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Analysis of long-term stable isotopic composition in German precipitation



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SUMMARY

Stable water isotopes in precipitation (¹⁸O and ²H) have been frequently used as environmental tracers to understand processes and timescales in hydrology and climate research. Capturing changes of isotopic composition over time and investigating long-term processes requires long-term data set analysis. In Germany, we have one of the world's densest national networks for long-term isotopic analysis of precipitation covering up to 36 years of time series at 28 locations. These data were used to identify the average as well as the temporal evolution of isotopic composition in German precipitation and how it is related to meteorological and geographical parameters. We found that individual Local Meteoric Water Lines (LMWL) and the long-term averages of δ^{18} O and δ^{2} H depend on latitude and elevation. More variable isotopic compositions and more enriched averages were found at the coast compared to more stable compositions and depleted averages in the South, South-East and at higher elevations. This continentality effect was strongly influenced by seasonal isotope-temperature dependencies. Removing the seasonality and looking at the changes over time compared to long-term averages indicate similar patterns for temperatures and ¹⁸O at some locations. We concluded that temperature and isotopes are in equilibrium in inland air masses only. The trend in temperature evolution was consistent on the national level, and temperature increases were observed in almost all stations. In contrast, temporal patterns of ¹⁸O revealed different patterns and increases were only observed in 20 out of 28 locations. Therefore, changes in isotopic composition in precipitation are not only influenced by large scale processes (i.e. temperature) but also by local factors which need to be further investigated.

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1. Introduction

The distributions and average values of stable isotopes of the water molecules, oxygen-18 (¹⁸O) and deuterium (²H), in precipitation are a valuable tool to study spatial and temporal processes in climate research and hydrology. It has become increasingly apparent that long-term data sets are required to fully understand processes of the water cycle in terms of climate variability and climate change. Therefore, long-term data sets of stable isotopes in precipitation must be investigated in more detail to adequately account for spatial and temporal changes and influencing factors. This is the scope of the present study.

Both, ¹⁸O and ²H in precipitation vary in space and time depending on the source of water as well as the conditions (humidity and temperature) during cloud formations and rain

out. The absolute ratio and seasonal fluctuations in the distribution of water isotopes in precipitation measured at an individual location reflect its local temperature, continentality, latitude and altitude (Clark and Fritz, 1997). These geographical effects can be well observed in the mean isotope compositions on the global (Bowen et al., 2005; Rozanski et al., 1993; Terzer et al., 2013), regional (Gat and Carmi, 1970; Harvey and Welker, 2000; Tian et al., 2007), national (Dutton et al., 2005; Liu et al., 2010a,b; Longinelli and Selmo, 2003; Schotterer et al., 2010; Schürch et al., 2003), and local scale (Datta et al., 1991). These effects rely on temperature dependent equilibrium and on kinetic nonequilibrium fractionation (e.g. due to humidity). Due to equilibrium fractionation processes in the atmosphere, there exists an empirical average linear relationship of $\delta^2 H$ and $\delta^{18} O$ water isotopes in precipitation ($\delta^2 H = 8 \cdot \delta^{18} O + 10$) (Craig, 1961; Dansgaard, 1964). This relationship is referred to as the Global Meteoric Water Line (GMWL), and reflects depleted isotope contents (lower ratios) in colder regions/seasons and more enriched isotopes contents (higher ratios) in warmer regions/seasons. The





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isotopic ratio is dependent on local climatic conditions of condensation and rainout, so the relationship between δ^2 H and δ^{18} O may differ from the global distribution resulting in Local Meteoric Water Lines (LMWL) with, for example, lower slopes in regions with high fractions of evaporation during rainout (Clark and Fritz, 1997). Another measure to identify site specific changes in isotope signatures is the Deuterium excess (d-Excess), which is defined as $d = \delta^2 H - 8 \cdot \delta^{18}O$ and is 10‰ on a global average (Craig, 1961). It may vary locally due to kinetic fractionation processes, being low in regions with high humidity and vice versa (Dansgaard, 1964), and be used to indicate different origins of the water (e.g. tropic or polar) or fractions of re-evaporated water (e.g. marine or continental).

Due to the spatial and temporal variability of δ^2 H and δ^{18} O in precipitation, these isotopes have been used to identify sources of water such as atmospheric moisture (Liu et al., 2010b; Sjostrom and Welker, 2009; Vachon et al., 2010; Welker, 2000), groundwater recharge (Abbott et al., 2000; Adomako et al., 2010; Blasch and Bryson, 2007), and plant water uptake (Brooks et al., 2010). In other hydrological applications, water isotopes were used to determine partitioning between event and pre-event water in streamflow (Klaus and McDonnell, 2013), catchment transit times (McGuire and McDonnell, 2006), to constrain precipitation runoff models (Fenicia et al., 2008; Seibert and McDonnell, 2002) or unsaturated flow and transport models (Stumpp et al., 2009).

The relationships between water isotope ratios, and meteorological and geographical parameters, allows for the production of regional (Longinelli and Selmo, 2003; Vachon et al., 2010) and global isotope maps (Bowen et al., 2005; Bowen and Revenaugh, 2003; Bowen and Wilkinson, 2002; Terzer et al., 2013). Similarly, empirical, linear relationships between temperature and δ^{18} O or δ^{2} H in precipitation (Dansgaard, 1964) have been used to reconstruct paleoclimatic conditions from stable isotope analysis in tree rings (Masson-Delmotte et al., 2005) and ice cores (Barbante et al., 2006; Jouzel et al., 1997; Rozanski et al., 1997). Clearly, stable isotope ratios and relationships are not only important to understand the past trends (Lykoudis and Argiriou, 2011), but have potential to show ongoing changes in the climate system (Rozanski et al., 1992).

Information about water isotopes in precipitation is also of benefit to climate circulations models (Joussaume et al., 1984; Lee et al., 2007; Sturm et al., 2005) and hydrological process and modelling studies because isotopes can be used as environmental tracers to calibrate and validate such models. Climate models require high resolution data for validation typically, with grid sizes ranging from 0.2° to 4° (Taylor et al., 2012).

Besides high resolution data, modelling studies require high quality and continuous long-term data of climatic and isotopic parameters. To date, few such data are available worldwide for continuous long-term analysis with high spatial resolution. Most of these data are embedded in the Global Network for Isotopes in Precipitation (GNIP), which is a joint program of the International Atomic Energy Agency (IAEA) and the World Metrological Organisation (WMO), and are available online (http://wwwnaweb.iaea.org/napc/ih/IHS_resources_isohis.html#wiser). Out of this dataset, Rozanski et al. (1992) investigated temporal trends of isotopes in precipitation at twelve stations world-wide over 15-30 years. Most notably, Rozanski et al. (1992) showed that four stations in Switzerland increased in both temperature and δ^{18} O compared to the long-term annual average at the end of the study period in the late 80s. However, it remains unknown whether these trends are periodic signals or long-term effects of generally increased (or decreased) isotope values in regions with higher (or lower) air temperatures attributed to climate change. Furthermore, it is still unclear how and how fast regional climatic variability is reproduced in the isotope values and also if a response to any climatic changes can be identified in the isotopic composition. Such analysis requires continuous long-term datasets with high spatial resolution, which are available on a national level for Germany. This unique dataset includes monthly water isotopes in precipitation of 14–36 years from 28 meteorological stations in Germany which equates to an average spatial resolution of $0.5^{\circ} \times 0.5^{\circ}$.

The objective of this study is to (1) present the data to the public and, particularly, to use these continuous long-term data sets (2) to identify the national distribution of average water isotopes in German precipitation, (3) to relate the information of the individual isotopic distributions to geographical and meteorological parameters and (4) to investigate the variability of isotopic composition and its influencing factor temperature and amount of precipitation over time. Results of this study can be further used for statistical trend analysis, to improve our knowledge on controlling factors of regional and local parameters on the average isotopic composition, for identifying impacts and regions of climate change, for large scale hydrological modelling and for the improvement of climate models.

2. Methods

Monthly or event based samples of precipitation were collected at 28 meteorological stations across Germany between 1974 and present (Fig. 1). Most of these stations (16) are sampled on a monthly basis and are part of the GNIP since 1978. Since 1997, monthly samples of nine stations and precipitation event based samples of three stations operated by Germany's National Meteorological Service (Deutscher Wetterdienst, DWD) have been collected. A list of the stations and sampling records used in this study are given in Table 1.

The ratio of isotopes (R_{sample}) is given in the delta notation as δ -value (%), which is the relative deviation of the sample from a standard ($R_{standard}$):

$$\delta[\%] = \frac{R_{\text{Sample}} - R_{\text{Standard}}}{R_{\text{Standard}}} \cdot 1000 \tag{1}$$

V-SMOW (Vienna-Standard Mean Ocean Water) was used as the reference standard for both ¹⁸O and ²H. Positive δ -ratios indicate enrichment of the heavier isotope relative to the standard; negative ratios indicate depletion. More details about principles of isotope hydrology and measurement techniques are given by the IAEA (1983) and Leibundgut et al. (2009). Precipitation samples were analysed for $\delta^{18}O$ (±0.15‰) and $\delta^{2}H$ (±1‰) ratios without any pre-treatment of the samples using dual-inlet mass spectrometry (at Helmholtz Zentrum München, Germany). Weighted average monthly values and long-term averages ($\overline{\delta}$) were calculated from event based and monthly precipitation amount (*P*) and isotope values (δ), respectively:

$$\overline{\delta}[\%] = \frac{\sum_{i=1}^{N} \delta_i \cdot P_i}{\sum_{i=1}^{N} P_i}$$
(2)

The amount of precipitation was recorded at each station. Missing precipitation data and mean monthly temperatures were derived for each station from Germany's National Meteorological Service (DWD). These data were not available for the entire observation time at the stations Emmerich, Koblenz, Neubrandenburg and Weil am Rhein. Accordingly, a complete and unique long-term record of water isotopes in precipitation and meteorological parameters (amount of precipitation and surface air temperature) is available on a national level for 24 stations.

From the $\delta^2 H - \delta^{18}O$ relationships, Local Meteoric Water Lines (LMWL) were calculated at the national (across Germany) scale as well as at local scale for each location individually. Further, long-term averages were calculated for $\delta^2 H$, $\delta^{18}O$ and d-Excess and

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