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Molecular diffusion of stable water isotopes in polar firn as a proxy for past temperatures

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Abstract

Polar precipitation archived in ice caps contains information on past temperature conditions. Such information can be retrieved by measuring the water isotopic signals of δ^{18} O and δ D in ice cores. These signals have been attenuated during densification due to molecular diffusion in the firn column, where the magnitude of the diffusion is isotopologue specific and temperature dependent. By utilizing the differential diffusion signal, dual isotope measurements of δ^{18} O and δ D enable multiple temperature reconstruction techniques. This study assesses how well six different methods can be used to reconstruct past surface temperatures from the diffusion-based temperature proxies. Two of the methods are based on the single diffusion lengths of $\delta^{18}O$ and δD , three of the methods employ the differential diffusion signal, while the last uses the ratio between the single diffusion lengths. All techniques are tested on synthetic data in order to evaluate their accuracy and precision. We perform a benchmark test to thirteen high resolution Holocene data sets from Greenland and Antarctica, which represent a broad range of mean annual surface temperatures and accumulation rates. Based on the benchmark test, we comment on the accuracy and precision of the methods. Both the benchmark test and the synthetic data test demonstrate that the most precise reconstructions are obtained when using the single isotope diffusion lengths, with precisions of approximately 1.0 °C. In the benchmark test, the single isotope diffusion lengths are also found to reconstruct consistent temperatures with a root-mean-squaredeviation of $0.7 \,^{\circ}$ C. The techniques employing the differential diffusion signals are more uncertain, where the most precise method has a precision of 1.9 °C. The diffusion length ratio method is the least precise with a precision of 13.7 °C. The absolute temperature estimates from this method are also shown to be highly sensitive to the choice of fractionation factor parameterization.

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1. INTRODUCTION

Polar precipitation stored for thousands of years in the ice caps of Greenland and Antarctica contains unique information on past climatic conditions. The isotopic composition of polar ice, commonly expressed through the δ notation has been used as a direct proxy of the relative

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depletion of a water vapor mass in its journey from the evaporation site to the place where condensation takes place (Epstein et al., 1951; Mook, 2000). Additionally, for modern times, the isotopic signal of present day shows a good correlation with the temperature of the cloud at the time of precipitation (Dansgaard, 1954, 1964) and as a result it has been proposed and used as a proxy of past temperatures (Jouzel and Merlivat, 1984; Jouzel et al., 1997; Johnsen et al., 2001).

The use of the isotopic paleothermometer presents some notable limitations. The modern day linear relationship

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between δ^{18} O and temperature commonly referred to as the "spatial slope" may hold for present conditions, but studies based on borehole temperature reconstruction (Cuffey et al., 1994; Johnsen et al., 1995) as well as the thermal fractionation of the δ^{15} N signal in polar firn (Severinghaus et al., 1998; Severinghaus and Brook, 1999) have independently underlined the inaccuracy of the spatial isotope slope when it is extrapolated to past climatic conditions. Even though qualitatively the δ^{18} O signal comprises past temperature information, it fails to provide a quantitative picture on the magnitudes of past climatic changes.

Johnsen (1977) and Whillans and Grootes (1985) and Johnsen et al. (2000) set the foundations for the quantitative description of the diffusive processes the water isotopic signal undergoes in the porous firn layer from the time of deposition until pore close-off. Even though the main purpose of Johnsen et al. (2000) was to investigate how to reconstruct the part of the signal that was attenuated during the diffusive processes, the authors make a reference to the possibility of using the assessment of the diffusive rates as a proxy for past firn temperatures.

The temperature reconstruction method based on isotope firn diffusion requires data of high resolution. Moreover, if one would like to look into the differential diffusion signal, datasets of both δ^{18} O and δ D are required. Such data sets have until recently not been easy to obtain especially due to the challenging nature of the δD analysis (Bigeleisen et al., 1952; Vaughn et al., 1998). With the advent of present commercial high-accuracy, highprecision Infra-Red spectrometers (Crosson, 2008; Brand et al., 2009), simultaneous measurements of δ^{18} O and δ D have become easier to obtain. Coupling of these instruments to Continuous Flow Analysis systems (Gkinis et al., 2011; Maselli et al., 2013; Emanuelsson et al., 2015; Jones et al., 2017) can also result in measurements of ultra-high resolution, a necessary condition for accurate temperature reconstructions based on water isotope diffusion.

A number of existing works have presented past firn temperature reconstructions based on water isotope diffusion. Simonsen et al. (2011) and Gkinis et al. (2014) used high resolution isotopic datasets from the NorthGRIP ice core (NGRIP members, 2004). The first study makes use of the differential diffusion signal, utilizing spectral estimates of high-resolution dual δ^{18} O and δ D datasets covering the GS-1 and GI-1 periods in the NorthGRIP ice core (Rasmussen et al., 2014). The second study presents a combined temperature and accumulation history of the past 16,000 years based on the power spectral density (PSD hereafter) signals of high resolution δ^{18} O measurements of the NorthGRIP ice core. More recently, van der Wel et al. (2015) introduced a slightly different approach for reconstructing the differential diffusion signal and testing it on dual δ^{18} O, δ D high resolution data from the EDML ice core (Oerter et al., 2004). By artificially forwarddiffusing the δD signal the authors estimate differential diffusion rates by maximizing the correlation between the δ^{18} O and δD signal. In this work we attempt to test the various

approaches in utilizing the temperature reconstruction technique.

We use synthetic, as well as real ice core data sets that represent Holocene conditions from a variety of drilling sites on Greenland and Antarctica. Our objective is to use data sections that originate from parts of the core as close to present day as possible. By doing this we aim to minimize possible uncertainties and biases in the ice flow thinning adjustment that is required for temperature interpretation of the diffusion rate estimates. Such a bias has been shown to exist for the NorthGRIP ice core (Gkinis et al., 2014), most likely due to the Dansgaard and Johnsen (1969) ice flow model overestimating the past accumulation rates for the site. In order to include as much data as possible, approximately half of the datasets used here have an age of 9–10 ka. This age coincides with the timing of the early Holocene Climate Optimum around 5-9 ka (HCO hereafter). For Greenlandic drill sites, temperatures were up to 3 °C warmer than present day during the HCO (Dahl-Jensen et al., 1998). Another aspect of this study is that it uses water isotopic data sets of δ^{18} O and δ D measured using different analytical techniques, namely Isotope Ratio Mass Spectroscopy (IRMS hereafter) as well as Cavity Ring Down Spectroscopy (CRDS hereafter). Two of the data sets presented here were obtained using Continuous Flow Analysis (CFA hereafter) systems tailored for water isotopic analysis (Gkinis et al., 2011). All data sections are characterized by a very high sampling resolution typically of 5 cm or better.

2. THEORY

2.1. Diffusion of water isotope signals in firn

The porous medium of the top 60–80 m of firn allows for a molecular diffusion process that attenuates the water isotope signal from the time of deposition until pore closeoff. The process takes place in the vapor phase and it can be described by Fick's second law as (assuming that the water isotope ratio signal (δ) approximates the concentration):

$$\frac{\partial \delta}{\partial t} = D(t) \frac{\partial^2 \delta}{\partial z^2} - \dot{\varepsilon}_z(t) z \frac{\partial \delta}{\partial z}$$
(1)

where D(t) is the diffusivity coefficient, $\dot{\varepsilon}_z(t)$ the vertical strain rate and z is the vertical axis of a coordinate system, with its origin being fixed within the considered layer. The attenuation of the isotopic signal results in loss of information. However, the dependence of $\dot{\varepsilon}_z(t)$ and D(t) on temperature and accumulation presents the possibility of using the process as a tool to infer these two paleoclimatic parameters. A solution to Eq. (1) can be given by the convolution of the initial isotopic profile δ' with a Gaussian filter \mathcal{G} as:

$$\delta(z) = \mathcal{S}(z)[\delta'(z) * \mathcal{G}(z)]$$
⁽²⁾

where the Gaussian filter is described as:

$$\mathcal{G}(z) = \frac{1}{\sigma\sqrt{2\pi}} e^{\frac{-z^2}{2\sigma^2}},\tag{3}$$

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