeled approximately as a Rayleigh process [Smith et 301 al., 2005, though there are some complications due to dynamical response to topography 302 [Galewsky, 2009] and microphysical effects. The increase in $\delta^{18}O_{precip}$ on the downslope 303 at x=315-320 km is associated with the fractionation of evaporating rain once it passes 304 downstream of the orographic cloud (see also Fig. 2b-c.) As shown in previous studies 305 [e.g. Stewart, 1975; Lawrence et al., 1998; Bony et al., 2008; Risi et al., 2008], evaporation 306 in subsaturated conditions tends to enrich the rain and deplete the vapor, as the lighter 307 H_2O will more quickly move from the liquid to the surrounding vapor. 308

3.2. Sensitivity to CDNC

Next, the sensitivity of the reference simulation to changes in CDNC (as a proxy for aerosol variations) is shown. This is interesting both as a way to understand whether aerosol impacts on orographic precipitation [e.g., *Rosenfeld et al.*, 2008] could impact the isotopic composition as well, and as an example of how changing the microphysical processes which contribute to precipitation could impact the amount, distribution and isotopic composition of orographic precipitation.

Three cases with increasing values of CDNC are shown in Fig. 1, which illustrates potential changes in the orographic cloud with CDNC. As the CDNC value increases, the conversion of cloud to rain and the riming of cloud liquid by snow become less efficient, resulting in an increase in both the amount of cloud liquid and the leeward region it spans.

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While there is already leeward spillover of cloud liquid in the 25 $\rm cm^{-3}$ case, cloud liquid 319 extends an additional 15 km down the leeward side in the 800 $\rm cm^{-3}$ experiment, with the 320 region of maximum mass mixing ratio (red filled contours) also reaching approximately 321 5 km further downstream. This shift in the leeward extent of cloud liquid is mirrored in 322 the isotopic composition of precipitation in Fig. 3b, where the increase in $\delta^{18}O_{precip}$ due to 323 rain evaporation occurs farther downstream as CDNC increases. On the windward slope, 324 the location of the leading edge of the cloud does not change in all of the warm 800 m 325 experiments. Increases in CDNC have little impact on the location of snow and cloud 326 ice: both the horizontal and vertical extent of the glaciated cloud remain the same. The 327 mass mixing ratio, however, does decrease very slightly (note change in contours over the 328 mountain peak region) as the CDNC increases, which is opposite to and of much smaller 329 magnitude than the trend found for cloud liquid. The isotopic signatures of cloud liquid 330 and combined cloud ice/snow are similar to those of the reference simulation (Fig. 2) and 331 are not shown. 332

As in the reference simulation, most of the precipitation in the simulations with vary-333 ing CDNC falls as rain (see Tab. 1) with small, similar amounts of accumulated snow 334 and graupel. The third column in Tab. 1 indicates that the accumulated precipitation 335 decreases as CDNC increases, and is reduced by more than half between the 25 $\rm cm^{-3}$ 336 and 800 cm^{-3} experiments. Fig. 3a shows that the location of the maximum precipita-337 tion shifts leeward as CDNC increases, which has been previously observed in wintertime 338 orographic precipitation [Jirak and Cotton, 2006; Saleeby et al., 2011]. Among the dif-339 ferent mountain heights and temperatures considered here, the magnitude of the shift 340 is strongest and most obvious for the W800m experiments, where there is a difference 341

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 $_{342}$ of approximately 10 km between the precipitation peaks in the 25 cm⁻³ and 800 cm⁻³ $_{343}$ simulations. This shift is also evident in the spillover calculations in Tab. 1.

The domain can be broken down into three smaller regions: upstream of the peak (up 344 to 290 km into the domain), around the peak (290–310 km) and downstream of the peak 345 (310 km onwards). As Fig. 3a indicates, most of the precipitation falls in the first and 346 second regions. The influence of CDNC on precipitation is also most pronounced in these 347 regions. However, the CDNC impact on the $\delta^{18}O_{precip}$ is slightly different. Fig. 3b illus-348 trates that the largest $\delta^{18}O_{precip}$ difference between simulations occurs in the first region, 349 but over the second region, variation in the isotopic signal is small (< 1%). In the third 350 region, downstream of the mountain peak, the accumulated precipitation is relatively 351 unchanged between simulations, but there is some separation in the $\delta^{18}O_{precip}$ of approxi-352 mately 2% at x=310 km before the effects of rain evaporation enter further down the lee 353 slope. The slopes of $\delta^{18}O_{precip}$ across the peak differ, with the steepest change in $\delta^{18}O_{precip}$ 354 across the peak in the simulation with the largest precipitation (CDNC= 25 cm^{-3}) as one 355 would expect due to the effect of rainout [Smith et al., 2005]. This leads the 25 cm⁻³ 356 simulation to have the largest upstream-downstream difference in $\delta^{18}O_{precip}$ around the 357 peak (x=290-310 km). 358

3.3. Microphysical pathways

To better understand the changes in precipitation and its isotopic composition across the mountain, we consider the budget for the total mass of precipitating hydrometeors (rain, snow and graupel combined) in these simulations, integrated in time and over the whole domain or a sub-region of the domain. Since isotopic composition is unchanged by exchanges between rain, snow and graupel by freezing, melting or aggregation, we focus

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³⁶⁴ on the sources which determine the isotopic composition of the precipitation: autocon-³⁶⁵ version/accretion of cloud liquid or cloud ice, riming of cloud liquid and exchanges with ³⁶⁶ vapor by deposition or sublimation/evaporation. In this budget, surface precipitation, P, ³⁶⁷ is a sink of hydrometeor mass and is balanced by various microphysical sources of rain, ³⁶⁸ snow and graupel as well as advection and storage of these hydrometeors:

$$P = Q_{LAUT} + Q_{LACC} + Q_{IAUT} + Q_{IACC} + Q_{RIM} + Q_{DEP} + Q_{SUB} + Q_{ADV} - Q_{STOR}.$$

$$(1)$$

Here, the sources of hydrometeor mass include microphysical processes, such as autocon-370 version of cloud liquid (LAUT), accretion of cloud liquid (LACC), autoconversion of cloud 371 ice (IAUT), accretion of cloud ice (IACC), riming of cloud liquid (RIM), vapor deposi-372 tion onto ice (DEP), sublimation of ice (SUB), along with those associated with moisture 373 flux convergence (labeled ADV for advection) and storage (STOR). The storage term is 374 negative because increases in hydrometeors in the domain over time come at the expense 375 of surface precipitation. Each of these terms are integrated over the duration of the sim-376 ulations and over the domain or a subset of the domain in the horizontal direction and 377 then normalized by the mountain half-width (20 km). Note that, because the Thomp-378 son microphysical scheme produces little cloud ice, much of the vapor deposition onto ice 379 phase hydrometeors that occurs in the domain contributes directly to snow growth. Other 380 microphysical schemes would likely have stronger vapor deposition onto cloud ice, so that 381 the autoconversion/accretion of cloud ice would be relatively more important and vapor 382 deposition relatively less important. Also, note that the net tendency of vapor deposi-383 tion (including deposition, sublimation and rain evaporation) has been averaged over the 384 simulation and then partitioned into regions of deposition and sublimation/evaporation 385 according to the sign of the mean tendency. 386

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³⁸⁷ A similar budget can be written for the mass of the heavy isotopologues, and the isotopic ³⁸⁸ composition of those contributions can be computed from the ratio of the contribution ³⁸⁹ to heavy isotope mass, e.g., $H_2^{18}O$, to that for the standard isotope, $H_2^{16}O$. The $\delta^{18}O$ of ³⁹⁰ hydrometeor mass generated by each process may then be computed as for precipitation ³⁹¹ itself, $\delta^{18}O_{precip}$.

In Fig. 4, the total precipitation and the contributions of the dominant microphysical 392 processes to precipitation and its isotopic composition are shown in three regions: the peak 393 (x=295-305 km) and the regions upwind and downwind of the peak. In the following, 394 the sources of precipitation in each region are analyzed. Note that the precipitation 395 produced in each region may fall to the surface there or be transported downstream. In 396 Figs. 4b–d, the contribution of each process in each region has been normalized by the 397 total precipitation in the domain for each case. These normalized contributions can be 398 interpreted as weights, which can be applied to the characteristic isotopic composition 399 from each process to determine $\delta^{18}O_{nrecin}$. 400

In the upwind region (Fig. 4b), riming of cloud droplets contributes most to the growth 401 of precipitating hydrometeors, with vapor deposition onto ice making the second largest 402 contribution in most cases. The 25 cm^{-3} simulation differs in the importance of auto-403 conversion and accretion of cloud liquid. Riming and autoconversion of cloud liquid both 404 have a direct dependence on the size of cloud droplets and therefore on CDNC. Accretion 405 of cloud liquid may also depend indirectly on CDNC if less rain is generated through 406 autoconversion as CDNC increases. As autoconversion and accretion of cloud liquid fall 407 off with increasing CDNC, the contribution from vapor deposition increases. The isotopic 408 signatures of the liquid processes are more enriched than the vapor deposition by approxi-409

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⁴¹⁰ mately 4-9‰ (Fig. 4f), and thus the precipitation in the lower CDNC simulations is more ⁴¹¹ enriched than the higher CDNC simulations. Overall, the microphysics explain the de-⁴¹² crease in both the accumulated precipitation and the isotopic content of the precipitation ⁴¹³ as seen in Fig. 3.

Riming of cloud liquid over the mountain peak is the largest source of precipitation in 414 the three regions and itself produces enough hydrometeor mass to account for half of the 415 surface precipitation in all cases except $CDNC=25 \text{ cm}^{-3}$ (Fig. 4c). Accretion of cloud 416 liquid and vapor deposition onto ice also contribute to precipitation over the peak. Similar 417 to the upwind region, accretion of cloud liquid decreases with increasing CDNC, though 418 more modestly, but riming actually increases. In this region, vapor deposition onto ice 419 is still the most depleted source term. However, the ice produced by vapor deposition is 420 more enriched above the peak than in the upstream region. The average δ^{18} O differences 421 between the ice produced by riming and vapor deposition over the peak range between 422 2-4‰ (Fig. 4g). Therefore, the variation in the source terms of precipitation with CDNC 423 over the peak produce little change in the $\delta^{18}O_{precip}$ formed there, in part because the 424 isotopic composition of the sources are more similar. 425

Precipitation production on the leeward slope derives predominantly from vapor deposition onto ice in addition to relatively small contributions from ice autoconversion and accretion of cloud liquid (Fig. 4d). One significant difference in this downwind region compared to the other two regions, is the presence of a large sink of precipitation mass caused by sublimation and rain evaporation. This pocket of sublimation/evaporation is expected due to subsidence and thus warming of air as it flows over the mountain peak. Though all of the microphysical source terms increase with CDNC in this region, removal

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of precipitation due to sublimation/evaporation essentially balances out the source terms, and the accumulated precipitation remains relatively constant in the different simulations (Fig. 3a). The smaller range of the δ^{18} O in the accumulated precipitation can be attributed to the similarity of the source terms in each simulation, except for that due to sublimation/evaporation which becomes more depleted with increasing CDNC.

3.4. Sensitivity to mountain height and temperature

The sensitivity of domain-integrated precipitation amount and its isotopic composition 438 to CDNC changes was also studied for a number of mountain heights (800 m, 1500 m 439 and 3000 m) and two temperature profiles (with $T_{sfc} = 0^{\circ}C$ and $7^{\circ}C)^{1}$. To understand 440 how precipitation and its isotopic content are related across these simulations, Fig. 5 441 shows their relationship when integrated over the whole domain (Fig. 5a) and over the 442 regions upstream of the peak, over the peak and downstream of the peak (Figs. 5b-d, 443 respectively). These regions are defined as above in section 3.3. As seen in Fig. 5a, 444 the response of total (domain-integrated) precipitation and its isotopic content to CDNC 445 changes — where it exists — is modest in comparison to that due to mountain height 446 and temperature. The only significant response of $\delta^{18}O_{precip}$ to CDNC occurs for small 447 precipitation amounts (< 5 mm) in the upwind region of W800m (Fig. 5b). Otherwise, 448 the change in isotopic content due to temperature exceeds that due to CDNC by a factor 449 of approximately 10 for the ranges of temperature and CDNC explored here. The weaker 450 sensitivity of precipitation to CDNC changes with increasing precipitation is reminiscent 451 of the work of Muhlbauer et al. [2010] for mixed-phase clouds and Miltenberger et al. 452 [2015] for warm clouds. The possibility remains that a model setup that yields weaker 453 precipitation might show a stronger sensitivity of precipitation to CDNC changes, as in 454

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⁴⁵⁵ *Miltenberger et al.* [2015]. However, the change in $\delta^{18}O_{precip}$ due to CDNC is unlikely to ⁴⁵⁶ increase far beyond the range seen in the reference case (W800m).

Since the response to CDNC is weak in many cases, the present section focuses on a 457 single CDNC value (200 $\rm cm^{-3}$) across the range of mountain heights and temperatures 458 to understand the responses to mountain height and temperature seen in Fig. 5. The 459 changing configuration of the orographic cloud with mountain height and temperature is 460 shown in Fig. 6. The orographic cloud produced in the C800m experiment is quite similar 461 to that of the reference simulation in terms of vertical extent (Fig. 6d). However, the 462 extent of the cloud and snow on the lee slope changes with cloud liquid ending closer to 463 the peak and the cloud ice/snow reaching farther down the slope. For the higher mountain 464 heights (Figs. 6b-c, e-f), the liquid cloud is shallower in the colder simulations while the 465 snow has a similar vertical extent. These higher mountain heights also produce more 466 ice/snow than the reference simulation, and in the cold temperature experiments, there is 467 more cloud ice/snow than liquid. Note that the 3000 m mountain wave response depends 468 on temperature, with the isotherms downstream of the mountain suggesting a stronger 469 downslope flow in the colder simulation. 470

Fig. 7 shows the isotopic composition of water vapor, cloud liquid and combined cloud ice/snow for the three mountain heights with the colder temperature profile ($T_{sfc} = 0^{\circ}$ C). Unlike in the reference simulation, these simulations have little rain, and its isotopic composition is not shown. As noted in section 2.3, the water vapor at the surface upwind of the mountain (Figs. 7a–c) is 8‰ more depleted than that of the reference simulation (Fig. 2a). The water vapor isotopic composition becomes increasingly asymmetric for the higher mountains due to rainout [*Smith et al.*, 2005], and the thin layer of downslope

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flow is visible in the water vapor isotopic composition for the 3000 m mountain (Fig. 7c). 478 The cloud liquid is almost entirely confined to the upstream side of the mountain, and its 479 isotopic content (Figs. 7d-f) is tied to the water vapor through the assumption of vapor-480 liquid isotopic equilibrium. The combined cloud ice/snow (Figs. 7g-i) is more depleted 481 than cloud liquid at the same altitude, and this difference increases with mountain height. 482 On the lee side of the mountain, the snow reaches to the base of the mountain in each case 483 and becomes more depleted with mountain height, as the snow has formed from vapor 484 that either originates at higher altitudes or has been depleted through precipitation. 485

As suggested by the sensitivity of total precipitation shown in Fig. 5, the distribution 486 of precipitation and its isotopic content across the mountain changes much more substan-487 tially with mountain height and temperature than with CDNC (Fig. 8). The precipitation 488 amount increases and shifts upstream with increasing mountain height, and $\delta^{18}O_{precip}$ on 489 the lee slope becomes more depleted with mountain height in agreement with the snow 490 isotopic composition shown in Figs. 7g–i. The lee slope difference in $\delta^{18}O_{precip}$ between 491 the 800 m and 3000 m mountain heights at x=315 km reaches 11% and 13% in the 492 warm and cold simulations, respectively. Similar differences are seen in the precipitation 493 integrated in the lee of the peak in Fig. 5d. The stronger dependence of $\delta^{18}O_{precip}$ on 494 mountain height in the cold simulations mirrors that seen in total precipitation and its 495 isotopic content in Fig. 5 and suggests that the isotopic lapse rate, the change in $\delta^{18}O_{precip}$ 496 with altitude, itself depends on temperature. 497

⁴⁹⁸ As in section 3.3, the relative contributions of different microphysical pathways to the ⁴⁹⁹ formation of precipitating hydrometeors are shown in Fig. 9b to understand better the ⁵⁰⁰ influence of mountain height and temperature on isotopic composition, which was seen

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⁵⁰¹ in Fig. 5. Most of the precipitation occurs windward of the peak in the sensitivity ⁵⁰² simulations, and thus the source terms plotted in Fig. 9b are similar to the breakdown of ⁵⁰³ upwind precipitation. A supplemental Fig. S2 shows the full breakdown of precipitation ⁵⁰⁴ sources by region as in Fig. 4.

The $\delta^{18}O_{precip}$ is the most enriched in the reference simulation (W800m) compared to 505 all other simulations (Fig. 9c), and this is also the case where the sources of riming and 506 accretion are largest and vapor deposition smallest. The contribution to precipitation from 507 riming decreases with increasing mountain height and decreasing temperature (Fig. 9b), 508 while the contribution of vapor deposition increases. Note that these contributions are 509 normalized by total precipitation, which itself increases with mountain height. While 510 there is considerable variation in the isotopic composition of the precipitation sources 511 with mountain height and temperature, this variation is systematic in the most important 512 contributors to precipitation: riming, vapor deposition and sublimation. As the mountain 513 height increases or the temperature falls, these processes form precipitating hydrometeors 514 from more depleted water vapor in the drier air found at colder temperatures and/or 515 further aloft. Despite the variation with mountain height and temperature seen in Fig. 9d, 516 a clear separation exists between the isotopic compositions contributed by riming and 517 vapor deposition to precipitation, and the shift towards the formation of snow by vapor 518 deposition at colder temperatures and higher mountains is reflected in the more depleted 519 isotopic compositions in those experiments. 520

It is evident, particularly in the cold temperature experiments, that precipitation source significantly influences the $\delta^{18}O_{precip}$ signal, and that the decreasing $\delta^{18}O_{precip}$ signal with increasing mountain height is not a simple reflection of temperature. The solid line

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in Fig. 5a-b represents the regression of the warm temperature experiments' domain-524 integrated precipitation and $\delta^{18}O_{precip}$. The dashed line in Figs. 5a–b is the same as the 525 solid line, but shifted down by 8‰, which represents the surface vapor δ^{18} O difference 526 between the warm and cold temperature profiles (see Figs. 2a and 7a). The cold 800 m 527 simulations fall on this dashed line in Fig. 5, but as the mountain height increases, the 528 $\delta^{18}O_{precip}$ values of the cold temperature simulations fall well below this line, implying that 529 precipitation is more depleted than what is expected from the 8% offset in the upwind 530 sounding in the cold 1500 m and 3000 m experiments. As noted above, this suggests that 531 the dependence of $\delta^{18}O_{precip}$ on altitude is itself a function of temperature. This can be 532 explained by the combination of three effects. First, the changing sources of precipitation 533 also contribute with a shift from riming to vapor deposition with decreasing tempera-534 ture. For the C3000m case, the domain-averaged $\delta^{18}O_{precip}$ is close to the $\delta^{18}O$ signatures 535 of vapor deposition itself. Second, the nonlinearity in the relationship between isotopic 536 composition and height plays a role here, as the gap between δ^{18} O for the warm and cold 537 simulations increases with height due to the curvature of the Rayleigh curve (Fig. S1). 538 Last, the changing structure of the mountain wave with mountain height and temperature 539 may also impact the distribution of precipitation and also its isotopic composition. 540

4. Discussion and Conclusions

Orographic precipitation is an important water resource, and in this work we have attempted to provide new perspective on how different atmospheric regimes may influence the formation of precipitation. The isotopic composition of orographic precipitation also provides additional information about the sources of water vapor and the microphysical processes that produce this precipitation. In the present study, the microphysical controls

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⁵⁴⁶ on the isotopic composition of wintertime orographic precipitation have been explored in ⁵⁴⁷ idealized simulations of flow over a two-dimensional mountain using an isotope-enabled ⁵⁴⁸ version of WRF. One reference simulation was performed along with sensitivity experi-⁵⁴⁹ ments that varied CDNC, temperature and mountain height to study the responses in the ⁵⁵⁰ microphysical processes, their respective isotopic composition and the $\delta^{18}O_{precip}$.

One of the main goals of this work has been to study the isotopic signatures of pre-551 cipitation and cloud microphysical processes and determine if there is a distinct isotopic 552 signal associated with those processes. With an idealized setup using different mountain 553 heights, warm and cold temperature profiles, and increasing CDNC, our simulations show 554 that there is a distinct difference in the δ^{18} O signatures of microphysical processes. The 555 $\delta^{18}O_{precip}$ reflects the relative contributions from each of the sources, and thus hydrome-556 teors that form from isotopically lighter sources lead to more depleted precipitation. In 557 all of the simulations, precipitation grows mainly by riming of cloud liquid, vapor de-558 position onto ice, or a combination of the two processes. The $\delta^{18}O$ difference between 559 riming and vapor deposition ranges between 3-8% in all simulations and is independent 560 of the environmental temperature. The distinct isotopic signals of the two sources persist 561 despite wide variation in the isotopic composition of these sources with mountain height 562 and temperature. This difference is related mainly to the altitude of the growth processes 563 within the cloud, as vapor deposition occurs both near the surface and in air with more 564 depleted water vapor aloft, and riming predominately happens near the mountain surface. 565 The sensitivity of $\delta^{18}O_{precip}$ to mountain height and temperature reflects, in part, the 566 changing sources of precipitating hydrometeors. The dominant source of precipitation 567 shifts from riming for smaller mountains and the warmer temperature profile to vapor 568

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deposition for higher mountains and colder temperatures. The more depleted isotopic 569 composition of the precipitating hydrometeors generated by vapor deposition contributes 570 to the decrease of $\delta^{18}O_{precip}$ with mountain height and temperature. It is notable that the 571 relationship between precipitation amount and $\delta^{18}O_{precip}$ driven by increasing mountain 572 height differs with temperature, and that the difference in $\delta^{18}O_{precip}$ between the warm and 573 cold simulations increases with mountain height. This suggests that the isotopic lapse rate 574 is itself a function of temperature, and that this temperature dependence partly results 575 in a shift in the microphysical pathways through which precipitating hydrometeors grow. 576 Additional factors that could also contribute to the temperature dependence of the isotopic 577 lapse rate include the nonlinearity of the Rayleigh curve and changes in the patterns of 578 airflow over the mountain. 579

We have attempted to illuminate how $\delta^{18}O_{precip}$ depends on the processes responsible 580 for the growth of precipitating hydrometeors. The decrease in $\delta^{18}O_{precip}$ with increasing 581 mountain height and colder temperature profiles is largely driven by the formation of hy-582 drometeors from more depleted water vapor in the drier air further aloft or at colder tem-583 peratures. However, the pathways through which precipitating hydrometeors are formed 584 also plays a role, as the more enriched precipitating hydrometers produced by riming 585 contribute less to surface precipitation and the more depleted hydrometeors produced 586 by vapor deposition onto ice contribute more. The weaker dependence of $\delta^{18}O_{precip}$ on 587 CDNC, where it exists, can be explained in a similar manner. While the weak dependence 588 on CDNC suggests a similarly weak dependence on aerosol concentrations, the domain-589 integrated signal in the strongest case is roughly equivalent to a 1°C shift in temperature 590 along the Rayleigh curve. Such a change might be visible in paleoclimate records of pre-591

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⁵⁹² cipitation if there were systematic shifts in aerosol concentrations on longer timescales. ⁵⁹³ However, if the majority of precipitation was produced in colder conditions, the sensitiv-⁵⁹⁴ ity to CDNC might not be visible, as is the case in the more strongly precipitating cases ⁵⁹⁵ here. While not considered here, mixed-phase orographic precipitation does respond to ⁵⁹⁶ changes in ice nuclei concentrations [e.g., *Fan et al.*, 2014], and the associated shifts in ⁵⁹⁷ microphysical processes could also impact the isotopic composition of precipitation.

The results have potential implications for research and field campaigns looking to study 598 the influence of different atmospheric regimes on orographic precipitation, such as IFRACS 599 (a 2014 campaign led by Doug Lowenthal, Gannet Haller and colleagues at the Desert 600 Research Institute: https://www.eol.ucar.edu/field_projects/ifracs), ISPA-III 601 [Ward and Cotton, 2011] and StormVEx [Mace et al., 2010]. As liquid processes are most 602 responsive to CDNC, locations where precipitation primarily forms through accretion of 603 cloud liquid and/or riming are likely to experience decreased accumulation, a shift in the 604 location of, and a decrease in the $\delta^{18}O_{precip}$. However, the sensitivity depends on both the 605 mountain height and the region above the mountain surface in which precipitation forms 606 and grows. The model could be beneficial to those planning observational campaigns in 607 terms of choosing locations to collect samples. For example, those interested in studying 608 the influence of aerosols on snowfall could identify the regions where precipitation is likely 609 to be most sensitive or least sensitive to aerosol loading. 610

In this idealized modeling study, we were able to distinguish isotopic signatures of the microphysical growth processes. As the climatology will vary between locations or even seasonally at one location, the model can be used to identify the isotopic signatures of microphysical processes in specific locations, which would help to determine growth

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pathways of measured precipitation. The next steps are to use the isotope-enabled microphysics scheme in a realistic setting to study snowfall events at Storm Peak Lab in Colorado observed during the Isotopic Fractionation in Snow (IFRACS) campaign. In this future work, we hope to expand upon our current research by studying the isotopic signatures of the microphysical growth processes that produce the observed precipitation.

Appendix A: Incorporating Isotopologues Into the Microphysics

Stable water isotopologues were added to the Thompson microphysics scheme in the 620 WRF model by duplicating all microphysical processes (e.g., freezing, melting, vapor de-621 position, evaporation) with additional process rates for the water isotopologues following 622 Blossey et al. [2010, App. B]. Except for the sublimation of ice, which is assumed to be 623 non-fractionating, all exchanges between vapor and condensate involve fractionation. The 624 fractionation/equilibration of water isotopologues from rain is included, along with the 625 fractionation of water vapor as it is deposited onto ice phase hydrometeors. Water vapor 626 and cloud liquid are assumed to be in isotopic equilibrium. While a detailed description 627 of the isotopic treatment including all of these processes can be found in appendix B of 628 Blossey et al. [2010], we give a brief summary below that emphasizes those processes that 629 play important roles in the cold and mixed-phase clouds central to this study. 630

For most processes, especially those in which whole hydrometeors are moved from one microphysical category to another (e.g., freezing of cloud droplets to form ice), the heavy isotopologues of water are transferred in proportion to their concentration in the source hydrometeor. For example, the freezing of cloud liquid droplets (wfz) to form cloud ice transfers heavy isotopologues to cloud ice as follows:

$$\left. \frac{dr'_i}{dt} \right|_{wfz} = \left. \frac{dr_i}{dt} \right|_{wfz} \mathcal{R}_c \tag{A1}$$

where $\mathcal{R}_c = r'_c/r_c$ is the isotopic ratio of cloud liquid, and r_c and r_i are the mass mixing ratios of cloud liquid and cloud ice, respectively. The mass mixing ratios of heavy isotopologues are denoted with primes, e.g. r'_c . Further, it is assumed that the isotopic ratio is uniform in each hydrometeor category, so that large and small raindrops have the same isotopic composition, for example. The latter assumption will not hold exactly in reality

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and is a source of error; the computation complexity of allowing such variation in a bulk scheme could be considerable.

For the few microphysical processes that result in fractionation (the unequal transfer 643 of heavy and light isotopologues between phases), those processes are represented as 644 described in *Blossey et al.* [2010, App. B]. In general, the lower vibrational energy of 645 the heavier isotopologues of water cause them to prefer the condensed phases (liquid, ice) 646 to the vapor phase, so that their concentrations in vapor are smaller than in the condensed 647 phases. When comparing concentrations of isotopologues, the words "heavier" or "more 648 enriched" are used to describe concentrations of heavy isotopologues that are higher, while 649 "lighter" or "more depleted" are used for smaller concentrations of heavy isotopologues. 650 We supply here a summary of how these processes might affect the isotopic composition 651 of water in mixed-phase clouds. 652

The efficient exchange between small liquid water droplets in clouds and the surrounding water vapor leads many microphysical schemes (including Thompson) to assume that incloud water vapor mixing ratios are equal to the saturation mixing ratio when cloud liquid is present. The complementary condition for heavy isotopologues is that the isotopic ratios of cloud liquid and water vapor are in isotopic equilibrium:

$$\mathcal{R}_c = \alpha_l \, \mathcal{R}_v \tag{A2}$$

where \mathcal{R}_v is the isotopic ratio of water vapor and α_l is the equilibrium fractionation coefficient for liquid [*Majoube*, 1971]. As the equilibration time for isotopic composition of small liquid water droplets is on the order of a few seconds [*Ciais and Jouzel*, 1994], this is in general a good assumption and is included in our implementation.

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Given the potentially large supersaturation with respect to ice, we need to consider the non-equilibrium processes driven by gradients of water vapor between the environment and ice particles which leads to vapor deposition onto those particles. The relatively smaller diffusivities of the heavy isotopologues modifies the transfer of water to the particle surface, so that the deposition of heavy isotopologues may be written as

$$\frac{dr'_i}{dt}_{dep} = \alpha_s \alpha_k \mathcal{R}_v \, \frac{dr_i}{dt}_{dep} \tag{A3}$$

⁶⁶⁷ [*Ciais and Jouzel*, 1994], where α_s is the equilibrium fractionation coefficient for ice [*Ma-*⁶⁶⁸ *joube*, 1970; *Merlivat and Nief*, 1967] and the kinetic fractionation coefficient, α_k , repre-⁶⁶⁹ sents the effects of the relative diffusion of the heavy and light isotopologues [*Jouzel and* ⁶⁷⁰ *Merlivat*, 1984].

The effects of these two processes on the isotopic composition of liquid and ice in 671 mixed-phase clouds is depicted in Fig. A1. Here, the variation in the saturation ratios 672 with respect to liquid and ice is depicted in the left panel as a function of temperature. 673 In keeping with the assumption in the microphysical scheme, the saturation ratio with 674 respect to liquid is one, while the ice saturation ratio grows with decreasing temperature. 675 The isotopic content of the vapor, cloud liquid and ice formed through vapor deposition 676 is shown in the right panel. Equations A2 and A3 have been used to compute isotopic 677 composition, except for cloud liquid water, whose value is fixed to the relationship observed 678 in mixed-phase orographic clouds by Lowenthal et al. [2011], 679

$$\delta^{18} \mathcal{O}_c = 0.9T - 10.12 \tag{A4}$$

where T is temperature in degrees Celsius. While ice formed through vapor deposition is more enriched than cloud liquid at the same temperature, close to 0°C, increasing the

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⁶⁶² supersaturation with respect to ice and decreasing temperature causes a stronger kinetic ⁶⁶³ effect during deposition onto cloud ice. Note that vapor deposition onto ice in a liquid ⁶⁶⁴ class is most efficient at colder temperatures, peaking close to -15°C [*Rogers and Yau*, ⁶⁸⁵ 1989, p. 161]. As a result, the typical isotopic composition of ice formed through vapor ⁶⁸⁶ deposition is often more depleted than that of cloud liquid closer to 0°C. Note that these ⁶⁸⁷ relationships will not hold once the liquid water is removed and the cloud is fully glaciated.

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Notes

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^{1.} Each simulation is labeled according to its temperature and mountain height. For example, W800m denotes the reference case with "W" denoting the warmer sounding with $T_{sfc} = 7^{\circ}C$ and, in other runs, "C" the colder sounding with $T_{sfc} = 0^{\circ}C$.

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Figure 1. Contoured temperature (black) and mixing ratios of cloud liquid water (shaded) and combined cloud ice/snow (contoured) for the 800 m warm temperature simulations, averaged over the last four hours of each simulation. Cloud droplet number concentrations of (a) 25 cm⁻³, (b) 200 cm⁻³, and (c) 800 cm⁻³ are shown to illustrate sensitivity to CDNC. Units are kg kg⁻¹ for hydrometeor mixing ratios and K for temperature.

Figure 2. Average over the last four hours of δ^{18} O of (a) vapor, (b) rain, (c) cloud liquid and (d) ice/snow for reference simulation with a CDNC of 200 cm⁻³.

Figure 3. (a) Precipitation accumulated over the 12 hours of the simulation and (b) the associated $\delta^{18}O_{precip}$ for the reference simulation and its sensitivity to CDNC changes. The mountain peak is located at 300 km.

Figure 4. Breakdown of (a) accumulated precipitation in the reference simulation over the whole domain and the regions upwind of the peak, over the peak and downwind of the peak, as well as (e) the respective average δ^{18} O values. The contributions from precipitation sources normalized by the total, domain-integrated precipitation in the three subregions are shown in (b–d) and the corresponding isotopic signatures in (e–g). The accumulated precipitation and its sources are integrated over the domain and normalized by the mountain half-width (20 km). Sources are: autoconversion of cloud liquid (laut), accretion of cloud liquid by rain (lacc), autoconversion of cloud ice (iaut), riming of cloud liquid (rim), vapor deposition onto ice (dep), and sublimation of ice/evaporation of rain (sub).

Figure 5. Scatter plot of area-integrated precipitation vs $\delta^{18}O_{precip}$ for all mountain heights, temperatures and CDNC values. These quantities are presented both for the whole domain (a) and for regions upstream of the peak (b), over the peak (c) and to the lee of the peak (d). For each experiment, the CDNC value is indicated by the color and size of the symbol, while the mountain height and temperature are shown by the shape of the symbol. The grey solid line in (a) and (b) is the regression of the domain averages in the warm temperature experiments and the dashed line is the solid line shifted down by 8‰. The regression is not shown in (c) and (d) because the isotopic composition over and downwind of the peak depends on the precipitation amount upstream.

Figure 6. As in figure 1, but for the 200 cm⁻³ simulations from the (a) 800 m, (b) 1500 m,
(c) 3000 m warm temperature experiments and (d) 800 m, (e) 1500 m, and (f) 3000 m cold temperature experiments.

Figure 7. Based on averages over the last four hours of each simulation, δ^{18} O of (top) vapor, (middle) cloud liquid and (bottom) ice/snow for the (a,d,g) 800 m, (b,e,h) 1500 m, and (c,f,i) 3000 m cold temperature experiments with a CDNC of 200 cm⁻³.

Figure 8. Distribution of precipitation accumulated over the 12 hour simulations in (a) warm experiments and (b) cold experiments. Corresponding $\delta^{18}O_{precip}$ for (c) warm experiments and (d) cold experiments. All profiles are based on the 200 cm⁻³ simulations. The mountain peak is located at 300 km. Note that the axis limits for $\delta^{18}O_{precip}$ have been shifted by 8 ‰ from the warm to cold simulations to account for the difference in the isotopic composition of vapor at the surface in the two cases.

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Case	CDNC	Total Prec	Snow	Graupel	Rain	Spillover
W800m	25	38.6mm	7.8%	7%	85.2%	37%
W800m	100	32.1mm	13.5%	12.9%	73.6%	59%
W800m	200	28.5mm	14.3%	13.7%	72%	56%
W800m	400	23.7mm	14.8 %	14.2%	70.8%	62%
W800m	800	18.3mm	13.8%	13%	73.1%	68%
C800m	25	34.2mm	92.2%	2.6%	5.2%	45%
C800m	100	32.2mm	96.3%	3%	0.7%	50%
C800m	200	30.9mm	96.9%	2.9%	0.2%	53%
C800m	400	29.3mm	97.5%	2.5%	_	56%
C800m	800	26.6mm	98.1%	1.9%	_	61%
W1500m	25	113.6mm	60.9%	7%	32.1%	31%
W1500m	100	110.5mm	64.1%	11%	24.9%	33%
W1500m	200	106.6mm	65.8%	13%	21.2%	35%
W1500m	400	104.8mm	67%	13.7%	19.3%	36%
W1500m	800	105.3mm	67.1%	14%	18.9%	37%
C1500m	25	91.3mm	96.1%	0.5%	3.4%	32%
C1500m	100	92.6mm	98.3%	0.6%	1.1%	33%
C1500m	200	90.7mm	98.7%	0.7%	0.6%	34%
C1500m	400	91.7 mm	99.1%	0.7%	0.2%	35%
C1500m	800	87.3mm	99.4%	0.6%		38%
W3000m	25	235.8mm	67.8%	3.2%	29%	27%
W3000m	100	237.8mm	69.1%	3.8%	27.1%	27%
W3000m	200	233.9mm	71%	4.3%	24.7%	28%
W3000m	400	232.1mm	72.1%	4.8%	23.1%	29%
W3000m	800	227.9mm	74%	5.3%	20.7%	31%
C3000m	25	182.3mm	97.3%	0.5%	2.2%	20%
C3000m	100	183.2mm	98.7%	0.3%	1%	21%
C3000m	200	166.7mm	99%	0.3%	0.7%	22%
C3000m	400	175.1mm	99.4%	0.3%	0.3%	22%
C3000m	800	172.2mm	99.6%	0.3%	0.03%	22%

Table 1: Breakdown of the major statistics for all runs. Columns indicate the case (height and cold (C) or warm (W) initial temperature profile), cloud droplet number concentration (CDNC) in cm^{-3} , normalized (by the mountain half-width) domain-integrated precipitation over 12 hour simulation, and the percent of snow, graupel and rain. Spillover is the ratio of the total leeward precipitation to the total mountain precipitation.

Figure 9. Breakdown of (a) total precipitation, snow and graupel and (c) corresponding δ^{18} O values for all experiments. The contributions from (b) precipitation sources normalized by the total precipitation and (d) the corresponding isotopic signatures for all experiments. Sources from left to right are: autoconversion of cloud liquid (LAUT), accretion of cloud liquid by rain (LACC), autoconversion of cloud ice (IAUT), accretion of cloud liquid (RIM), vapor deposition onto ice (DEP), and sublimation of ice/evaporation of rain (SUB). All values are based on the 200 cm⁻³ simulations.

Figure A1. [Left panel:] the saturation ratios with respect to cloud liquid (blue) and ice (green) as a function of temperature. [Right panel:] δ^{18} O of in-cloud water vapor (black), cloud liquid (blue) and vapor deposition onto ice particles (green) as a function of temperature.

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