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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 ($\delta^{18}\text{O}$, $\delta^2\text{H}$; 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 rain¹⁻⁴. Recent studies⁵⁻⁸ have observed lower $\delta^{18}\text{O}$ 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 concept¹⁻⁹, wherein adiabatic cooling of an air mass results in
25 successive condensation (rainout) with progressively lower $\delta^{18}\text{O}$ 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 $\delta^{18}\text{O}$ of deep convective
28 rain forming at higher altitudes and a lack of isotopic exchange with near surface moisture due
29 to rapid fall velocities¹⁻⁴, or from in-cloud re-evaporation of rain and isotopic exchange with
30 ^{18}O -depleted moisture at low altitudes^{5-6,9}. Likewise, sub-cloud evaporation and isotopic
31 exchange with ambient moisture are described as accentuating the ‘amount effect’ by increasing
32 $\delta^{18}\text{O}$ when precipitation amounts are low, particularly under drier and warmer conditions¹⁻⁹.
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^{3,5,7,9-13}. A comprehensive framework
36 that adequately explains observed isotope distributions in tropical and midlatitude precipitation,
37 however, is still lacking⁴.

38 A key assumption underpinning current interpretations of precipitation isotope
39 distribution is that a change in $\delta^{18}\text{O}$ of precipitation directly represents a change in one or more
40 of the parameters that may influence the isotopic composition, such as moisture source,
41 temperature, rainout, or post-condensation isotope exchange. It is important to note that in spite
42 of potentially large differences in these parameters, $\delta^{18}\text{O}$ of 50°N to 50°S precipitation normally

43 has a range of ~ -30 to 0% (*ref. 16*), and most or all of this variability may be observed in
44 single or consecutive storm events^{1,6,8,14,15}. Recently, precipitation $\delta^{18}\text{O}$ has been correlated
45 globally with a single climate parameter¹⁷ (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 types¹⁸⁻²³ – stratiform and convective – with significantly
49 different characteristics of temporal and spatial variability²⁰⁻²⁵ 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.

54

55 **Rain type fractions and stable isotope ratios**

56 Figures 1a and 1b show mean monthly $\delta^{18}\text{O}$ with respect to stratiform rain fractions and
57 precipitation amount from 28 tropical and two midlatitude locations (Table 1). We retrieved
58 monthly stratiform fractions (1998-2014) from the TRMM Precipitation Radar 2A25 (version 7)
59 data product^{21,26,27} and (for midlatitudes) synoptic cloud observations²⁴ (Methods). Monthly
60 $\delta^{18}\text{O}$ and precipitation amounts are from the IAEA/WMO global network of isotopes in
61 precipitation (GNIP) database¹⁶ (Methods). The locations listed in Figure 1 span a broad
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 $\delta^{18}\text{O}$ 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^{21,26,27}.
71 Some of this variability is reduced by taking climatological means, but probably not in relation
72 to $\delta^{18}\text{O}$ 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^{21,27}. 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 $\delta^{18}\text{O}$ – precipitation amount correlation is quite variable (Fig. 1b), even though
80 at each tropical location there is a general trend of decreasing $\delta^{18}\text{O}$ with increasing precipitation.
81 Stratiform fractions and precipitation amount (Fig. 1c) show an opposite correlation compared
82 to $\delta^{18}\text{O}$ and even low amounts of precipitation have low $\delta^{18}\text{O}$ when the stratiform fraction is
83 high. More importantly, a significant increase in tropical monthly precipitation (~400–900 mm)
84 occurs with a limited $\delta^{18}\text{O}$ variability that is consistent with the narrow range in stratiform
85 fractions.

86

87 **Convective – stratiform isotope differences**

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

90 (nimbostratus), vertical air motions are weak¹⁸⁻²³ with mean upward air velocity ($\sim 0.2 \text{ m s}^{-1}$)
91 much lower than typical ice and snow terminal fall velocity ($\sim 1 \text{ m s}^{-1}$). Condensation nuclei (ice
92 particles) form or are introduced near cloud tops and grow, initially by vapor deposition
93 (diffusion) and later by aggregation, while falling slowly towards the surface¹⁸⁻²³. Below the
94 freezing level, melting occurs in an $\sim 500 \text{ m}$ thick layer^{18,19} (‘bright band’ observed in vertical
95 radar reflectivity profiles) and as raindrops fall further, they may partially evaporate, under
96 conditions of subsidence, or grow by accretion and coalescence, under conditions of uplift^{18,19}.
97 The time for condensation, growth and rainfall for stratiform precipitation is about 1-3 hours¹⁹.

98 Hydrometeor growth by vapor diffusion above the freezing level, where tropospheric
99 moisture would have a $\delta^{18}\text{O}$ of ~ -50 to -40% (*ref.* 28), isotopic homogenization in the melting
100 layer, and equilibration with lower altitude moisture²⁹ as relatively small rain drops¹⁹ (on
101 average $\sim 1 \text{ mm}$ in diameter) fall slowly to the surface, all would result in low $\delta^{18}\text{O}$ of stratiform
102 rain. Because of the isotopic exchange below the melting level, near-surface temperature would
103 have a greater influence, resulting in a range of $\delta^{18}\text{O}$. When the melting level is near or at the
104 ground surface ($\sim 1 \text{ km}$ or lower), as in midlatitude winters, limited or negligible exchange with
105 ambient moisture^{3,9,10,28,29} would produce relatively more negative $\delta^{18}\text{O}$ of stratiform
106 precipitation.

107 In contrast, condensation particles in convective clouds (cumulus or cumulonimbus)
108 form near the cloud base¹⁸⁻²³ and grow as they are lifted in strong updrafts ($1\text{--}10 \text{ m s}^{-1}$). The
109 time from initiation of condensation to rainfall in a single convective cell may be as little as
110 thirty minutes¹⁹. When convection is shallow, precipitation forms by ‘warm’ processes¹⁹⁻²¹,
111 wherein condensation occurs below the freezing level and rain drops grow by collision and
112 coalescence. In deep convection, condensation nuclei form as ice particles just above the
113 freezing level, and grow rapidly in updrafts by riming as super-cooled water (mostly from the

114 planetary boundary layer) is frozen at successively higher altitudes¹⁹⁻²³. Entrainment of
115 surrounding air above the freezing level may also contribute minor proportions of moisture for
116 riming growth in convective updrafts¹⁹. As particles become large enough (e.g. graupel) to
117 overcome upward air motion, they descend in strong downdrafts, melt rapidly below the 0°C
118 level, as opposed to a ‘melting layer’ in stratiform precipitation, and fall as large rain drops (>2
119 mm diameter) without significant evaporation or growth¹⁹⁻²³.

120 Convective updrafts are fed by boundary layer moisture, with $\delta^{18}\text{O}$ similar to that of
121 ocean evaporation (~ -12 to -10‰), particularly in oceanic or coastal locations, and slightly
122 lower more inland depending upon the contribution from evaporated soil moisture⁴. The liquid-
123 vapor isotope fractionation^{4,9} is $\sim 10.7 - 9.8\text{‰}$ (0 to 10°C) and shallow convective or ‘warm’
124 rain would likely have a $\delta^{18}\text{O}$ that is about $-1 \pm 1 \text{‰}$. In deep convective precipitation, ice
125 particles from boundary layer moisture just above the freezing level would have $\delta^{18}\text{O}$ of $\sim 0\text{‰}$
126 or higher as ice-vapor isotope fractionation⁹ (at 0 to -10°C) is $\sim 4\text{‰}$ greater than that for liquid-
127 vapor. In addition, depending upon the nature of ice particle surfaces, the kinetic isotope
128 fractionation between ice-vapor or ice-liquid could be even larger³⁰. As ice particles are lifted in
129 updrafts, further accretion (riming) of supercooled boundary layer moisture would also result in
130 relatively higher $\delta^{18}\text{O}$. Entrained environmental air at higher altitudes may have a low $\delta^{18}\text{O}$, but
131 because of its low moisture content, it is unlikely to significantly decrease the $\delta^{18}\text{O}$ of
132 hydrometeors³¹. Ice particles or graupel falling in downdrafts will melt beneath the 0°C level
133 and the large size and fall velocity¹⁹ (~ 5 to 10 m s^{-1}) of the rain drops will inhibit isotopic
134 exchange with ambient moisture^{29,31}, preserving the high $\delta^{18}\text{O}$ acquired during ice and riming
135 growth in updrafts. This is consistent with precipitation tritium (^3H) contents (see below).

136

137 **Tritium in convective rain**

138 Differences in ice formation, growth, and melting in convective and stratiform clouds
139 resulting in different $\delta^{18}\text{O}$ are evident also in the maximum height of high reflectivity, 40-dBZ
140 echo recorded by TRMM precipitation radar^{19,32,33}. 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^{19,32}.
144 Figures 3a and 3b show the $\delta^{18}\text{O}$ 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 $\delta^{18}\text{O}$ with low stratiform
147 fractions occurs with echo heights >5 km (i.e., deep convection) or <~1 km (warm rain). For
148 higher stratiform fractions (>~30%), a wider range of lower $\delta^{18}\text{O}$ occurs with the 40-dBZ echo
149 at ~2-4 km.

150 Figures 4a and 4b show ^3H for selected tropical and midlatitude locations with respect to
151 stratiform fractions and $\delta^{18}\text{O}$. Tritium contents are also derived from the GNIP database¹⁶ and
152 are expressed in tritium units (1TU = 10^{-18} $^3\text{H}/\text{H}$; Methods). As noted earlier, riming growth in
153 deep convective updrafts may include entrained moisture at higher altitudes (~6-8 km) where
154 ^3H contents are expected to be very high^{29,34} (>~ 10^4 TU) compared to those near surface
155 (presently ~5 TU or less). Even minor amounts of this high altitude moisture in hydrometeors
156 (~5% of total water budget) would substantially increase precipitation ^3H contents²³, resulting in
157 a positive $^3\text{H} - \delta^{18}\text{O}$ correlation. Isotopic exchange and equilibration at lower altitudes (as in
158 stratiform rain) will decrease precipitation ^3H contents to ambient levels^{29,34}.

159 At Dhaka and New Delhi, the 40-dBZ echo heights are >~6 km (Fig. 3). Deep
160 convection with similar echo heights is also known to occur at Vienna and Krakow³⁵. All of
161 these locations show an increasing ^3H with decreasing stratiform fraction or increasing $\delta^{18}\text{O}$

162 (Figs. 4a, 4b), indicating that higher $\delta^{18}\text{O}$ did not result from isotopic exchange or in-cloud rain
163 re-evaporation at lower altitudes. Lower ^3H and a lack of $^3\text{H} - \delta^{18}\text{O}$ 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
169 clouds in the tropics and midlatitudes¹⁸⁻²⁴ where mesoscale convective systems and extra-
170 tropical cyclones produce most of the precipitation¹⁹. Average annual stratiform fractions (on a
171 volume basis) are ~35-55% in the tropics²¹ and are higher in the midlatitudes^{19,35}. Smaller, less
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
174 events both in the tropics and midlatitudes^{1,6,8,15}. Complex patterns⁴⁻⁸ of precipitation $\delta^{18}\text{O}$
175 decreasing, increasing, and then decreasing again with time are commonly observed⁴⁻⁸ during
176 storm events and may easily result from successive storms with stratiform precipitation trailing
177 convective^{19,36}. A progressive decrease, increase or no change in $\delta^{18}\text{O}$ with time^{5,6,14,15} may
178 occur when stratiform precipitation trails, leads, or overlaps convective precipitation³⁶.

179 The $\delta^{18}\text{O}$ – stratiform fraction correlation shown in Figure 1 for monthly data is found
180 also at storm event or interannual time scales. Recent measurements of isotope ratios in the
181 equatorial Indian Ocean^{4,15} and in California, USA⁸ both indicate that $\delta^{18}\text{O}$ is low ($\sim -20\text{‰}$)
182 when precipitation is dominantly stratiform and high ($\sim 0\text{‰}$) when it is mostly convective.
183 Significant interannual variability occurs in response to regional or global phenomenon, such as
184 the El Niño–Southern Oscillation (ENSO)³⁷, and the character of precipitation is affected
185 differently across the Pacific²¹, 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 $\delta^{18}\text{O}$ (-4.9‰ ; average -6.0‰), while at Bellavista, fractions were
190 higher (49%; average 30%) and $\delta^{18}\text{O}$ lower (-5.5‰ ; average -2.9‰).

191 The negative correlation between $\delta^{18}\text{O}$ and stratiform rain fractions (Fig. 1a) is also
192 consistent with the previously observed positive correlation between $\delta^{18}\text{O}$ and moisture
193 residence time¹⁷ because residence time would be lower, and stratiform fractions higher, in
194 well-organized, mesoscale convective systems and extra-tropical cyclones^{19,21}.

195 We conclude that $\delta^{18}\text{O}$ 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^{38,39}.

202 Precipitation partitioning based on isotope distributions should help to improve climate
203 models²⁵ and the simulation of convection by providing observational constraints for
204 tropospheric heating profiles and rain types^{18-22,41}. 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 $\delta^{18}\text{O}$ recorded in
208 proxy archives of paleo-precipitation⁴¹, such as speleothems and tropical ice, should reflect a
209 change in the proportions of convective and stratiform precipitation. Lower or higher $\delta^{18}\text{O}$, 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.

214
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345

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.

349

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

351 **METHODS**

352 **Stratiform fractions**

353 *Tropical locations:* Stratiform rain fractions were calculated using NASA's TRMM PR 2A23 (rain
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 ~2 km
358 above mean ground level height. Stratiform and convective conditional rain rates were estimated from
359 mean radar reflectivity (≥ 18 dBZ) using 2A25 V7 initial stratiform and convective Z-R relations²⁷.
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
364 the algorithm predicts are raining are very small in count (1.7% of all raining pixels) and likely don't
365 contribute much to rainfall near the surface; this rain appears to mostly exist at higher levels and has an
366 anvil appearance²⁷. These pixels are not used in our stratiform-convective rain calculations. Therefore,
367 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 base²⁴:
371 (http://badc.nerc.ac.uk/view/badc.nerc.ac.uk__ATOM__dataent_ukmo-midas). Precipitation is
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**

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

386 $\delta^{18}\text{O}$ and precipitation amount were retrieved from the global network of isotopes in
387 precipitation (GNIP) database¹⁶ http://www-naweb.iaea.org/napc/ih/IHS_resources_gnip.html. Tropical
388 locations in the 30°N – 30°S latitude range, with a minimum of four consecutive years of isotope data
389 in the 1998-2014 period, were selected. Isotope data from Jambi and Bukit stations in Indonesia were
390 obtained within the framework of “Daily basis precipitation sampling network for water isotope
391 analysis” at the Institute of Observational Research for Global Change, Japan Agency for Marine-Earth
392 Science and Technology, by N. Kurita and K. Ichiyanagi. In addition, data for two midlatitude

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

397 Tritium (^3H) is reported in Tritium Units (TU) where $1 \text{ TU} = 10^{-18} \text{ } ^3\text{H}/\text{H}$. Analyses of water
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 ± 0.1 to ± 0.5 TU. Tritium is produced in
401 the stratosphere by cosmic ray spallation and reaches the troposphere by moisture ‘leakage’ or
402 exchange and its abundance decreases towards the Earth’s surface^{20,26}. In addition, a latitudinal
403 gradient (decreasing ^3H contents towards the equator) exists due to the fact that stratospheric exchange
404 occurs mostly in mid to high latitudes^{20,21}. Stratospheric ^3H injection by thermo-nuclear bomb tests
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
407 1990s, but vertical differences in tropospheric ^3H contents remained.

408 Precipitation ^3H contents were also retrieved from the GNIP database for 1998–2012, except for
409 New Delhi where data were available for 1964–1978. During this period, atmospheric ^3H was declining
410 rapidly due to the wash out of ‘bomb’ tritium³⁴. As a result, absolute ^3H values for New Delhi cannot
411 be compared directly with other locations. The $^3\text{H} - \delta^{18}\text{O}$ correlations for New Delhi shown in Fig. 4b
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
414 by declining concentrations. The lower ^3H at Dhaka compared to Vienna or Krakow reflects the
415 latitudinal gradient in tropospheric ^3H concentration because the stratosphere-troposphere exchange

416 that brings tritium to the troposphere occurs mostly at higher latitudes^{29,34}, as well as differences in the
417 height of convective entrainment.

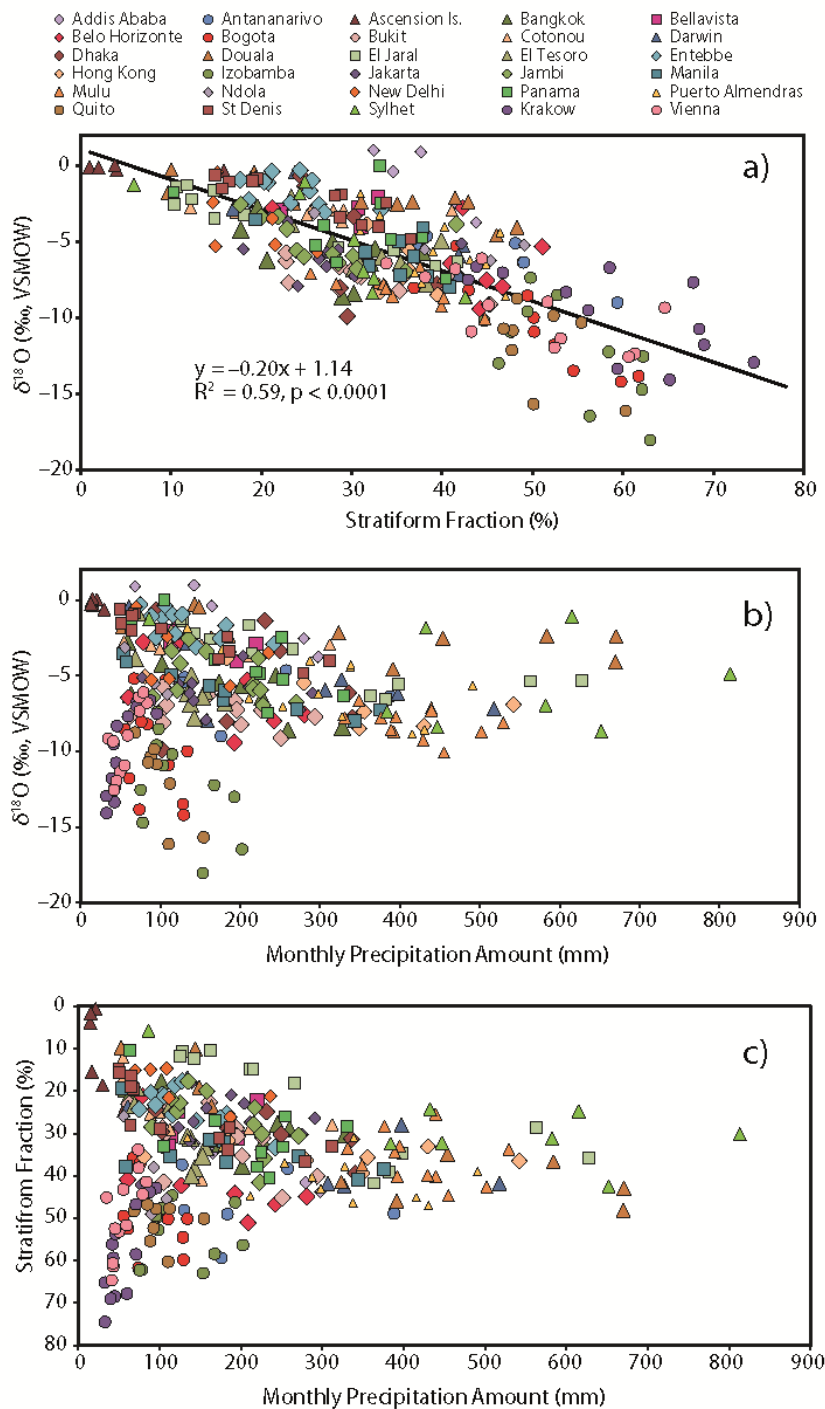
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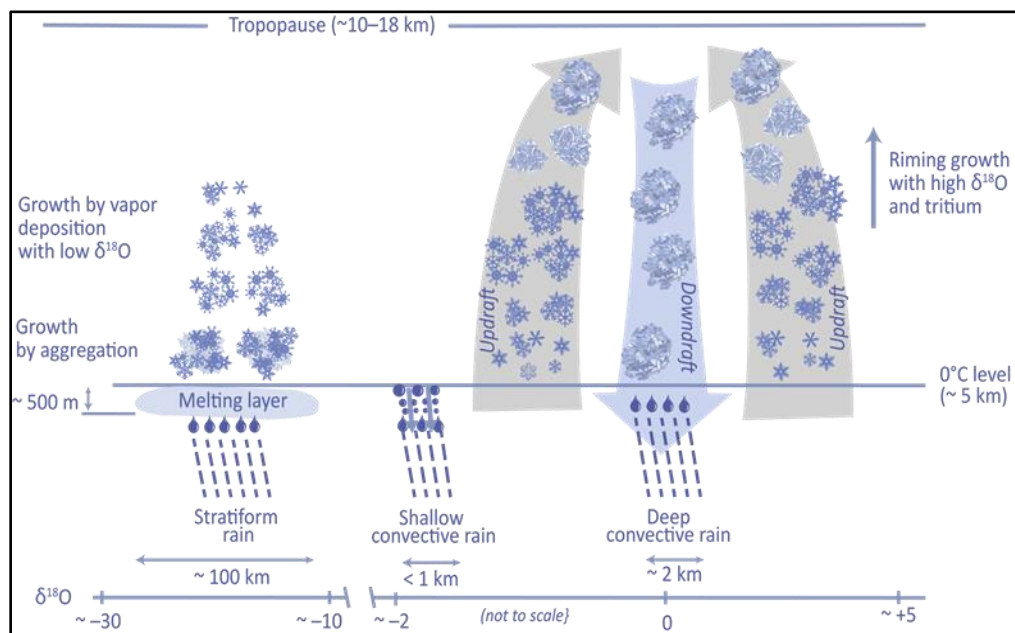
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Figure 1. Correlation of mean monthly $\delta^{18}\text{O}$, stratiform fraction and precipitation amount in tropical and midlatitude precipitation. The remainder of the rain fraction is convective (Methods). Solid line in panel a) is based on a linear regression of all data points shown. The vertical axis in c) is shown in reverse scale and trends in b) and c) are similar but opposite with respect to precipitation amount



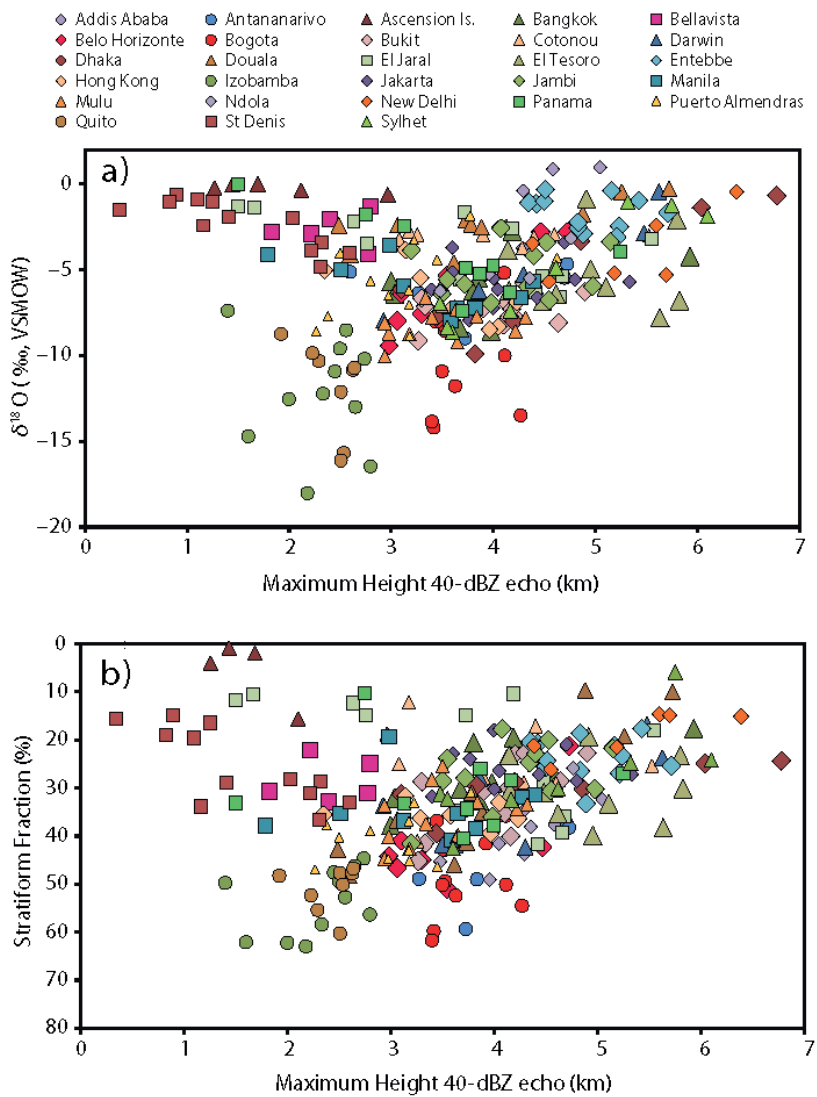
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426 **Figure 2.** Schematic representation of differences in dynamical and microphysical processes in
427 convective and stratiform precipitation resulting in isotope variations. (Adapted after Houze Jr.^{18,19})
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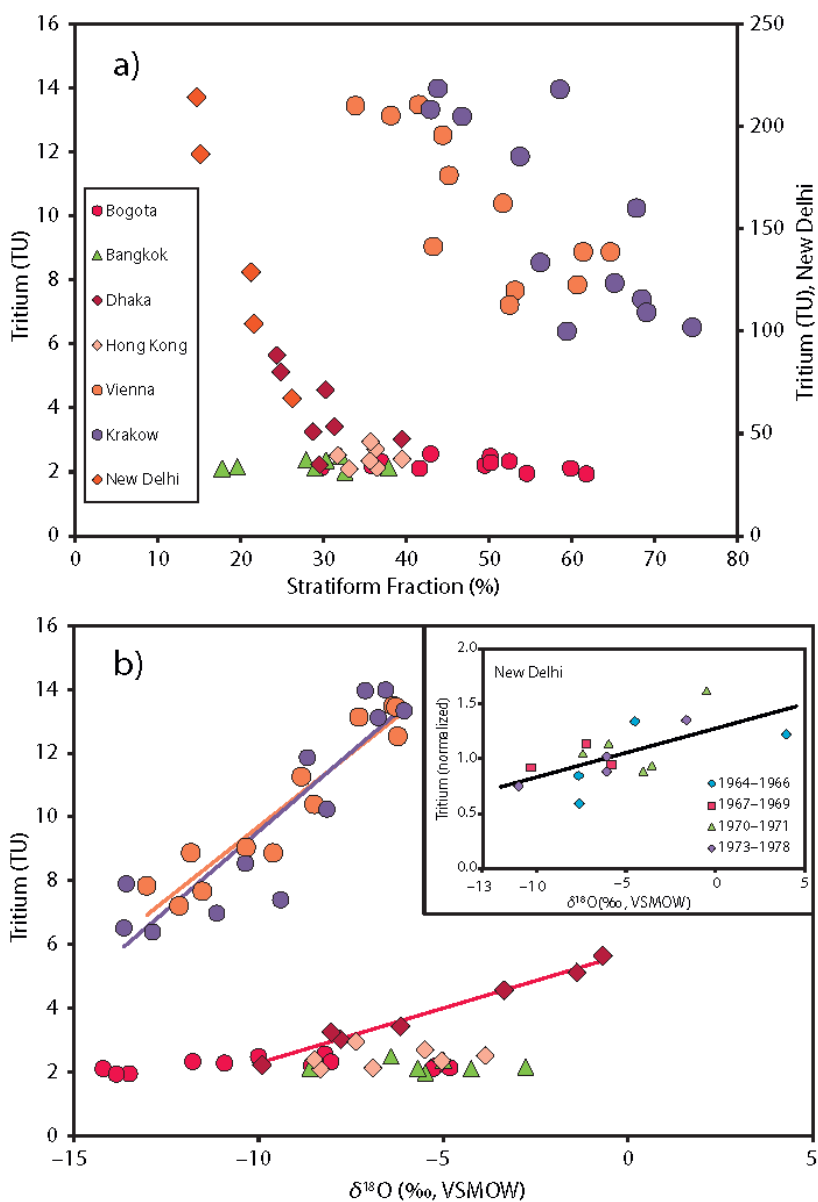
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434 **Figure 3.** Correlation of mean monthly $\delta^{18}\text{O}$ and stratiform fractions with the maximum height of 40-
 435 dBZ echo. Precipitation with low stratiform fractions (or high convective fractions) has higher $\delta^{18}\text{O}$ but
 436 low or high echo heights reflecting shallow (warm rain) or deep convection.
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440 **Figure 4.** Correlation of monthly precipitation tritium (^3H) content with stratiform fractions and $\delta^{18}\text{O}$.
 441 In 4a, note the much higher tritium at New Delhi because 1964-1978 data are used, while 2000-2012
 442 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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Table 1. Geographical coordinates and related information for locations used in this study.

Code	Location	Lat.	Long.	Alt. (m)	Average Annual		Months with Precip > 50 mm*
					Precip (mm)	T (°C)	
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

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