A Statistical Method for Generating Temporally Downscaled Geochemical Tracers in Precipitation

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Early Online Release: This preliminary version has been accepted for publication in *Journal of Hydrometeorology,* may be fully cited, and has been assigned DOI 10.1175/JHM-D-20-0142.1. The final typeset copyedited article will replace the EOR at the above DOI when it is published.

ABSTRACT

Sampling intervals of precipitation geochemistry measurements are often coarser than those required by fine-scale hydrometeorological models. This study presents a statistical method to temporally downscale geochemical tracer signals in precipitation so that they can be used in high-resolution, tracer-enabled applications. In this method, we separated the deterministic component of the time series and the remaining daily stochastic component, which was approximated by a conditional multivariate Gaussian distribution. Specifically, statistics of the stochastic component could be explained from coarser data using a newly identified power-law decay function, which relates data aggregation intervals to changes in tracer concentration variance and correlations with precipitation amounts. These statistics were used within a copula framework to generate synthetic tracer values from the deterministic and stochastic time series components based on daily precipitation amounts. The method was evaluated at 27 sites located worldwide using daily precipitation isotope ratios, which were aggregated in time to provide low resolution testing datasets with known daily values. At each site, the downscaling method was applied on weekly, biweekly and monthly aggregated series to yield an ensemble of daily tracer realizations. Daily tracer concentrations downscaled from a biweekly series had average (+/- standard deviation) absolute errors of 1.69‰ (1.61‰) for δ^{2}H and 0.23‰ (0.24‰) for δ^{18}O relative to observations. The results suggest coarsely sampled precipitation tracers can be accurately downscaled to daily values. This method may be extended to other geochemical tracers in order to generate downscaled datasets needed to drive complex, fine-scale models of hydrometeorological processes.

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

1. Introduction

Naturally occurring chemical signatures in precipitation (e.g. Bailey et al. 2018, Bowen et al. 2019, Gibson et al. 2005, Kendall and McDonnell 2012, Moerman et al. 2013, West et al. 2010, Wiederhold 2015) are frequently used as hydrometeorological tracers, especially when inferring transport or chemical transformations through terrestrial, aquatic, and atmospheric environments (e.g. Abbott et al. 2016, Brooks et al. 2014, Good et al. 2015, Gupta et al. 2020, Kanner et al. 2014, Remondi et al. 2018). Tracer-enabled modeling allows for process-level inference based not only on the size of fluxes, but also on the spatial and temporal transport and mixing of the geochemical signatures associated with the fluxes, thereby facilitating improved understanding and multi-response model evaluation (Bowen and Good 2015, Krause et al. 2005, McGuire and McDonnell 2006, Sprenger et al. 2019, Turnadge and Smerdon 2014). Researchers have used tracers within global climate models to evaluate processes that are challenging to observe (e.g. ageostrophic circulations, convection and turbulence) or are modeled at sub-grid scales and are therefore not explicitly simulated but parameterized (e.g. Gupta et al. 2020, Orbe et al. 2020, Rosa et al. 2012). For instance, isotope-enabled general circulation models (GCMs) have explicitly simulated water isotope ratios within the critical zone on sub-daily time scales (e.g., a version of the Community Earth System Model (iCESM1); Brady et al. 2019, Nusbaumer et al. 2017, Wong et al. 2017) and provide outputs which have been evaluated against observational datasets at various scales (e.g., Hoffmann et al. 2000, Nusbaumer et al. 2017, Risi et al. 2012, Steen-Larsen et al. 2016, Wong et al. 2017).

In many modeling applications, observed and modeled temporal resolutions are different and, in these cases, a downscaling method is required in order to use observed datasets within a model to evaluate processes with dynamic fluctuations over short temporal intervals (Ebtehaj and

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

Foufoula-Georgiou 2013). The temporal resolution at which many geochemical tracers are collected, a result of analytical or logistical cost or the need to aggregate them in time to achieve a measurable signal due to low tracer concentrations, contrasts with the time steps typical of many hydrometeorological models (Rosa et al. 2012, Gupta et al. 2020). Accordingly, a method is needed to generate higher frequency datasets of precipitation chemistry from low frequency collections.

Statistical downscaling leverages relationships observed in both fine- and coarse-scale measurements to predict fine-scale variations where only coarse-scale data are available (Ebtehaj and Foufoula-Georgiou 2013, Goncu and Albek 2016). Extensive work has focused on downscaling precipitation rate, including through use of temporal neural networks (e.g. Coulibaly et al. 2005), stochastic methods (e.g. Bordoy and Burlando 2013, D'Onofrio 2013, Poduje and Haberlandt 2017), and conditional multivariate statistical models (e.g. Yang et al. 2010). However, past studies have not temporally downscaled precipitation chemistry data, as is warranted for tracer applications.

Precipitation stable isotope ratios (δ^2H and δ^{18}O) are an ideal test case for developing a downscaling method that can benefit tracer applications, if the downscaling can preserve multi-scale statistical properties (Ebtehaj and Foufoula-Georgiou 2013). Not only is such downscaling in demand, decades of research demonstrate patterns in precipitation isotope ratios that could be leveraged in downscaling; specifically, precipitation amount often covaries with isotopic composition, attributable to the interplay of diverse climatological, physiographical, and meteorological factors in the evaporation, condensation, and transport of atmospheric moisture (e.g. Aggarwal et al. 2016, Aggarwal et al. 2012, Bowen et al. 2019, Ingraham 1998, Konecky et al. 2019, Lee and Fung 2008, Moore et al. 2016, Risi et al. 2008, West et al. 2010). This (typically

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

inverse) covariation between precipitation rates and isotope ratios, often referred to as an "amount effect", represents partially systematic variations at sub-seasonal, monthly, and event time scales (Celle-Jeanton et al. 2001, Conroy et al. 2016, Craig 1961, Craig and Gordon 1965, Gat 1996, Lee and Fung 2008, Moore et al. 2013, Tharammal et al. 2017). If these amount effects share statistical similarities across various time scales, they could support downscaling methods to predict short-term fluctuations. Hypothetically, relationships inferred from sporadic or brief datasets could be used to predict short term variations in precipitation isotopic composition. Those patterns could be superimposed on the longer timescale seasonal patterns, which tend to follow regional patterns (Bowen et al. 2019, Dansgaard 1964, Feng et al. 2009, Allen et al. 2019), to potentially generate realistic, continuous, high-frequency time series of precipitation isotope ratios.

In this study, we developed and evaluated a downscaling method that uses the statistical structure of observed stable isotope time series to downscale and generate stable isotope time series at finer resolutions. We used daily observations of precipitation amounts and isotope ratios from 27 monitoring stations across the globe. The daily data were artificially aggregated to weekly, biweekly and monthly scales, using amount-weighted running means to simulate coarser-scale datasets on which to apply the method. These aggregated time series were evaluated for statistical trends, specifically characterizing how the time-series means, standard deviations and correlation structures changed as the temporal sampling interval increased. Then, the statistical downscaling method was applied on each of the weekly, biweekly and monthly aggregated tracer time series to generate downscaled tracer values. An ensemble of downscaled realizations was generated at each site, the statistics of which were compared to those of the original daily observations. Our objective was to generate downscaled realizations that accurately preserved the observed daily δ^2H and δ^{18}O

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

means and standard deviations and the correlation structure between precipitation amount, δ^2H, and δ^{18}O, so that these realizations could be suitable for various potential modeling applications.

2. Data and Methods

a. Site Information and Tracer Datasets

Daily precipitation stable water isotope time series were downloaded from the International Atomic Energy's (IAEA) Global Network of Isotopes in Precipitation (GNIP) and Water Isotope System of Data Analysis, Visualization and Electronic Retrieval (WISER) database (IAEA/WMO 2020). Each time series was filtered to ensure precipitation values were greater than zero and had corresponding δ^2H and δ^{18}O isotope ratios. All time series with greater than one year of observations were selected, resulting in the 27 datasets used in the subsequent analysis; details pertaining to each site are included in Table S1 located in the *Supplementary Materials*. A minimum time series length of one year was chosen because we wanted to account for site-specific seasonal precipitation patterns in the generated downscaled tracer time series. We acknowledge seasonality is usually characterized over time scales greater than one year, however for this analysis we decided on a minimum of one year so the downscaling method could be applied to as many datasets as possible. In the *Discussion*, the downscaling method's performance was evaluated against the number of years represented in the time series and the frequency of collection, i.e. the number of recorded precipitation events divided by the total number of days represented in the time series. The time series lengths ranged from 1.22 to 15.94 years, with an average of 5.34 years. The total number of samples in a time series ranged from 33 to 1026, with an average of ~210. The site with 33 samples (Barasat, Kolkata; Table S1) was sampled over 1.33 years.

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

All hydrogen and oxygen isotope ratios of precipitation were denoted as δ^2H and δ^{18}O, defined by

$$\delta\ (‰) = \frac{R_{sample} - R_{std}}{R_{std}}\ 1000 \qquad \text{Eq. 1}$$

where δ was the isotope ratio in delta notation, R_{sample} was the ratio of concentrations between the rare and abundant isotopologues, and R_{std} was the isotopic ratio standard; for this analysis, that standard was the Vienna Standard Mean Ocean Water (VSMOW). The site locations and average stable water isotope observations were represented in Figure 1. The 27 sites have an average mean (+/- standard deviation) daily observed precipitation of 22.80 (21.38) mm, δ^2H of -37.77 (24.62) ‰ and δ^{18}O of -6.03 (3.21) ‰. The maximum recorded total daily precipitation ranged from 43.0 to 317.5 mm across sites. At the 27 sites, the observed isotope ratios ranged from -228.0 to 43.35 ‰ for δ^2H and -30.50 to 8.81 ‰ for δ^{18}O. The site list included geographic locations across different climates and with uniform and seasonally varying precipitation amounts.

Isotope ratios are often evaluated relative to the Global Meteoric Water Line (GMWL), which is defined as $\delta^2\mathrm{H} = 8\delta^{18}\mathrm{O} + 10\ ‰$ (Craig, 1961). Deuterium excess (*d*-excess $(‰) = \delta^2\mathrm{H} - 8\delta^{18}\mathrm{O}$) measures the deviation of a water sample's composition from the GMWL (Dansgaard, 1964) and is a useful secondary tracer in that it varies with respect to the evaporation and mixing history of airmasses (e.g., Benetti et al. 2014, Fröhlich et al. 2002, Pfahl and Sodemann 2014). One can use *d*-excess to understand both the source of precipitation and the evolution of moisture during transport (Fröhlich et al. 2002, Good et al. 2014). We aimed to preserve a site's *d*-excess in the downscaled time series because it can be informative for a variety of hydrological and meteorological applications.

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

b. Constructing Low-resolution Datasets

We aggregated each of the 27 GNIP site's datasets using a moving, precipitation amount-weighted average (Eq. 2). This provided us with datasets of low-resolution tracer time series on which to apply the downscaling method to generate downscaled daily estimates to compare with the observed daily values. The precipitation amount-weighted average was defined as

$$\overline{\delta^2 H_t} = \frac{\sum_{i=1}^{t} P_i \delta^2 H_i}{\sum_{i=1}^{t} P_i} \ \ and \ \ \overline{\delta^{18} O_t} = \frac{\sum_{i=1}^{t} P_i \delta^{18} O_i}{\sum_{i=1}^{t} P_i} \qquad \text{Eq. 2}$$

where P was total precipitation (mm), $\delta^2 H$ (‰) and $\delta^{18} O$ (‰) were the daily observed stable water isotope tracer values at time *t* (days), and $\overline{\delta^2 H_t}$ (‰) and $\overline{\delta^{18} O_t}$ (‰) were the *t*-day average tracer value within the specified aggregated temporal interval. $\overline{\delta^2 H_t}$ and $\overline{\delta^{18} O_t}$ values populated a time series at *t* level of aggregation. We focused on downscaling time series aggregated at *t* values of 7-, 14-, and 28-days (weekly, biweekly, and monthly).

Time series statistics were evaluated across a range of temporal intervals. Moerman et al. (2013) investigated the correlation structure between precipitation amount and δ^{18}O at Mulu Meteo, Sarawak (Table S1) at daily to 12-week (84-days) time scales. Following their approach, we evaluated trends in the mean (μ), standard deviation (σ), and Pearson correlation coefficient (ρ) at different temporal intervals to capture the time series response and prediction accuracy. The ρ measures the linear correlation between two variables and has a value between -1 and 1, where 1 is a total positive linear correlation, 0 is no linear correlation, and -1 is total negative linear correlation. At the daily scale for the 27 GNIP sites, the average (+/- standard deviation) ρ(P, δ^2H) was -0.18 (+/- 0.18), ρ(P, δ^{18}O) was -0.20 (+/- 0.17) and ρ(δ^2H, δ^{18}O) was 0.96 (+/- 0.03).

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

c. Statistical Precipitation Tracer Downscaling Method

1) REMOVAL OF THE DETERMINISTIC TIME SERIES COMPONENT

Each aggregated weekly, biweekly and monthly time series (Eq. 2) was treated as an example of a low-resolution dataset on which to apply the downscaling method. We considered each tracer time series to have a deterministic component and a stochastic component. In the first step, the deterministic component was characterized by the seasonality in the precipitation signal and was removed from each set of observations. Isotope ratios in precipitation frequently have been observed to exhibit distinct seasonal signals. These can be approximated as a combination of sinusoidal functions through Fourier decomposition (Allen et al. 2018, Allen et al. 2019, Dutton et al. 2005, Feng et al. 2009, Halder et al. 2015, Vachon et al. 2007, Wilkinson and Ivany 2002). Sinusoidal functions effectively describe the collinear structure and fluctuations in the covariation of δ^2H and δ^{18}O relative to the GMWL (Figure 1; Allen et al. 2018, Craig 1961, Dansgaard 1964). The sine curve parameters (amplitude, phase, and offset) are often predictable in space (Allen et al. 2018, Jasechko et al. 2016) and succinctly represent temporal dynamics because they express continuous, cyclic time series. Allen et al. (2019) used monthly isotopes in precipitation GNIP datasets from across the globe to capture patterns in the precipitation isotope seasonality using sinusoidal functions. When predicting the isotope seasonality, the values of the sine parameters can be described as functions of climate and geography. Additionally, sine curves are useful when describing the propagation of cyclic signals, this has been done to infer catchment-scale mixing processes using the dampening ratio of seasonal isotope amplitudes in streamflow versus precipitation (Kirchner 2016a, 2016b; also see Clow et al. 2018, von Freyberg et al. 2018, Gallart et al. 2020, Jacobs et al. 2018, Jasechko et al. 2016, Lutz et al. 2018, Song et al. 2017).

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

We fitted sinusoidal functions to each of the site's daily to 12-week aggregated time series to describe the deterministic components using a non-linear, least squares fitting routine, "curve_fit" in Python's (v3.7.6) SciPy Library (v1.2.1), following the methods from Allen et al. (2018). We used a time-weighted fit routine (i.e., not amount weighted and each daily sample had equal weight) to approximate the parameters of the sinusoidal function (Eq. 3) because our ultimate goal related to predicting daily precipitation variations in isotopic composition, regardless of whether or not they are associated with larger events. The sine functions were defined with a fixed period of one year and

$$Precipitation\ \delta^2H \text{ or } \delta^{18}O(f) = A\,sin(2\pi f - \phi) + b, \quad \text{Eq. 3}$$

where f was the fractional year and b was an offset parameter (Allen et al. 2018). All fitted amplitudes (A) and phases (ϕ) were bounded so the amplitude values were positive and the phase ranged between $-\pi$ and π. The presence of large seasonal isotope cycles enables the quantification of mixing, transport and turnover of water in landscape and/or biota. Amplitude dampening reflects mixing processes, phase shifts reflect advective travel times and offset differences reflect proportional contributions of different seasons' precipitation (Kirchner 2016a, 2016b). The defined sinusoidal functions were subtracted from the daily to 12-week aggregated series, thus removing the deterministic time series components.

2) GENERATION OF STOCHASTIC TRACER REALIZATIONS

Next, the daily statistics of the stochastic hydrogen (δ^2H^*) and oxygen ($\delta^{18}O^*$) isotope time series were estimated by using the relationship between the observed daily stochastic statistics

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

and the stochastic signal's statistics across a range of aggregation intervals (t) multiplied by each site's specific precipitation frequency (λ; defined as the number of days with precipitation divided by the total number of days in a time series). The statistics of the stochastic signal at aggregation interval t were denoted with * as μ_t^*, σ_t^*, and ρ_t^* and estimates of these at the daily (t=1) resolution were denoted as $\hat{\mu}_1^*, \hat{\sigma}_1^*,$ and $\hat{\rho}_1^*$. After removal of the deterministic component, the stochastic signals had mean isotope values of approximately zero across all ranges of $t\lambda$ (Figure 2.a,b). Consequently, we assumed the stochastic signal to behave as a purely random mean zero process ($\hat{\mu}_1^* = 0$), which was further substantiated using tests for independence, autocorrelation, and normality on the stochastic signal (refer to *Sections 2.d* and *3.d*).

The time series standard deviations were greatest at daily time scales and decreased with increasing $t\lambda$ as a power law function (Figure 2.c,d). This decrease resulted from the averaging and weighting of individual daily tracer concentrations by precipitation amounts over longer temporal intervals. By the Central Limit Theorem and the Law of Large Numbers, as the sampling size increases, the sampling distribution converges to a normal distribution where the standard deviation decreases at a rate of $1/n^{0.5}$, where n is a number of samples. It should be noted, the results from the Central Limit Theorem and the Law of Large Numbers holds as long as the signal is purely stochastic and there are no trends or heteroscedasticity in the time series. It was assumed a similar relationship was held between the daily standard deviation of days with precipitation tracer values (σ_1^*, ‰) and the series of known t-day aggregation intervals (days) with their corresponding standard deviations in time (σ_t^*, ‰). $t\lambda$ estimated the expected number of precipitation events in each aggregation level because precipitation does not occur every day (e.g. $n \approx t\lambda$). We expressed this relationship as

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

$$\sigma_t^* = \frac{\hat{\sigma}_1^*}{(t\lambda)^a}, \qquad \text{Eq. 4}$$

where *a* is a site-specific parameter defining the rate decrease in σ_t^* with increasing *t*. We used the non-linear, least-squares fitting routine, "curve_fit", in Python's (v3.7.6) SciPy Library (v1.2.1) to estimate the *a* and $\hat{\sigma}_1^*$ parameters in Eq. 4. a was constrained between 0.2 and 0.5 in order to bound the curve fitting routine. When $\hat{\sigma}_1^*$ was compared with σ_1^*, a values below 0.2 often underpredicted $\hat{\sigma}_1^*$ and above 0.5 often overpredicted $\hat{\sigma}_1^*$. The initial value predicted for *a* was set at 0.3, however varying this had negligible influence on the final *a* parameter estimates and the *a* parameter estimates were not strongly related to observed standard deviation (for δ^2H^* and $\delta^{18}O^*$ $R^2 < 0.002$ and p-value > 0.75). To estimate the daily standard deviation at a site with a biweekly (t = 14) sampling frequency, first λ must be calculated and the time series can be aggregated to 28-, 42-, 56-, 70-, and 84-day intervals (Eq. 2) for 2*t* to 6*t*, giving 6 points to fit Eq. 4. Weekly time series were aggregated from 2*t* to 12*t* (12 points), while monthly time series were aggregated from 2*t* to 3*t* (3 points). This quantified the decrease in σ_t^* from the available data resolution out to 12 weeks (84-days), and allows *a* and $\hat{\sigma}_1^*$ to be estimated.

The ratio of ρ_t^* divided by ρ_1^* across λt was relatively invariant and centered around one (Figure 2.e-g). Thus, Pearson correlation coefficients at a *t*-day aggregation interval (ρ_t^*) were used to describe the daily correlations ($\hat{\rho}_1^*$) between precipitation amount and the stochastic signal's δ^2H^* and $\delta^{18}O^*$ values.

Pseudo-random numbers were generated using a Gaussian copula (Sklar 1959), defined by the estimated daily statistics, $\hat{\mu}_1^*$'s, $\hat{\sigma}_1^*$'s, and $\hat{\rho}_1^*$'s, and conditioned on the observed daily precipitation amounts. Other copula families are possible (e.g. Archimedean copula, Gumbel copula); however, here the Gaussian copula was used because it offered a simple approach for

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

modeling the dependence of multivariate states (Schneider and Ramos 2014). In probability theory and statistics, the marginal distribution of a subset of a collection of random variables is the probability distribution of one random variable without any reference to other random variables. Copula models separate the dependency structure of multiple random variables from their marginal distributions by mapping each variable through its cumulative distribution functions (CDF) to the unit interval (i.e. closed interval [0,1]). This captures the dependence between the variables using a copula or coupling term, allowing a different marginal distribution for each variable while capturing the multivariate dependencies (Schneider and Ramos 2014, Sklar 1959). Here, a copula captured the multivariate dependencies between precipitation amount, $\delta^2\mathrm{H}^*$ and $\delta^{18}\mathrm{O}^*$. Refer to *Supplemental Material* for further detail on the definition of the Gaussian copula used here. Models using copula techniques have captured the spatial and temporal patterns of precipitation characteristics (Kuhn et al. 2007, Gao et al. 2018), temporally downscale precipitation datasets (Gyasi-Agyei 2011, So et al. 2017), to forecast precipitation events (Bárdossy and Pegram 2009, Khedun 2014) and across other hydrological disciplines (e.g. temperature and rainfall dynamics (Cong and Brady 2012, Schölzel and Friederichs 2008), extreme-value stochastic rainfall events (Kuhn et al. 2007, Laux et al. 2011, Huang et al. 2012), drought distributions from monthly rainfall (Laux et al. 2009), hydraulic conductivity of aquifer systems (Haslauer et al. 2012), and groundwater recharge from precipitation events (Jasechko and Taylor 2015).

For each observed precipitation amount, values of $\delta^2\mathrm{H}^*$ and $\delta^{18}\mathrm{O}^*$ (a 2-number sample representing the stochastic signal) were drawn from a multivariate Gaussian distribution using Python's (v3.7.6) SciPy Library (v1.2.1) with parameters described by $\hat{\rho}_1^*(P, \delta^2H^*)$, $\hat{\rho}_1^*(P, \delta^{18}O^*)$ and $\hat{\rho}_1^*(\delta^2H^*, \delta^{18}O^*)$ (refer to Eq. 9 and 10 in the *Supplemental Material*). The covariates used here were precipitation amount and its isotopic composition, however it should be

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

noted the covariates can change depending on the method's application and data availability. Next, Gaussian CDF values were calculated for each of the generated series. The resulting uniform values were then used to resample from the coarse resolution empirical distribution of isotope ratios for each site, formed by the deseasonalized time series. Each of these values was then rescaled by $\hat{\sigma}_1^*/\sigma_t^*$. The resulting stochastic time series were daily $\delta^2\mathrm{H}^*$ and $\delta^{18}\mathrm{O}^*$ values conditioned on observed precipitation amounts with means of zero, standard deviations of $\hat{\sigma}_1^*$ and Pearson correlation coefficients of $\hat{\rho}_1^*$.

3) FULL SYNTHETIC TIME SERIES GENERATION

The deterministic component, the sinusoidal function from Eq. 3, was added to each generated stochastic time series. The result was a downscaled tracer time series which captured site-specific daily precipitation amount effects, seasonal signals and stochastic variability. Finally, we applied a residual correction on the downscaled synthetic series to preserve the observed aggregated weighted tracer values. For each synthetic value within each aggregation interval, an interval-specific, single correction factor was subtracted from the downscaled values so that there was no difference between that period's downscaled synthetic values aggregated for that interval and the observed coarse-resolution interval's value. In doing so, the precipitation-weighted values of the synthetic time series then equaled the known aggregated value. This property is particularly important as it closes the tracer mass balance. The statistical downscaling method applied to a dataset with a biweekly sampling frequency was summarized and visualized in *Section (ii)* of the *Supplemental Material*.

d. Evaluation of Precipitation Tracer Downscaling Mythology

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

The statistical method was iterated over 100 times generating an ensemble of downscaled isotope time series at each of the 27 GNIP locations. The large number of time series generated for each ensemble allowed for us to quantify the performance of the downscaling method. Each ensemble was expected to capture the observed site-specific tracer means and standard deviations and the correlation coefficients between precipitation amount, δ^2H and $\delta^{18}O$. The statistical downscaling method was evaluated using multiple techniques, detailed in the subsequent paragraphs.

After removing the deterministic components, the stochastic time series were expected to have means of approximately zero, a predicable decrease in standard deviation (Eq. 4) and Pearson correlation coefficients at low temporal resolutions appropriately defining daily covariate structures. To test this, σ_1^* and ρ_1^* of the observed datasets were compared to $\hat{\sigma}_1^*$ and $\hat{\rho}_1^*$ of downscaled ensembles using root-mean squared error (RMSE), mean bias error (MBE), and R-squared (R^2). Autocorrelations with lags ranging from 1- to 20-days (Figure 6, refer to *Section 3.c*) and tests for normality were calculated for the stochastic signal of the observed datasets and downscaled ensembles.

The average of ensemble means ($\overline{\hat{\mu}_1}$) and standard deviations ($\overline{\hat{\sigma}_1}$) for each isotope ratio and the Pearson correlation coefficients ($\overline{\hat{\rho}_1}$) between precipitation amount and each isotope ratio were compared to the observed daily statistics. R^2 values were calculated for the downscaled ensembles and observed daily site statistics. Each site's observed *d*-excess was evaluated against the downscaled ensemble's *d*-excess. Lastly, we compared the absolute error between the downscaled ensemble and observed time series means to various site-specific and time series characteristics.

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

3. Results

a. Evaluation of Estimated Daily Stochastic Signal Statistics

The estimated daily stochastic signal statistics from weekly, biweekly and monthly aggregation intervals accurately described the observed statistics (Figure 3). The method best predicted $\hat{\sigma}_1^*$ when applied to a weekly series, while the worst approximations of $\hat{\sigma}_1^*$ occurred when it was applied to a monthly series. We expected the weekly time series to best predict $\hat{\sigma}_1^*$ because it better characterizes the change in tracer concentration variance as more values of *t* were used to fit the $\hat{\sigma}_1^*$ and *a* parameters in Eq. 4. For all 27 GNIP sites, the δ^2H $\hat{\sigma}_1^*$ had RMSEs of 2.73 ‰ (MBE = -1.92 ‰) for a weekly series, 5.21 ‰ (MBE = -3.72 ‰) for a biweekly series, and 7.83 ‰ (MBE = -6.02 ‰) for a monthly series. The δ^{18}O $\hat{\sigma}_1^*$ had RMSEs of 0.35 ‰ (MBE = -0.77 ‰) for a weekly series, 0.64 ‰ (MBE = -0.48 ‰) for a biweekly series, and 0.98 ‰ (MBE = -0.24 ‰) for a monthly series. For weekly, biweekly and monthly series, $\hat{\rho}_1^*$(P, δ^2H), $\hat{\rho}_1^*$(P, δ^{18}O), and $\hat{\rho}_1^*$(δ^2H, δ^{18}O) had low RMSEs ranging from 0.01 to 0.18 ‰ and MBEs ranging from -0.01 to 0.03 ‰ across all sites. $\hat{\rho}_1^*$(P, δ^2H) and $\hat{\rho}_1^*$(P, δ^{18}O) were more likely to be overestimated for sites with ρ_1^*(P, δ^2H) and ρ_1^*(P, δ^{18}O) near zero, most likely a result of a site's weak amount effect that can become less significant and sometimes positive as a time series is aggregated. More data could improve estimates of $\hat{\rho}_1^*$. The *Discussion* provides further detail on methods for potentially improving statistical estimates at sites where errors were more apparent.

b. Evaluation of the Downscaled Tracer Realizations

The average of each ensemble's means ($\overline{\hat{\mu}_1}$) and standard deviations ($\overline{\hat{\sigma}_1}$) for each isotope ratio and the Pearson correlation coefficients ($\overline{\hat{\rho}_1}$) between precipitation amount and its corresponding isotope ratios were compared to the observed daily statistics at each site before

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

applying the residual correction (Figure 4). The downscaled time series with the most accurate $\overline{\hat{\rho}_1}$ were calculated when the method was applied to a weekly time series. After applying a residual correction on each realization in an ensemble, the residual corrected downscaled series accurately captured the μ_1, σ_1, and ρ_1 (Figure 5), though slightly altered R^2 values. The R^2 between μ_1 and $\overline{\hat{\mu}_1}$ and σ_1 and $\overline{\hat{\sigma}_1}$ were similar for the original downscaled (Figure 4.a-d) and residual corrected ensembles (Figure 5.a-d). The residual correction increases the R^2 between ρ_1 and $\overline{\hat{\rho}_1}$ (Figure 4.e-g, Figure 5.e-g), especially when it is applied on a downscaled weekly series. For a downscaled weekly series, the R^2 of $\rho_1(P, \delta^2 H)$ and $\overline{\hat{\rho}_1}(P, \delta^2 H)$ and $\rho_1(P, \delta^{18} O)$ and $\overline{\hat{\rho}_1}(P, \delta^{18} O)$ increased from 0.88 to 0.93 with a residual correction. For applications where model outputs are directly compared to observation datasets, a residual correction should be applied to generate tracer ensembles which are comparable to the coarser resolution observed values. The average bias between the downscaled and observed time series means and standard deviations were summarized in Table 1. The residual correction on the downscaled ensembles reduced bias in the standard deviations, but had little effect on the means. The *Discussion* provides further detail on potential methods for adding informative covariates (e.g. air temperature) to the downscaled time series estimates at sites where errors were more apparent.

c. Conserved Processes with the Method

An analysis of the observed time series demonstrates strong autocorrelation; when the seasonal signal is removed, the observed autocorrelation is nearly all removed (Figure 6). In fact, the median autocorrelation of the observed time series stochastic signals falls below 5 % after 3-day lags and are approximately zero at 4-day lags, supporting the assumption that the sinusoidal function adequately described the deterministic component and the residual was stationary (i.e.

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

white noise). The Shapiro-Wilk and the D'Agostino's K^2 normality tests suggest that we could not reject the assumption of normality in the weekly, biweekly and monthly time series (p-value > 0.05). Histograms of the stochastic signals for both isotope ratios across all 27 sites are provided in the *Supplemental Materials* (Figure S3). Next, we calculated autocorrelations from 1- to 20-day lags of the residual corrected downscaled ensembles. The autocorrelations mimicked the observed temporal trends and memory of the daily time series. Autocorrelations for δ^2H and δ^2H^* (Figure 6) and are highly correlated with trends observed in the autocorrelations for $\delta^{18}O$ and $\delta^{18}O^*$ (refer to *Supplemental Materials*, Figure S4). Based on the results from the autocorrelation analysis and normality tests, we concluded the addition of the seasonal signal to the generated stochastic time series captured the large majority of the observed tracer memory in the system.

The means and standard deviations in *d*-excess were accurately captured in the resulting downscaled time series (Figure 7). At each site, *d*-excess was calculated for the observed daily series and each ensemble from the downscaled weekly, biweekly and monthly time scales. The downscaled *d*-excess was over-estimated for the three sites with lowest observed *d*-excess, indicating potential effects to the downscaling method's performance when precipitation is predominantly composed of evaporated waters. These *d*-excess estimates provide a metric for evaluating the downscaled series relative to the GMWL and increases the applicability of this method for tracing meteorological forcing variables and their constituents through modeling environments. Alternative downscaling approaches that independently model δ^2H and $\delta^{18}O$ may not preserve *d*-excess signals and thus would provide precipitation predictions that should not be used in simulations that leverage the information provided by dual-isotope analyses.

4. Discussion

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

a. Method Evaluation for Select Site Characteristics

We compared the absolute error, calculated by taking the absolute value of the mean of the downscaled ensembles minus the observed mean, to the site's latitude, calculated rainfall frequency (λ), and total length of the time series in years (Figure 8). The largest absolute errors of the mean resulted from downscaling calculations that used monthly aggregated data, yielding average (+/- standard deviation) absolute errors of 2.26 ‰ (2.54 ‰) for δ^2H and 0.33 ‰ (0.35 ‰) for δ^{18}O. Linear regressions between site latitude and absolute errors in the means (derived from monthly, biweekly, and weekly ensembles) showed no strong correlations, suggesting that performance may be partially climate independent (Figure 8). Absolute errors were also not related to the strength of the seasonal isotopic variation, nor were they related to the overall variability in isotopic composition (as quantified by the standard deviation; Figure 9). Alternatively, a weak, but significant relationship was observed between absolute error and λ (R^2 = 0.25 and p-value = 0.0001 for δ^2H, R^2 = 0.23 and p-value = 0.0002 for δ^{18}O) and average recorded precipitation amount (R^2 = 0.12 and p-value = 0.009 for δ^2H, R^2 = 0.13 and p-value = 0.0003 for δ^{18}O) for downscaled weekly ensembles, but not for downscaled biweekly or monthly ensembles (Figures 8 and 9). Although not a site characteristic, time-series length significantly influenced absolute errors of downscaled biweekly and monthly ensembles. Longer time series spanning many years support better accounting for interannual variability and removing potential biases towards certain seasons. Nonlinear effects (e.g., continentality (Dansgaard 1964, Rozanski et al., 1993)) may be contributing to relatively high absolute errors, especially at the subtropics and mid-latitudes (Figure 8.a-b). When applying the downscaling method to datasets from these regions, one can adapt the copula framework to account for other influential site-specific characteristics (refer to *Section 2.b).*

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

b. Method Adaptation for Broader Applications

In this study, weekly, biweekly, and monthly data were used to generate daily observations, but more sophisticated applications could potentially be supported by different datasets. As a general rule, the deterministic time series component can be more accurately estimated with increased tracer sampling frequencies (Figure 8.c-d) and samples collected over longer time frames (Figure 8.e-f). Accurately representing the deterministic component increases the likelihood of a downscaled synthetic time series effectively representing the underlying seasonal patterns and interannual variability at a site. Depending on the application, one may increase or decrease the temporal downscaling intervals beyond daily or 12-week timescales. While not evaluated in this study, one could predict sub-daily datasets with appropriate observation datasets or known statistical properties (i.e., mean, standard deviation, covariance structure of precipitation and its tracer composition) of a site at sub-daily scales (e.g., diurnal cycle).

Theoretically, the downscaling methods used in this study can be expanded to higher dimensions and account for other tracer covariates including site conditions such as air temperature and relative humidity. At sites where the method under or overestimates the site statistics, other meteorological variables, such as air temperature, may correlate more strongly with isotope signals than precipitation amount. To do this, one needs to increase the number of covariates accounted for and the matrix dimensions within the copula framework (refer to *Supplemental Materials (i) Definition of a Gaussian Copula).* In these instances, adding more known dimensions to Equations 6 and 7 will incorporate additive information into the generated downscaled time series. Including additional known covariates within the copula framework may improve the representation of nonlinear effects at sites in the subtropics and mid-latitudes if meteorological variables (e.g.,

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

relative humidity, air temperature) are highly correlated with changes in tracer concentrations (Figures 8.a-b).

Not only would a downscaled time series facilitate running more detailed models that improve process understanding, but they also allow for better tracking of uncertainties associated with inferences drawn from those models. We compared the mean of the observed biweekly series and the mean of the downscaled biweekly ensemble aggregated to biweekly time scales using Eq. 2. The absolute error of the mean across all sites was 0.90 ‰ for δ^2H and 0.14 ‰ for δ^{18}O. This suggests models using downscaled tracers would mimic temporal trends observed at biweekly time scales, while also tracking processes and uncertainties only discernible at finer time scales. As expected, when the residual corrected downscaled biweekly ensemble was aggregated to biweekly time scales, the absolute error of the mean was approximately zero. To evaluate how the downscaling method compared to a naive downscale with no high-frequency statistical information, we created a daily time series where all precipitation events that occurred within each 14-day interval had the same isotopic composition equal to the observed biweekly values. The absolute error of the mean across all sites (+/- standard deviation) was 2.74 ‰ (2.24 ‰) for δ^2H and 0.39 ‰ (0.31 ‰) for δ^{18}O, which was higher than the absolute error of the mean calculated for all the downscaled biweekly ensembles (1.69 ‰ (1.61 ‰) for δ^2H and 0.23 ‰ (0.24 ‰) for δ^{18}O).

Due to limited data, all of the above analyses used the entire dataset to calculate the statistics, fit the models and apply the downscaling method. At sites with more than 5 years of data, we used the first 4 years to build a downscaling model to apply on the 5th year's precipitation time series. We generated an ensemble of 100 downscaled δ^2H and δ^{18}O time series at each site and compared it to the observed δ^2H and δ^{18}O from the 5th year of the time series. Based on

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

training and testing sizes, eight sites were used in this analysis and the absolute error of the mean (+/- standard deviation) for δ^2H was 4.80 ‰ (3.17 ‰), 4.89 ‰ (3.26 ‰) and 5.52 ‰ (3.50 ‰) downscaled from weekly, biweekly, and monthly series, respectively. The absolute error of the mean (+/- standard deviation) for $\delta^{18}O$ was 0.79 ‰ (0.59 ‰), 0.78 ‰ (0.63 ‰) and 0.85 ‰ (0.67 ‰) downscaled from weekly, biweekly, and monthly series, respectively. Based on these promising results, our downscaling method could be built using several years of precipitation data with a known concentration and then applied to years where only precipitation amount is available.

This method can be broadly applied to produce ensembles of downscaled datasets for various geochemical modeling applications. Ensembles decrease the risk of tying conclusions to one specific time series. The downscaled ensembles can be generated using the same statistics (like shown here) or multiple ensembles can be generated with varying statistical properties. Examples of different ensembles include time series generated from downscaled statistics estimated from different aggregation intervals (e.g. weekly and biweekly), employing a non-Gaussian copula framework (e.g. Gumbel copula, Extreme-value copula) to populate a conditioned stochastic signal's time series, and increasing dimensions of the copula framework by including additive meteorological variables (e.g. air temperature). Correspondingly, the geochemical tracer ensembles could be used for model selection and with numerous model and parameter sensitivity and uncertainty analyses. Ensembles could be useful in developing frameworks for model-data fusion by merging observational data with model outputs to improve model quality and characterize its uncertainty.

This downscaling approach could be extended across large spatial extents for use in global isotopic models or empirically based geographic simulations to represent sites with limited or no high-frequency observations available. To do this, one could generate downscaled geochemical

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

tracers correlating with precipitation inputs at the grid-scale. Lastly, the methodology can be applied to other geochemical tracers for understanding site-specific dynamics (e.g. chemical leaching, sediment transport and loading) or climatological applications (e.g. nitrogen deposition, carbon sequestration).

5. Conclusions

This statistical downscaling method generates datasets that maintain informative site-specific correlation structures between covariates and the geochemical tracer and retains the statistical properties of underlying processes (e.g., *d*-excess, amount effects). By modeling hydrologic dynamics using downscaled tracers, researchers can enhance understanding of physical processes without collecting fine temporal in-situ data. While an individual realization of this downscaling approach may generate reasonable estimates of true high-frequency values, iterating analyses using an ensemble of realizations allows for uncertainties in generated time series to be propagated through subsequent modeling and tracer-based analyses. The method is sufficiently general and can be applied for a variety of applications to generate downscaled ensembles for use in meteorological and hydrometeorological models to evaluate model performance, investigate system processes across spatial scales and is additive to model-data fusion frameworks.

Acknowledgements

The authors acknowledge the support of the United States National Science Foundation (DEB-1802885, DEB-1802880 and AGS-1954660) and thank the anonymous reviewers for their time and effort spent reviewing this manuscript. The authors state no conflict of interest.

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

Data Availability Statement

Python code is provided in the *Supplemental Material* to generate geochemical synthetic time series based on the user's site-specific time series statistics. The code is intended to be easily adaptable to higher dimensions or other user specific applications. Additional materials can be made available upon request.

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TABLES

Table 1. Average bias (predicted - observed statistic) (+/- standard deviation) for downscaled and *residual corrected downscaled* ensembles

	Bias in the Means (‰)				**Bias in the Standard Deviations (‰)**			
	$\boldsymbol{\delta^2\mathrm{H}}$	$\boldsymbol{\delta^2 H}$	$\boldsymbol{\delta^{18}\mathrm{O}}$	$\boldsymbol{\delta^{18} O}$	$\boldsymbol{\delta^2\mathrm{H}}$	$\boldsymbol{\delta^2 H}$	$\boldsymbol{\delta^{18}\mathrm{O}}$	$\boldsymbol{\delta^{18} O}$
Weekly	-0.10 (1.39)	*0.01* *(1.79)*	-0.02 (0.20)	*-0.03* *(0.26)*	0.02 (2.38)	*1.95* *(3.30)*	0.004 (0.34)	*0.25* *(0.46)*
Biweekly	-1.00 (2.20)	*-0.68* *(3.50)*	-0.16 (0.30)	*-0.12* *(0.46)*	1.04 (3.95)	*2.76* *(4.14)*	0.17 (0.56)	*0.39* *(0.59)*
Monthly	-1.43 (3.08)	*-0.13* *(5.86)*	-0.23 (0.43)	*-0.05* *(0.74)*	2.20 (8.75)	*3.28* *(8.81)*	0.28 (1.12)	*0.43* *(1.12)*

FIGURES

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

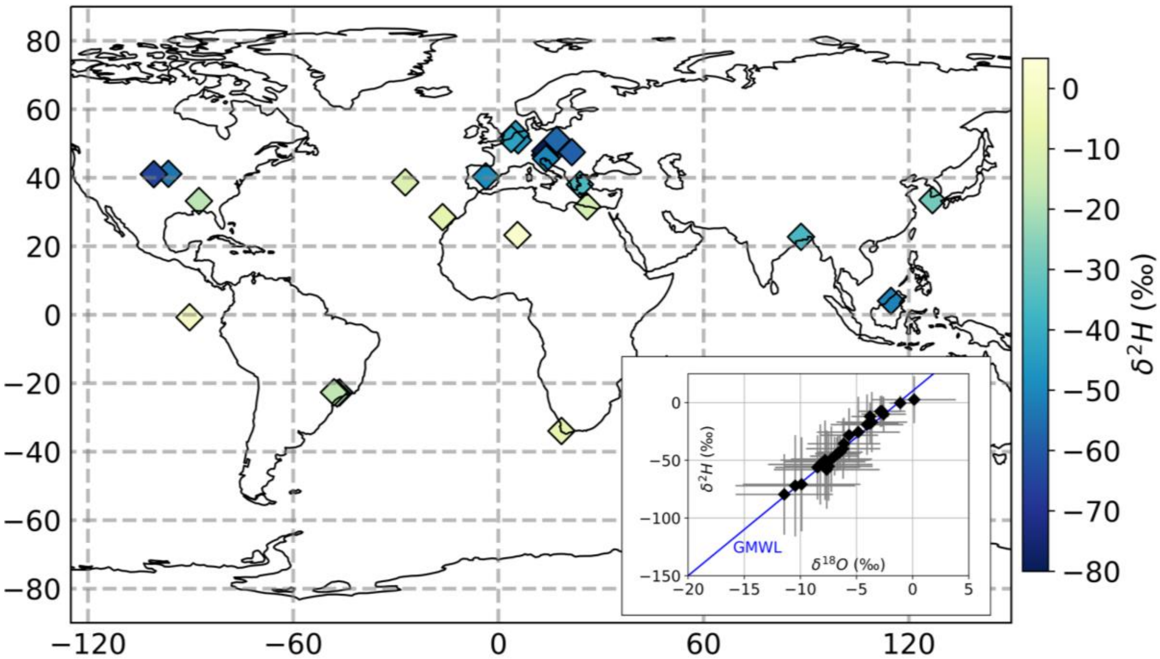

Figure 1. The large map displays the 27 GNIP site locations and their average $\boldsymbol{\delta^2 H}$ precipitation measurements. The smaller figure is a dual isotope plot with the mean and standard deviations of all daily precipitation stable water isotope measurements (δ^2H, δ^{18}O) at the 27 GNIP sites. The Global Meteoric Water Line (GMWL) is included in the subplot. Refer to Table S1 in the *Supplemental Material* for more site-specific characteristics.

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

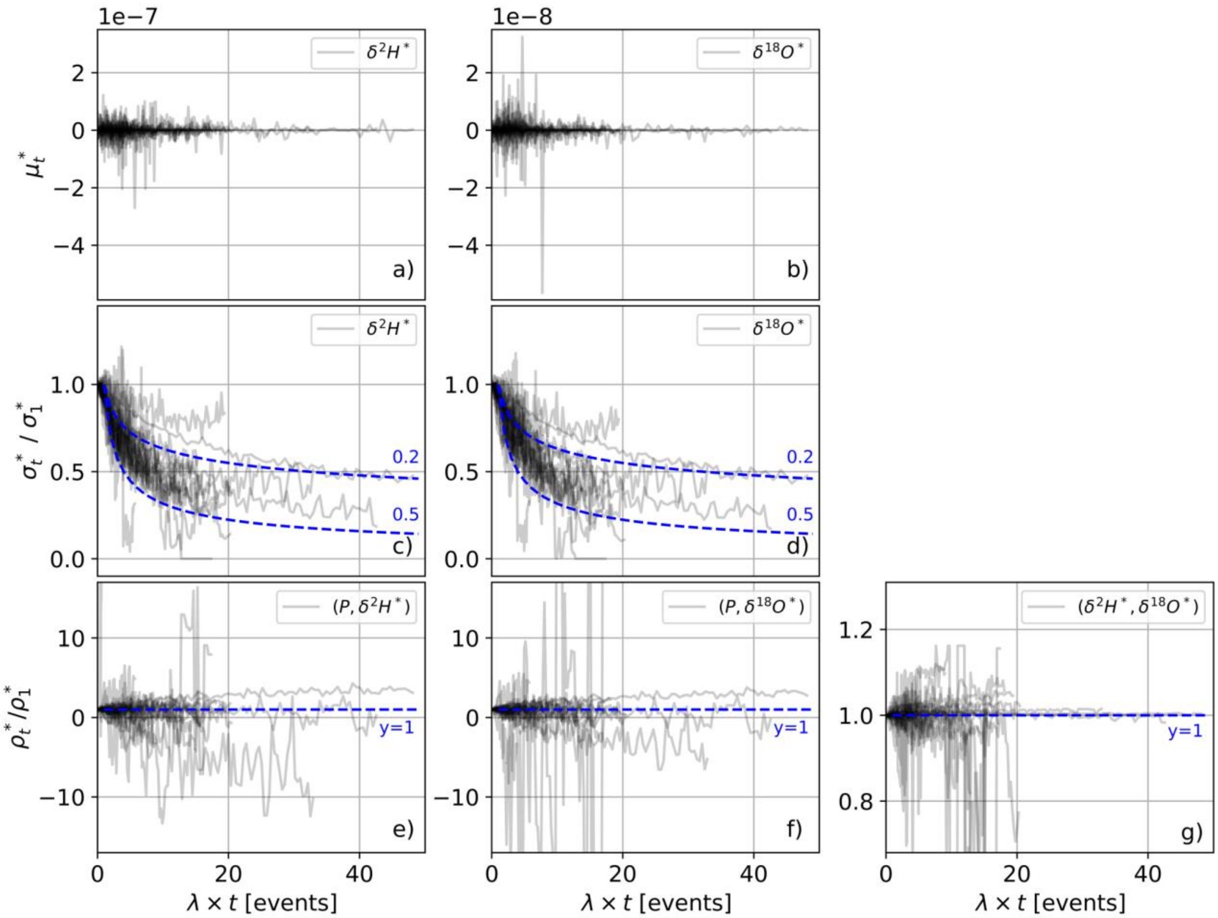

Figure 2. The x-axis is $\boldsymbol{\lambda}$ (recorded events / number of days in the time series) multiplied by daily to 12-week aggregation intervals (days) and the y-axes were the deviations for each of the 27 sites in their stochastic time series a,b) means ($\boldsymbol{\mu_t^*}$; note the scale of the y-axis), c,d) standard deviations at *t*-day ($\boldsymbol{\sigma_t^*}$) divided by the daily standard deviation ($\boldsymbol{\sigma_1^*}$) with blue dashed lines at $(\boldsymbol{\lambda n})^{\mathbf{0.5}}$ and $(\boldsymbol{\lambda n})^{\mathbf{0.2}}$, and e-g) Pearson correlation coefficients at *t*-day divided by daily ($\boldsymbol{\rho_t^*/\rho_1^*}$) with blue dashed lines at y-axis = 1. Refer to *Supplemental Materials* (Fig. S1) for larger ranges in y-axis values for ($\boldsymbol{\rho_t^*/\rho_1^*}$).

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

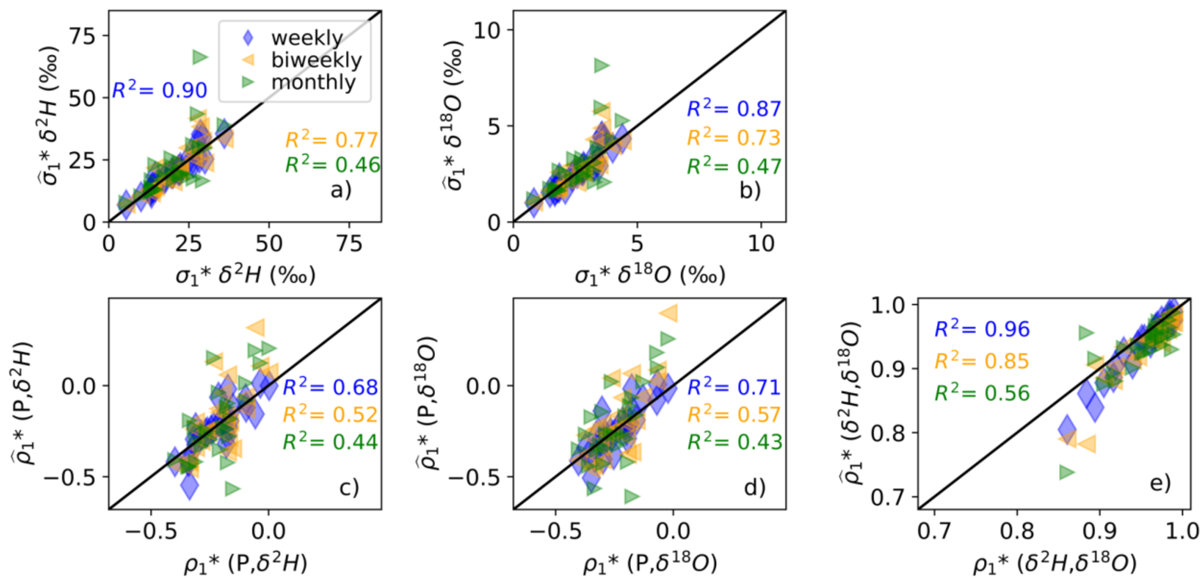

Figure 3. The estimated standard deviations (a,b) and Pearson correlation coefficients (c-e) of the stochastic signal from downscaled weekly, biweekly and monthly time series compared to the observed daily stochastic statistics. Each data point is one site location and the black lines are the 1:1 lines. The means were not shown because they are approximately zero (refer to Figure S2.a,b).

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

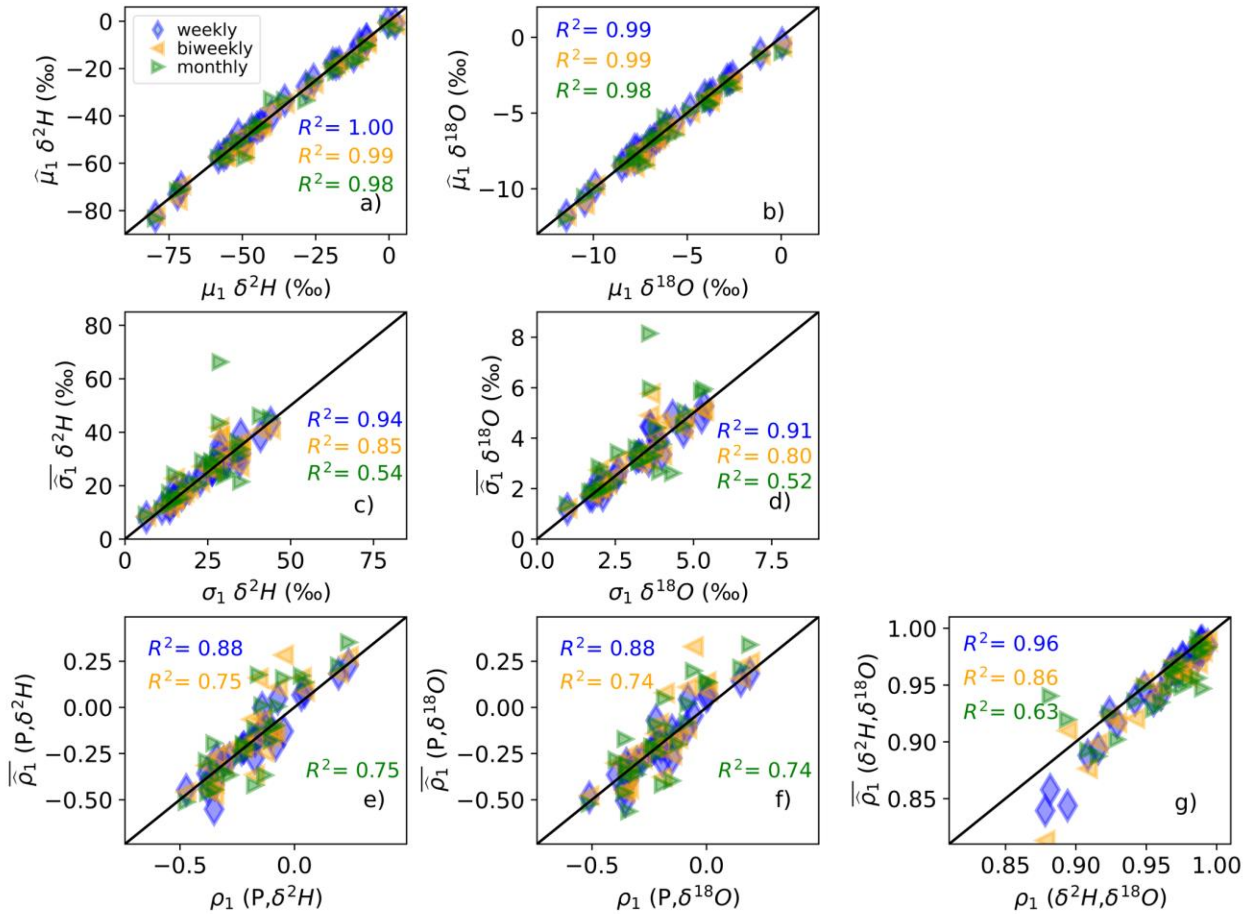

Figure 4. The average means (a,b), standard deviations (c,d) and Pearson correlation coefficients (e-g) of the downscaled ensembles from the weekly, biweekly and monthly time series compared to the observed daily site statistics. Each data point is one location and the black lines are the 1:1 lines.

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

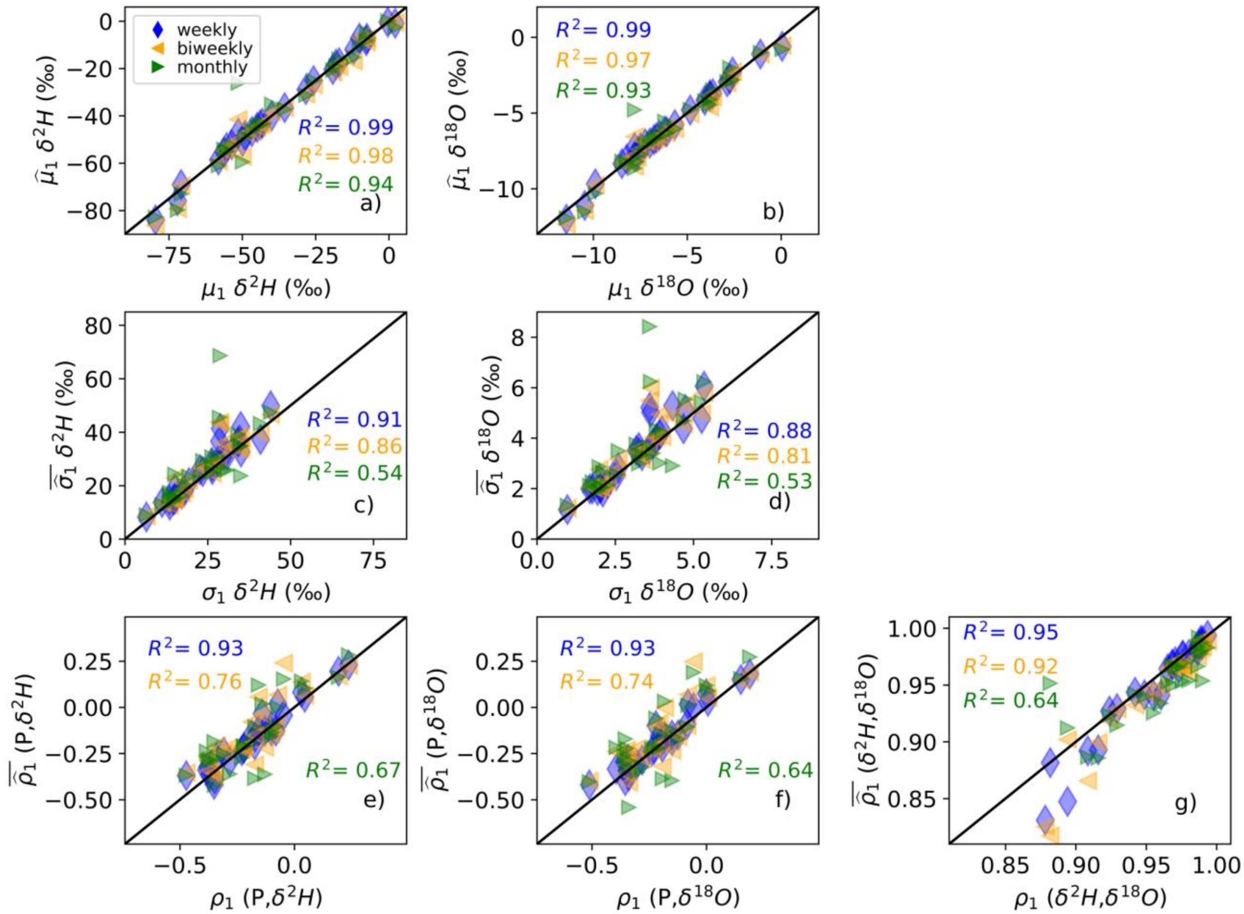

Figure 5. The average means (a,b), standard deviations (c,d) and Pearson correlation coefficients (e-g) of the residual corrected downscaled ensembles from the weekly, biweekly and monthly time series compared to the observed daily site statistics. Each data point is one location and the black lines are the 1:1 lines.

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

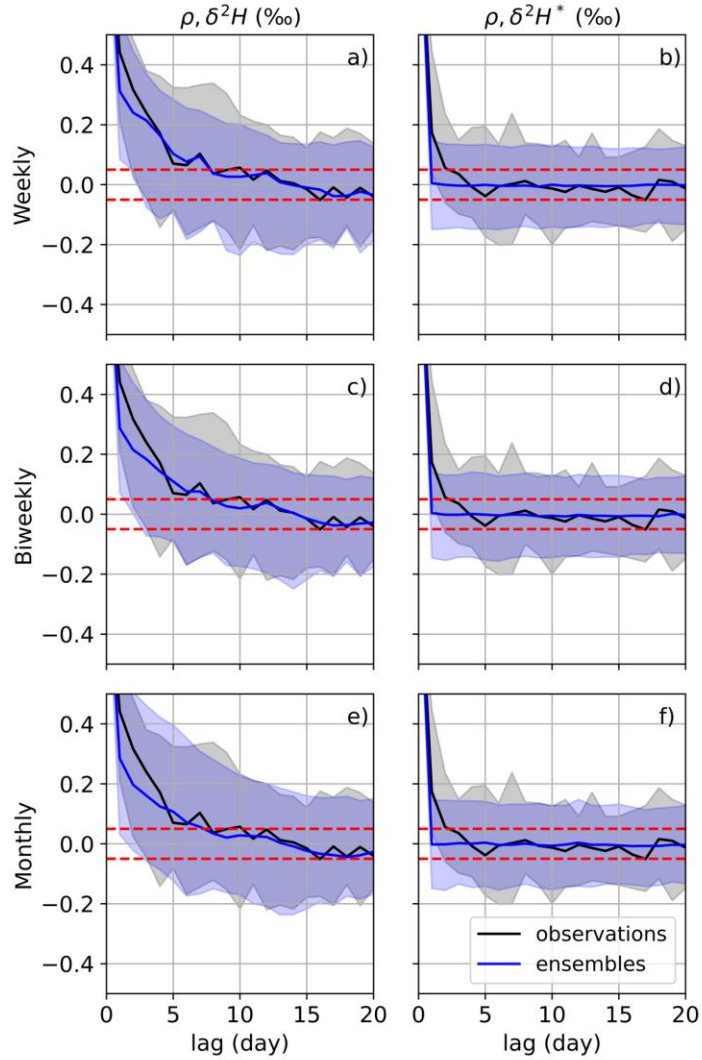

Figure 6. a,c,e) Median autocorrelation of the observed daily $\boldsymbol{\delta^2 H}$ datasets and the daily residual corrected ensembles (solid lines). b,d,f) Median autocorrelations of the $\boldsymbol{\delta^2 H}^*$ stochastic signals for the observations and the downscaled ensembles. The 5th to 95th percentiles of the observed and ensemble autocorrelations are represented as shaded regions. Horizontal red dashed line indicates where $\boldsymbol{\rho}$ is +/-5%.

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

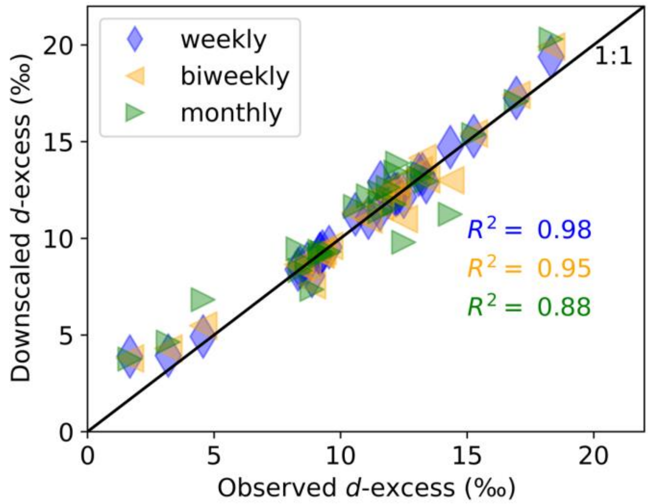

Figure 7. The average *d*-excess of the residual corrected downscaled ensemble at each site location compared to the average observed *d*-excess. Each data point is one site location and the black line is the 1:1 line.

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

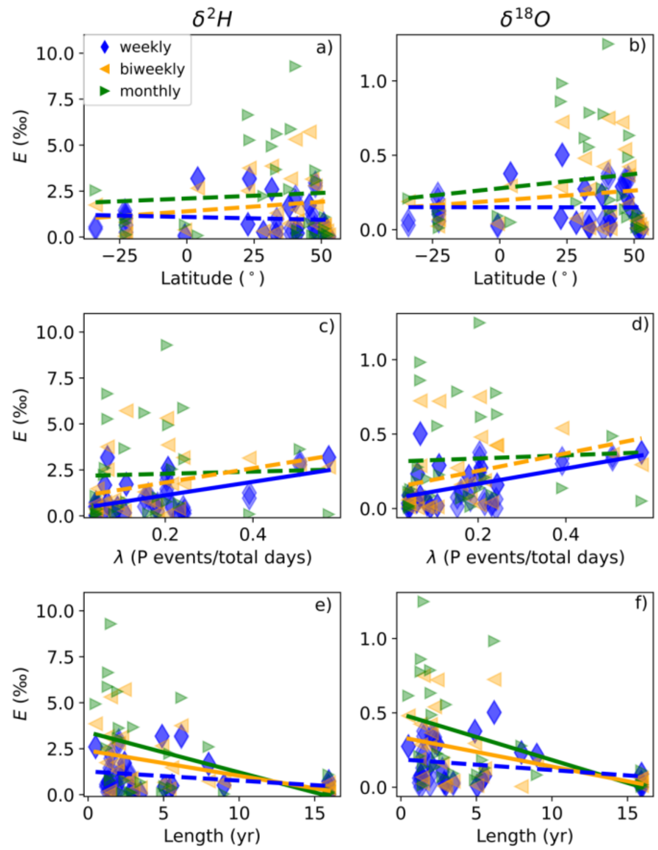

Figure 8. Absolute error (E) of the residual corrected ensemble means compared to various site-specific characteristics: a,b) latitude, c,d) $\boldsymbol{\lambda}$, and e,f) total length of the time series. Each data point is one site location, dashed lines represent p-values > 0.05, and solid lines represent p-values < 0.05.

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.

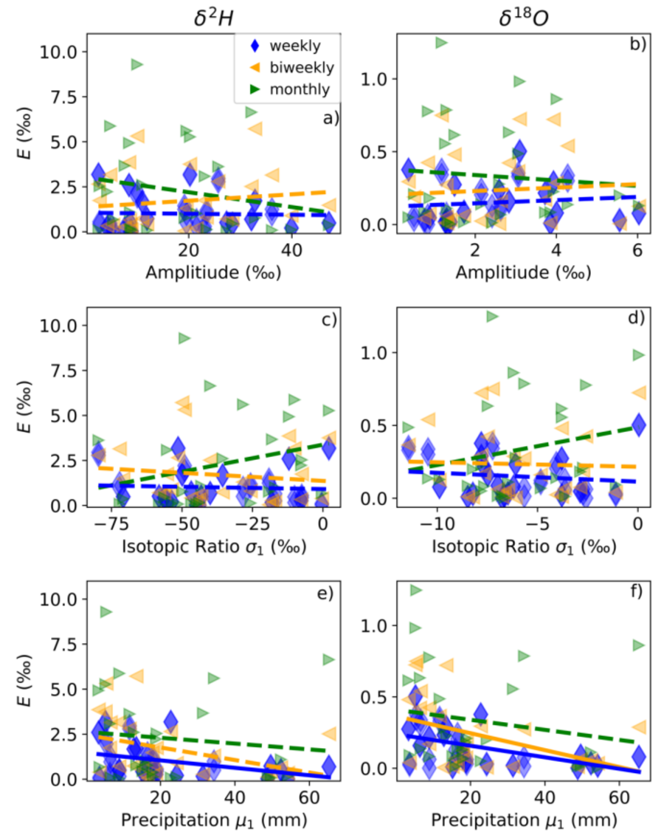

Figure 9. Absolute error (E) of the residual corrected ensemble means compared to various site-specific characteristics: a,b) the sinusoidal function's estimated amplitude, c,d) standard deviation of each isotope ratio and e,f) average daily precipitation. Each data point is one site location, dashed lines represent p-values > 0.05, and solid lines represent p-values < 0.05.

Accepted for publication in *Journal of Hydrometeorology*. DOI 10.1175/JHM-D-20-0142.1.