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## Proportions of Convective and Stratiform Precipitation Revealed in Water Isotope Ratios

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1 Tropical and midlatitude precipitation is fundamentally of two types, spatially-limited and 2 high-intensity convective or widespread and lower-intensity stratiform, owing to 3 differences in vertical air motions and microphysical processes governing rain formation. 4 These processes are difficult to observe or model and precipitation partitioning into rain 5 types is critical for understanding how the water cycle responds to climate changes. Here, 6 we combine two independent data sets – convective and stratiform precipitation fractions, 7 derived from the Tropical Rainfall Measuring Mission satellite or synoptic cloud 8 observations, and stable isotope and tritium compositions of surface precipitation, derived 9 from a global network – to show that isotope ratios reflect rain type proportions and are 10 negatively correlated with stratiform fractions. Condensation and riming associated with 11 boundary layer moisture produces higher isotope ratios in convective rain, along with 12 higher tritium when riming in deep convection occurs with entrained air at higher 13 altitudes. Based on our data, stable isotope ratios can be used to monitor changes in the 14 character of precipitation in response to periodic variability or changes in climate. Our 15 results also provide observational constraints for an improved simulation of convection in 16 climate models and a better understanding of isotope variations in proxy archives, such as 17 speleothems and tropical ice.

19	Stable isotope ratios ( $\delta^{18}$ O, $\delta^{2}$ H; Methods) have long been observed to be different in
20	precipitation from different types of clouds, such as those producing convective showers versus
21	those producing frontal, continuous rain <sup>1-4</sup> . Recent studies <sup>5-8</sup> have observed lower $\delta^{18}$ O in
22	precipitation from stratiform clouds, in hydrologically more organized cloud systems, or when a
23	"bright band" occurs in vertical radar reflectivity. These observations have all been interpreted
24	using a Rayleigh distillation concept <sup>1-9</sup> , wherein adiabatic cooling of an air mass results in
25	successive condensation (rainout) with progressively lower $\delta^{18}$ O of precipitation at lower
26	temperatures in midlatitudes or increasing rain amount ('amount effect') in the tropics. Existing
27	interpretations of the 'amount effect' consider it to result from the low $\delta^{18}$ O of deep convective
28	rain forming at higher altitudes and a lack of isotopic exchange with near surface moisture due
29	to rapid fall velocities <sup>1-4</sup> , or from in-cloud re-evaporation of rain and isotopic exchange with
30	<sup>18</sup> O-depleted moisture at low altitudes <sup>5-6,9</sup> . Likewise, sub-cloud evaporation and isotopic
31	exchange with ambient moisture are described as accentuating the 'amount effect' by increasing
32	$\delta^{18}$ O when precipitation amounts are low, particularly under drier and warmer conditions <sup>1-9</sup> .
33	Although simulations of isotope distributions in global climate models (GCMs) differentiate
34	between convective and non-convective precipitation, their isotope schemes are based
35	essentially on different variations of the Rayleigh concept <sup>3,5,7,9-13</sup> . A comprehensive framework
36	that adequately explains observed isotope distributions in tropical and midlatitude precipitation,
37	however, is still lacking <sup>4</sup> .
38	A key assumption underpinning current interpretations of precipitation isotope
39	distribution is that a change in $\delta^{18}$ O of precipitation directly represents a change in one or more

41 temperature, rainout, or post-condensation isotope exchange. It is important to note that in spite 42 of potentially large differences in these parameters,  $\delta^{18}$ O of 50°N to 50°S precipitation normally

of the parameters that may influence the isotopic composition, such as moisture source,

43	has a range of ~ $-30$ to 0‰ ( <i>ref. 16</i> ), and most or all of this variability may be observed in
44	single or consecutive storm events <sup>1,6,8,14,15</sup> . Recently, precipitation $\delta^{18}$ O has been correlated
45	globally with a single climate parameter <sup>17</sup> (atmospheric moisture residence time), which reflects
46	the degree of hydrological organization or structures of precipitating clouds. Tropical and
47	midlatitude precipitation sampled on the Earth's surface almost always consists of varying
48	proportions of two fundamental rain types <sup>18-23</sup> – stratiform and convective – with significantly
49	different characteristics of temporal and spatial variability <sup>20-25</sup> arising from differences in cloud
50	vertical air motions and microphysical processes during hydrometeor formation, growth, and
51	descent to the surface. These differences in rain formation impart characteristic isotope
52	signatures (as we will discuss below) and changing proportions of convective and stratiform
53	rain types may primarily be responsible for precipitation isotope variability.

55

## Rain type fractions and stable isotope ratios

Figures 1a and 1b show mean monthly  $\delta^{18}$ O with respect to stratiform rain fractions and 56 57 precipitation amount from 28 tropical and two midlatitude locations (Table 1). We retrieved monthly stratiform fractions (1998-2014) from the TRMM Precipitation Radar 2A25 (version 7) 58 data product<sup>21,26,27</sup> and (for midlatitudes) synoptic cloud observations<sup>24</sup> (Methods). Monthly 59  $\delta^{18}$ O and precipitation amounts are from the IAEA/WMO global network of isotopes in 60 precipitation (GNIP) database<sup>16</sup> (Methods). The locations listed in Figure 1 span a broad 61 62 latitudinal range (50°N–21°S) and hydro-meteorological conditions: small islands in the Indian, 63 Pacific, or Atlantic oceans, coastal or inland continental, sea-level to ~3000 m altitude, and low 64 to high, mean annual precipitation amounts (~166–5000 mm) or surface air temperatures (~7– 65 29°C).

66	The $\delta^{18}$ O increases with decreasing stratiform fractions (Fig. 1a) and a regression
67	including all locations indicates a strong negative correlation ( $R^2 \approx 0.6$ ; p<0.0001). This
68	correlation probably would be stronger, if not for somewhat independent variability and
69	uncertainty in rain fraction and isotope data. TRMM satellite coverage is limited to 0-2 swaths
70	per day in a 2.5° grid and may include only a few of the precipitation days in a month <sup><math>21,26,27</math></sup> .
71	Some of this variability is reduced by taking climatological means, but probably not in relation
72	to $\delta^{18}$ O because of the different spatial scales (point location versus a 2.5° grid cell) of the
73	isotope versus TRMM data. The uncertainty in estimated stratiform fraction is also greater
74	when shallow rain is significant and when rain amounts are low <sup>21,27</sup> . Rain types based on cloud
75	observations may overestimate the stratiform fractions (Methods). Additionally, isotope data
76	can be biased towards that of stratiform rain because a fixed location for isotope sampling may
77	not experience all convective rainfall events owing to the limited spatial extents of convective
78	clouds.

79 $\Box \Box \Box \delta^{18}$ O – precipitation amount correlation is quite variable (Fig. 1b), even though80at each tropical location there is a general trend of decreasing  $\delta^{18}$ O with increasing precipitation.81Stratiform fractions and precipitation amount (Fig. 1c) show an opposite correlation compared82to  $\delta^{18}$ O and even low amounts of precipitation have low  $\delta^{18}$ O when the stratiform fraction is83high. More importantly, a significant increase in tropical monthly precipitation (~400–900 mm)84occurs with a limited  $\delta^{18}$ O variability that is consistent with the narrow range in stratiform85fractions.

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### **Convective – stratiform isotope differences**

88 The low and high  $\delta^{18}$ O, respectively, of stratiform and convective rain are consistent 89 with dynamical and microphysical conditions of rain formation (Fig. 2). In stratiform clouds

(nimbostratus), vertical air motions are weak<sup>18-23</sup> with mean upward air velocity (~ $0.2 \text{ m s}^{-1}$ ) 90 91 much lower than typical ice and snow terminal fall velocity (~1 m s<sup>-1</sup>). Condensation nuclei (ice 92 particles) form or are introduced near cloud tops and grow, initially by vapor deposition (diffusion) and later by aggregation, while falling slowly towards the surface<sup>18-23</sup>. Below the 93 freezing level, melting occurs in an  $\sim 500$  m thick layer<sup>18,19</sup> ( 'bright band' observed in vertical 94 95 radar reflectivity profiles) and as raindrops fall further, they may partially evaporate, under conditions of subsidence, or grow by accretion and coalescence, under conditions of uplift<sup>18,19</sup>. 96 97 The time for condensation, growth and rainfall for stratiform precipitation is about 1-3 hours<sup>19</sup>.

98 Hydrometeor growth by vapor diffusion above the freezing level, where tropospheric 99 moisture would have a  $\delta^{18}$ O of ~ -50 to -40‰ (*ref.* 28), isotopic homogenization in the melting layer, and equilibration with lower altitude moisture<sup>29</sup> as relatively small rain drops<sup>19</sup> (on 100 average ~1 mm in diameter) fall slowly to the surface, all would result in low  $\delta^{18}$ O of stratiform 101 102 rain. Because of the isotopic exchange below the melting level, near-surface temperature would have a greater influence, resulting in a range of  $\delta^{18}$ O. When the melting level is near or at the 103 104 ground surface (~1 km or lower), as in midlatitude winters, limited or negligible exchange with ambient moisture<sup>3,9,10,28,29</sup> would produce relatively more negative  $\delta^{18}$ O of stratiform 105 106 precipitation.

107In contrast, condensation particles in convective clouds (cumulus or cumulonimbus)108form near the cloud base<sup>18-23</sup> and grow as they are lifted in strong updrafts  $(1-10 \text{ m s}^{-1})$ . The109time from initiation of condensation to rainfall in a single convective cell may be as little as110thirty minutes<sup>19</sup>. When convection is shallow, precipitation forms by 'warm' processes<sup>19-21</sup>,111wherein condensation occurs below the freezing level and rain drops grow by collision and112coalescence. In deep convection, condensation nuclei form as ice particles just above the113freezing level, and grow rapidly in updrafts by riming as super-cooled water (mostly from the

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114planetary boundary layer) is frozen at successively higher altitudes<sup>19-23</sup>. Entrainment of115surrounding air above the freezing level may also contribute minor proportions of moisture for116riming growth in convective updrafts<sup>19</sup>. As particles become large enough (e.g. graupel) to117overcome upward air motion, they descend in strong downdrafts, melt rapidly below the 0°C118level, as opposed to a 'melting layer' in stratiform precipitation, and fall as large rain drops (>2119mm diameter) without significant evaporation or growth<sup>19-23</sup>.

Convective updrafts are fed by boundary layer moisture, with  $\delta^{18}$ O similar to that of 120 121 ocean evaporation ( $\sim -12$  to -10%), particularly in oceanic or coastal locations, and slightly 122 lower more inland depending upon the contribution from evaporated soil moisture<sup>4</sup>. The liquidvapor isotope fractionation<sup>4,9</sup> is  $\sim 10.7 - 9.8\%$  (0 to 10°C) and shallow convective or 'warm' 123 rain would likely have a  $\delta^{18}$ O that is about  $-1\pm 1$  %. In deep convective precipitation, ice 124 particles from boundary layer moisture just above the freezing level would have  $\delta^{18}$ O of ~ 0‰ 125 or higher as ice-vapor isotope fractionation<sup>9</sup> (at 0 to  $-10^{\circ}$ C) is ~ 4‰ greater than that for liquid-126 127 vapor. In addition, depending upon the nature of ice particle surfaces, the kinetic isotope fractionation between ice-vapor or ice-liquid could be even larger<sup>30</sup>. As ice particles are lifted in 128 129 updrafts, further accretion (riming) of supercooled boundary layer moisture would also result in relatively higher  $\delta^{18}$ O. Entrained environmental air at higher altitudes may have a low  $\delta^{18}$ O, but 130 because of its low moisture content, it is unlikely to significantly decrease the  $\delta^{18}$ O of 131 132 hydrometeors<sup>31</sup>. Ice particles or graupel falling in downdrafts will melt beneath the 0°C level and the large size and fall velocity<sup>19</sup> (~5 to 10 m s<sup>-1</sup>) of the rain drops will inhibit isotopic 133 exchange with ambient moisture<sup>29,31</sup>, preserving the high  $\delta^{18}$ O acquired during ice and riming 134 growth in updrafts. This is consistent with precipitation tritium (<sup>3</sup>H) contents (see below). 135

136

## 137 **Tritium in convective rain**

138	Differences in ice formation, growth, and melting in convective and stratiform clouds
139	resulting in different $\delta^{18}$ O are evident also in the maximum height of high reflectivity, 40-dBZ
140	echo recorded by TRMM precipitation radar <sup>19,32,33</sup> . High reflectivity indicates large ice particles
141	(i.e., graupel) above the 0°C level (~5 km in the tropics) lifted in strong updrafts, a melting
142	layer of ice particles just below the 0°C level exhibiting a bright band characteristic of
143	stratiform clouds, or intense rain beneath the 0°C level as seen commonly over the oceans <sup>19,32</sup> .
144	Figures 3a and 3b show the $\delta^{18}$ O and stratiform rain fractions for tropical stations with respect
145	to the 40-dBZ echo height. We could not access similar data for Vienna and Krakow because
146	they are northward of the TRMM satellite orbital extent. High $\delta^{18}$ O with low stratiform
147	fractions occurs with echo heights >5 km (i.e., deep convection) or $<\sim1$ km (warm rain). For
148	higher stratiform fractions (>~30%), a wider range of lower $\delta^{18}$ O occurs with the 40-dBZ echo
149	at ~2-4 km.
150	Figures 4a and 4b show <sup>3</sup> H for selected tropical and midlatitude locations with respect to
150 151	Figures 4a and 4b show <sup>3</sup> H for selected tropical and midlatitude locations with respect to stratiform fractions and $\delta^{18}$ O. Tritium contents are also derived from the GNIP database <sup>16</sup> and
151	stratiform fractions and $\delta^{18}$ O. Tritium contents are also derived from the GNIP database <sup>16</sup> and
151 152	stratiform fractions and $\delta^{18}$ O. Tritium contents are also derived from the GNIP database <sup>16</sup> and are expressed in tritium units (1TU = 10 <sup>-18</sup> <sup>3</sup> H/H; Methods). As noted earlier, riming growth in
151 152 153	stratiform fractions and $\delta^{18}$ O. Tritium contents are also derived from the GNIP database <sup>16</sup> and are expressed in tritium units (1TU = 10 <sup>-18</sup> <sup>3</sup> H/H; Methods). As noted earlier, riming growth in deep convective updrafts may include entrained moisture at higher altitudes (~6-8 km) where
151 152 153 154	stratiform fractions and $\delta^{18}$ O. Tritium contents are also derived from the GNIP database <sup>16</sup> and are expressed in tritium units (1TU = 10 <sup>-18</sup> <sup>3</sup> H/H; Methods). As noted earlier, riming growth in deep convective updrafts may include entrained moisture at higher altitudes (~6-8 km) where <sup>3</sup> H contents are expected to be very high <sup>29,34</sup> (>~10 <sup>4</sup> TU) compared to those near surface
151 152 153 154 155	stratiform fractions and $\delta^{18}$ O. Tritium contents are also derived from the GNIP database <sup>16</sup> and are expressed in tritium units (1TU = 10 <sup>-18</sup> <sup>3</sup> H/H; Methods). As noted earlier, riming growth in deep convective updrafts may include entrained moisture at higher altitudes (~6-8 km) where <sup>3</sup> H contents are expected to be very high <sup>29,34</sup> (>~10 <sup>4</sup> TU) compared to those near surface (presently ~5 TU or less). Even minor amounts of this high altitude moisture in hydrometeors
151 152 153 154 155 156	stratiform fractions and $\delta^{18}$ O. Tritium contents are also derived from the GNIP database <sup>16</sup> and are expressed in tritium units (1TU = 10 <sup>-18</sup> <sup>3</sup> H/H; Methods). As noted earlier, riming growth in deep convective updrafts may include entrained moisture at higher altitudes (~6-8 km) where <sup>3</sup> H contents are expected to be very high <sup>29,34</sup> (>~10 <sup>4</sup> TU) compared to those near surface (presently ~5 TU or less). Even minor amounts of this high altitude moisture in hydrometeors (~5% of total water budget) would substantially increase precipitation <sup>3</sup> H contents <sup>23</sup> , resulting in
151 152 153 154 155 156 157	stratiform fractions and $\delta^{18}$ O. Tritium contents are also derived from the GNIP database <sup>16</sup> and are expressed in tritium units (1TU = 10 <sup>-18</sup> <sup>3</sup> H/H; Methods). As noted earlier, riming growth in deep convective updrafts may include entrained moisture at higher altitudes (~6-8 km) where <sup>3</sup> H contents are expected to be very high <sup>29,34</sup> (>~10 <sup>4</sup> TU) compared to those near surface (presently ~5 TU or less). Even minor amounts of this high altitude moisture in hydrometeors (~5% of total water budget) would substantially increase precipitation <sup>3</sup> H contents <sup>23</sup> , resulting in a positive <sup>3</sup> H – $\delta^{18}$ O correlation. Isotopic exchange and equilibration at lower altitudes (as in
151 152 153 154 155 156 157 158	stratiform fractions and $\delta^{18}$ O. Tritium contents are also derived from the GNIP database <sup>16</sup> and are expressed in tritium units (1TU = 10 <sup>-18</sup> <sup>3</sup> H/H; Methods). As noted earlier, riming growth in deep convective updrafts may include entrained moisture at higher altitudes (~6-8 km) where <sup>3</sup> H contents are expected to be very high <sup>29,34</sup> (>~10 <sup>4</sup> TU) compared to those near surface (presently ~5 TU or less). Even minor amounts of this high altitude moisture in hydrometeors (~5% of total water budget) would substantially increase precipitation <sup>3</sup> H contents <sup>23</sup> , resulting in a positive <sup>3</sup> H – $\delta^{18}$ O correlation. Isotopic exchange and equilibration at lower altitudes (as in stratiform rain) will decrease precipitation <sup>3</sup> H contents to ambient levels <sup>29,34</sup> .

- 162 (Figs. 4a, 4b), indicating that higher  $\delta^{18}$ O did not result from isotopic exchange or in-cloud rain 163 re-evaporation at lower altitudes. Lower <sup>3</sup>H and a lack of <sup>3</sup>H –  $\delta^{18}$ O correlation at Bogota, 164 Bangkok and Hong Kong (Figs. 4a, 4b) are consistent with lower heights of deep convection 165 and higher stratiform fractions (Fig. 3) at these locations.
- 166

## 167 Isotope variability and character of precipitation

168 Variable convective and stratiform fractions are an integral feature of precipitating clouds in the tropics and midlatitudes<sup>18-24</sup> where mesoscale convective systems and extra-169 tropical cyclones produce most of the precipitation<sup>19</sup>. Average annual stratiform fractions (on a 170 volume basis) are ~35-55% in the tropics<sup>21</sup> and are higher in the midlatitudes<sup>19,35</sup>. Smaller, less 171 172 organized convective systems have much lower stratiform fractions. Substantial variability in 173 rain type fractions ranging from near zero to nearly all stratiform rain may occur during storm events both in the tropics and midlatitudes<sup>1,6,8,15</sup>. Complex patterns<sup>4-8</sup> of precipitation  $\delta^{18}$ O 174 decreasing, increasing, and then decreasing again with time are commonly observed<sup>4-8</sup> during 175 176 storm events and may easily result from successive storms with stratiform precipitation trailing convective<sup>19,36</sup>. A progressive decrease, increase or no change in  $\delta^{18}$ O with time<sup>5,6,14,15</sup> may 177 occur when stratiform precipitation trails, leads, or overlaps convective precipitation<sup>36</sup>. 178

179The  $\delta^{18}$ O – stratiform fraction correlation shown in Figure 1 for monthly data is found180also at storm event or interannual time scales. Recent measurements of isotope ratios in the181equatorial Indian Ocean<sup>4,15</sup> and in California, USA<sup>8</sup> both indicate that  $\delta^{18}$ O is low (~ -20‰)182when precipitation is dominantly stratiform and high (~ 0‰) when it is mostly convective.183Significant interannual variability occurs in response to regional or global phenomenon, such as184the El Niño–Southern Oscillation (ENSO)<sup>37</sup>, and the character of precipitation is affected185differently across the Pacific<sup>21</sup>, with lower stratiform fractions in the west (e.g. Darwin, 130°E)

186	and higher in the east (e.g. Bellavista, 90°W). These opposite trends are seen in the mean annual
187	isotope ratios for 1998, a strong El Niño year (positive ENSO index), compared to average
188	values for 1999-2001. At Darwin, 1998 precipitation had lower stratiform fractions (29%;
189	average 33%) and higher $\delta^{18}$ O (-4.9‰; average -6.0‰), while at Bellavista, fractions were
190	higher (49%; average 30%) and $\delta^{18}$ O lower (-5.5%; average -2.9%).
191	The negative correlation between $\delta^{18}$ O and stratiform rain fractions (Fig. 1a) is also
192	consistent with the previously observed positive correlation between $\delta^{18}$ O and moisture
193	residence time <sup>17</sup> because residence time would be lower, and stratiform fractions higher, in
194	well-organized, mesoscale convective systems and extra-tropical cyclones <sup>19,21</sup> .
195	We conclude that $\delta^{18}$ O in tropical and midlatitude precipitation reflects the proportions
196	of convective and stratiform rain. Routine measurements of stable isotopes and tritium in sub-
197	daily or daily precipitation will help to better understand short-term variability in cloud
198	dynamics responsible for changes in rain fractions. In addition, isotope data could be used to
199	evaluate the relative importance of 'warm' process rain as a contribution to total precipitation,
200	as well as an independent, diagnostic tool to monitor variability in, and climate change impacts
201	on, the character of precipitation <sup>38,39</sup> .
202	Precipitation partitioning based on isotope distributions should help to improve climate
203	models <sup>25</sup> and the simulation of convection by providing observational constraints for
204	tropospheric heating profiles and rain types <sup>18-22,41</sup> . Isotope schemes in GCMs for convective and
205	stratiform precipitation that are consistent with microphysical processes depicted in Figure 2
206	should lead to model results being robust across temporal and spatial scales. Our results also
207	suggest that, similar to variations in modern precipitation, annual average $\delta^{18}$ O recorded in
208	proxy archives of paleo-precipitation <sup>41</sup> , such as speleothems and tropical ice, should reflect a
209	change in the proportions of convective and stratiform precipitation. Lower or higher $\delta^{18}$ O, for

210		example, may result from higher or lower stratiform fractions, which may or may not coincide
211		with wetter or drier conditions (Fig. 1). A re-examination of proxy isotope data in this light
212		should facilitate the development of more reliable models of past and future climate change
213		impacts on the water cycle.
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346 Author contributions: PKA initiated the project and wrote the paper; UR, DB, CS and AF provided

347 TRMM data; LAA processed isotope and related data; PB contributed cloud-based stratiform fractions;

348 all authors contributed to data evaluation and commented on the manuscript.

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350 **Competing interests:** There are no competing interests.

## 351 METHODS

352 Stratiform fractions

Tropical locations: Stratiform rain fractions were calculated using NASA's TRMM PR 2A23 (rain 353 354 type classification) and 2A25 (attenuation correction) orbital data products for 1998-2014. For each 355 major rain type (stratiform, deep convective, shallow convective), radar reflectivity data was binned by 356 latitude, longitude and height with a reflectivity bin spacing of 2 dBZ and spatial bin spacing of 2.5° by 357 2.5° from 35°N to 35°S. Rainfall was estimated at each grid point using PR data measured  $\sim 2$  km above mean ground level height. Stratiform and convective conditional rain rates were estimated from 358 mean radar reflectivity ( $\geq$  18 dBZ) using 2A25 V7 initial stratiform and convective Z-R relations<sup>27</sup>. 359 360 Unconditional rain rates could then be estimated using the probability of rainfall at each grid point 361 based on TRMM PR observations. Stratiform rainfall fraction here is defined as the ratio of stratiform 362 volumetric rainfall to total volumetric rainfall or the ratio of unconditional rain rates. 363 In TRMM rain type classification, rain is either stratiform or convective. The "other" pixels that the algorithm predicts are raining are very small in count (1.7% of all raining pixels) and likely don't 364

365 contribute much to rainfall near the surface; this rain appears to mostly exist at higher levels and has an
anvil appearance<sup>27</sup>. These pixels are not used in our stratiform-convective rain calculations. Therefore,
for our purposes here, 0% stratiform is essentially 100% convective with the exception of 'no-rain'
368 cases.

369 *Midlatitude locations:* For Vienna and Krakow, average monthly stratiform fractions for the period 370 1998-2014 were calculated from 3-hourly synoptic cloud observations in the MIDAS data base<sup>24</sup>: (http://badc.nerc.ac.uk/view/badc.nerc.ac.uk\_ATOM\_dataent\_ukmo-midas). Precipitation is 371 372 classified as convective when cumulus or cumulonimbus type clouds are present, and as stratiform 373 when stratus or nimbostratus are present. Conditions with mixed cloud types are separated in the 374 statistics. A day-night bias in the synoptic data could potentially affect the results. The stratiform 375 fraction is deemed more reliable and is derived as the amount of precipitation classified as stratiform 376 divided by total precipitation. Sensitivity studies with convective fraction derived as the ratio of the 377 amount of convective precipitation to total precipitation yield similar result. We note, however, that the 378 sum of convective and stratiform fractions generally is much less than 100% for many samples, giving 379 rise to a greater uncertainty in estimated fractions.

380

#### 381 Stable isotopes, precipitation amount and tritium

Stable isotope abundances in precipitation are expressed as  $\delta^{18}$ O or  $\delta^{2}$ H = (Rx/Rstd -1)\*1000, where R is the <sup>18</sup>O/<sup>16</sup>O or <sup>2</sup>H/H ratio in a sample (x) or standard (std) and the isotopic standard is VSMOW. Our analysis uses only oxygen isotope ratios ( $\delta^{18}$ O) because hydrogen isotope ratios are proportional to those of oxygen<sup>1,2</sup>.

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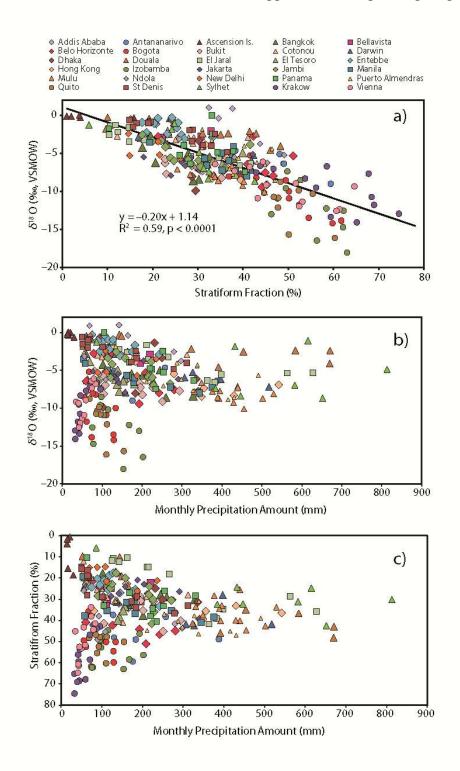
locations (Vienna and Krakow) were retrieved from the GNIP database. Monthly means were used to
calculate long-term arithmetic means (unweighted) for months with more than 50 mm precipitation
(Table 1), except for Ascension Island, Vienna and Krakow, where all months with consistent data over
multiple years were used.

Tritium (<sup>3</sup>H) is reported in Tritium Units (TU) where  $1 \text{ TU} = 10^{-18} \text{ }^{3}\text{H/H}$ . Analyses of water 397 398 samples for tritium is carried out using ultra-low level liquid scintillation counters after electrolytical 399 enrichment (or concentration) of tritium from a large sample by electrolysis. Current analytical 400 methods allow a typical uncertainty in tritium analysis of about  $\pm 0.1$  to  $\pm 0.5$  TU. Tritium is produced in 401 the stratosphere by cosmic ray spallation and reaches the troposphere by moisture 'leakage' or exchange and its abundance decreases towards the Earth's surface<sup>20,26</sup>. In addition, a latitudinal 402 403 gradient (decreasing <sup>3</sup>H contents towards the equator) exists due to the fact that stratospheric exchange occurs mostly in mid to high latitudes<sup>20,21</sup>. Stratospheric <sup>3</sup>H injection by thermo-nuclear bomb tests 404 405 before 1963 swamped the cosmogenic production and tropospheric tritium increased by orders of 406 magnitude. This 'bomb' tritium was gradually washed out of the stratosphere and troposphere by the 1990s, but vertical differences in tropospheric <sup>3</sup>H contents remained. 407

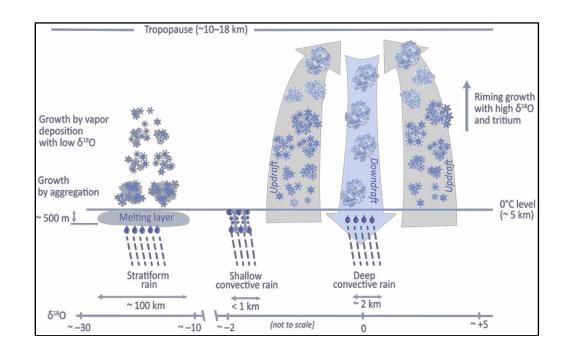
408 Precipitation <sup>3</sup>H contents were also retrieved from the GNIP database for 1998–2012, except for New Delhi where data were available for 1964–1978. During this period, atmospheric <sup>3</sup>H was declining 409 410 rapidly due to the wash out of 'bomb' tritium<sup>34</sup>. As a result, absolute <sup>3</sup>H values for New Delhi cannot be compared directly with other locations. The  ${}^{3}H - \delta {}^{18}O$  correlations for New Delhi shown in Fig. 4b 411 412 are based on data for both isotopes from the same time period (1964-1978), but are shown for smaller 413 time intervals after normalization by the average for each period so that the correlation is not masked by declining concentrations. The lower <sup>3</sup>H at Dhaka compared to Vienna or Krakow reflects the 414 415 latitudinal gradient in tropospheric <sup>3</sup>H concentration because the stratosphere-troposphere exchange

- 416 that brings tritium to the troposphere occurs mostly at higher latitudes<sup>29,34</sup>, as well as differences in the
- 417 height of convective entrainment.
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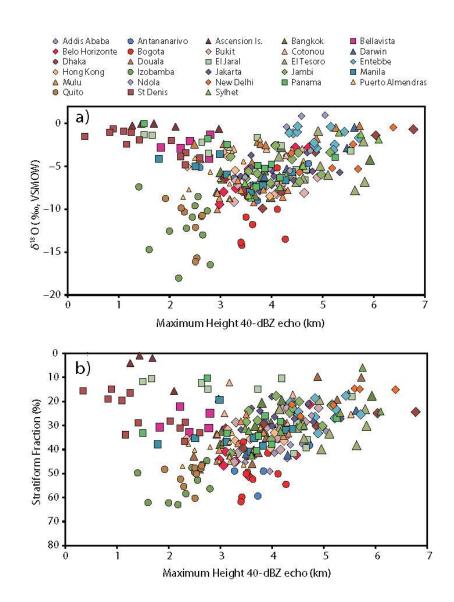
421 **Figure 1.** Correlation of mean monthly  $\delta^{18}$ O, stratiform fraction and precipitation amount in tropical 422 and midlatitude precipitation. The remainder of the rain fraction is convective (Methods). Solid line in 423 panel a) is based on a linear regression of all data points shown. The vertical axis in c) is shown in 424 reverse scale and trends in b) and c) are similar but opposite with respect to precipitation amount



- **Figure 2.** Schematic representation of differences in dynamical and microphysical processes in
- 427 convective and stratiform precipitation resulting in isotope variations. (Adapted after Houze Jr.<sup>18,19</sup>)



- 434 **Figure 3**. Correlation of mean monthly  $\delta^{18}$ O and stratiform fractions with the maximum height of 40-
- 435 dBZ echo. Precipitation with low stratiform fractions (or high convective fractions) has higher  $\delta^{18}$ O but
- 436 low or high echo heights reflecting shallow (warm rain) or deep convection.
- 437



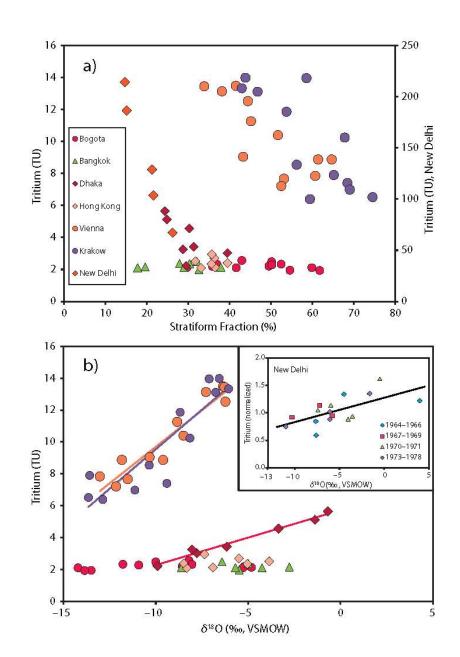
440 **Figure 4.** Correlation of monthly precipitation tritium (<sup>3</sup>H) content with stratiform fractions and  $\delta^{18}$ O.

In 4a, note the much higher tritium at New Delhi because 1964-1978 data are used, while 2000-2012

data are used for the other stations. Tritium values in 4b-inset are normalized by the mean annual

443 average for each period (Methods). The New Delhi regression line has an  $R^2$  of 0.42 with p<0.001. 444 Other regression lines have  $R^2$  of 0.81 (Krakow), 0.90 (Vienna), and 0.98 (Dhaka), all with p<0.00001.

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Fable 1. Geographica	l coordinates	and related	information	for locations	used in this study.
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				Alt.	Average Annual		Months with
Code	Location	Lat.	Long.	(m)	Precip (mm)	T (°C)	Precip > 50 mm*
6345000	Addis Ababa	9.00	38.73	2360	954	16.7	May-Sept
6708301	Antananarivo	-18.91	47.56	1295	1263	19.4	Jan-Apr; Nov-Dec
6190000	Ascension Is.	-7.92	-14.42	15	166	25.8	Mar-Aug
4845500	Bangkok	13.73	100.50	2	1661	28.5	Mar-Nov
8400101	Bellavista	-0.69	-90.33	194	763	22.5	Jan-May
8358301	Belo Horizonte	-19.87	-43.97	857	1190	22.5	Jan-Apr; Oct-Dec
8022200	Bogota	4.70	-74.13	2547	1029	13.3	Feb-Dec
9616301	Bukit (Sumatra)	-0.20	100.32	865	2005	21.9	Jan-Dec
6534403	Cotonou	6.42	2.33	14	1084	27.1	Mar-Jul; Sep-Nov
9412000	Darwin	-12.43	130.87	26	1902	27.3	Jan-Apr; Oct-Dec
4192300	Dhaka	23.95	90.28	14	1545	26.7	Apr-Oct
6491001	Douala	4.04	9.73	18	3787	27.2	Feb-Dec
7870802	El Jaral	14.94	-88.02	652	3586	22.6	Jan-Dec
8004401	El Tesoro	9.34	-75.29	175	1299	27.4	Mar-Dec
6370500	Entebbe	0.05	32.45	1155	1575	21.7	Jan-Dec
4500400	Hong Kong	22.32	114.17	66	2221	23.0	Apr-Oct
8404400	Izobamba	-0.37	-78.55	3058	1380	11.7	Jan-Jun; Aug-Dec
9674503	Jakarta	-6.29	106.67	45	1945	26.9	Jan-Sep; Nov-Dec
9619500	Jambi	-1.63	103.65	25	2126	26.9	Jan-Dec
9842900	Manila	14.52	121.00	14	1737	27.3	Apr-Dec
9644901	Mulu (Sarawak)	4.05	114.81	24	5026	27.0	Jan-Dec
6756100	Ndola	-13.00	28.65	1331	347	20.6	Jan-Feb; Nov-Dec
4218200	New Delhi	28.58	77.20	212	802	25.3	May-Oct
7880602	Panama	8.98	-79.53	5	1853	27.1	Apr-Dec
8437701	Puerto Almendras	-3.82	-73.38	98	3703	26.1	Jan-Dec
8407301	Quito	-0.17	-78.48	2789	808	15.2	Jan-May; Oct-Dec
6198001	St Denis	-20.90	55.48	70	1599	24.7	Jan-Dec
4190000	Sylhet	24.91	91.85	20	4012	25.7	Mar-Oct
1256500	Krakow	50.06	19.85	205	668	8.3	Jan-Dec
1103500	Vienna	48.25	16.36	198	684	10.3	Jan-Dec

451 \* except for Ascension Island, Vienna and Krakow, where all months with consistent data over multiple years
 452 were included.