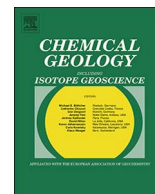




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Noble gas signatures in snow

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ABSTRACT

Noble gas concentrations in water are ideal probes to study surface and groundwater dynamics by providing indications of flow paths, connectivity between aquifers, and water residence times. Recent studies have pointed out anomalies in noble gas concentrations derived from groundwater in fractured systems, likely due to the presence of rapid infiltration and preferential flow paths. It has been suggested that such anomalies originate from conditions at high altitude when rainwater has had insufficient time to equilibrate with surface conditions. Potential sources also include snow, never previously investigated for its noble gas composition.

In order to document the noble gas signature in snow, noble gas concentrations and isotopic ratios were measured in samples collected between 2013 and 2016. Here, we outline a methodology for measuring noble gases in collected snow samples that involves a two-step procedure where He and Ne are measured independently from Ar, Kr and Xe.

Our results show that snow has elevated He concentrations with depleted concentrations of other noble gases with respect to air-saturated water (ASW). However, samples collected in 2013 show significant He and Ne depletion compared to those collected in 2014, 2015 and 2016. We suspect that, despite the well-controlled conditions of storage, the 2013 batch sample might have significantly re-crystallized, leading to a reduction in the characteristic diffusion length scale of the snow crystal structure. In addition, He and Ne concentrations display relatively low variability among all measured samples (< 14%), while Ar, Kr and Xe show large variability in their concentrations (> 40%). Our results confirm that He and Ne, which have small atomic radii, are likely dissolved within the ice/snow crystal lattice itself while the heavy noble gases (Ar, Kr and Xe) are likely accommodated by fluid inclusions, including air and quenched liquid water inclusions. Consequently, the smaller variability recorded in light noble gases may be due to the fact that He and Ne are hosted within plentiful host sites within the snow crystal lattice structure, whereas heavy noble gases rely on the presence of comparatively rare fluid inclusions.

1. Introduction

Dissolved noble gases in groundwater (He, Ne, Ar, Kr, and Xe) have been widely used to improve our knowledge of surface and groundwater dynamics by providing information on flow paths, connectivity between aquifers and water residence times (e.g., Andrews, 1985; Bottomley et al., 1984; Torgersen and Ivey, 1985; Mazor and Bosch, 1992; Castro et al., 1998a, 1998b; Patriarche et al., 2004; Castro et al., 2007; Wen et al., 2015). Noble gases are conservative tracers within groundwater systems and their concentrations in the recharge areas of aquifers are generally considered to be mostly a function of temperature (T), partial pressure (P, from the altitude of recharge area) and excess air (EA). Consequently, noble gas temperatures (NGTs) have commonly been regarded as a potentially robust proxy for past climate (Stute and Schlosser, 1993; Ballentine and Hall, 1999; Aeschbach-Hertig et al., 2002; Kipfer et al., 2002; Castro and Goblet, 2003; Kulongoski et al., 2003; Sun et al., 2010; Castro et al., 2012).

NGTs derived from groundwater in sedimentary systems are generally assumed to reflect subsurface conditions at the water table and thus they are usually interpreted as representing the mean annual air temperature (MAAT) and pressure conditions at the base of the aerated zone (Stute and Sonntag, 1992; Klump et al., 2007). However, the same does not hold true in fractured media (Manning and Solomon, 2003; Warrier et al., 2012). Indeed, due to the presence of preferential flow paths in fractured systems leading to rapid infiltration, it is likely that recharge water in these systems might reflect the temperature of the ground surface at the precise time of infiltration rather than the MAAT (Warrier et al., 2012, 2013). Alternatively, NGTs in fractured recharge areas might record conditions at high altitude if rainwater did not have enough time to equilibrate with surface conditions (Warrier et al., 2013). If this is the case, noble gases in fractured and karstic areas with thin soil cover will be recording seasonality and/or the nature of the original precipitation, and thus, should also provide us with

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information about the timing of recharge as well as information on recharge sources. Depending on the region considered, recharge sources will include fog, orographic rain, synoptic rain, snow and ice. The noble gas composition of these sources, however and that of natural fog and snow in particular, have not been investigated and remain largely unknown. Yet, in addition to glacial systems (e.g., Niu et al., 2015, 2017a, b), these end-members are critical to a proper understanding of the functioning of many groundwater flow systems.

Top et al. (1988) found that sea ice was generally enriched in He and highly depleted in Ar, Kr and Xe relative to ASW. More recently, Malone et al. (2010) confirmed this broad pattern of noble gas concentrations for ice samples from Lake Vida, Antarctica. Overall, ice samples were found to be enriched in He and slightly depleted in Ne with saturation average values relative to ASW of 1.38 and 0.82, respectively (Malone et al., 2010). In contrast, all heavier noble gases in ice are strongly depleted with respect to ASW, with relative saturations of 0.10, 0.06 and 0.05 for Ar, Kr and Xe, respectively. Although some studies of noble gas concentrations in glacial ice, glacial firn and ground ice have been performed (e.g. Severinghaus et al., 2003; Severinghaus and Battle, 2006; Utting et al., 2016), there has not yet been a systematic study of noble gases trapped within freshly produced snow. In many regions, freshly melted snow is a critical component of groundwater systems and snow at altitude is frequently a primary source material for rain. Acquiring knowledge of this end-member in terms of noble gas signatures and assessing its impact on the overall noble gas signature in these groundwater systems is thus critical not only to water resource management (e.g., Warriner et al., 2012) but also to paleoclimate reconstructions (e.g., Ma et al., 2004; Hall et al., 2005; Castro et al., 2012). In addition, this will also contribute to improving our understanding of the current extent of meltwater in ice sheets and alpine glaciers around the world (e.g., Niu et al., 2015, 2017a, 2017b).

The pattern of significant depletion of heavy noble gases seen in ice by Top et al. (1988) and Malone et al. (2010) points to ice formation as being a possible cause for the observed Ar, Kr and Xe concentration depletion previously observed in rainwater (Warriner et al., 2013; Niu et al., 2015). However, at this stage, the noble gas signature of snow, which forms under conditions that are distinct from those of bulk ice, remains unknown. Here, a methodology to measure both light and heavy noble gases in collected snow samples is outlined. The extremely fine-grain nature of this material poses significant experimental challenges. Our results confirm the basic pattern of elevated He concentrations along with depleted concentrations of heavy noble gases relative to ASW. In addition, important inferences can be made as to the nature of how noble gases are trapped within snow.

2. Experimental and analytical methods

2.1. Sampling collection and procedure

Seven snow samples were collected for noble gas analysis between

Table 1
Summary of snow samples.

Label	Sample type	Comment and details	Latitude (North)	Longitude (West)
Ann Arbor				
SA030315	Snow	t = 9:45 am, T = -5 °C	42° 16' 38.169"	83° 44' 6.833"
SA012513	Snow	t = 11:10 am; T = -9 °C	42° 16' 38.169"	83° 44' 6.833"
SA022213	Snow	t = 10:40 am. T = -6 °C	42° 16' 38.169"	83° 44' 6.833"
IA030315	Ice pellet	t = 1:50 pm; T = -1 °C	42° 16' 50.596"	83° 46' 4.169"
SA122415	Snow		42° 16' 50.596"	83° 46' 4.169"
Bloomfield Hills				
SB031714	Snow	T = -6 °C	42° 24' 30.92"	83° 22' 40.126"
SB020115	Snow		42° 24' 30.92"	83° 22' 40.126"
Milan				
SM022416	Snow	t = 10:30 am; T = 0 °C	42° 5' 3.958"	83° 41' 36.538"

2013 and 2016 at three different locations in the greater Detroit area, Michigan, for measurement of He, Ne, Ar, Kr and Xe concentrations and their isotopic ratios (Fig. S1; cf. Supporting information, Table 1). These include four snow samples collected in Ann Arbor at two different locations, two in Bloomfield Hills, and one in Milan. In addition, one ice pellet sample was collected in Ann Arbor (Table 1). Sample names indicate type (snow - S or ice - I), location (A - Ann Arbor; B - Bloomfield Hills; M - Milan) and collection date (mmddyy). Samples were collected during snow or ice fall, stored in sealed plastic bags and kept at -25 °C until analyzed. However, samples collected in 2013 show significant He and Ne depletion compared to those collected in 2014, 2015 and 2016.

2.2. Noble gas analysis

Noble gas measurements were performed in the Noble Gas Laboratory at the University of Michigan. The complete measurement procedure comprises estimation of He, Ne, Ar, Kr and Xe concentrations and isotopic ratios with standard errors for volume estimates of 1.5%, 1.3%, 1.3%, 1.5% and 2.2% for He, Ne, Ar, Kr and Xe, respectively. All snow and ice samples were melted under vacuum ($< 1 \times 10^{-4}$ Torr) in a sealed container attached to an automated noble gas extraction and purification line (Castro et al., 2009; Saar et al., 2005). Melted samples were stirred for 20 min while water vapor was used as a carrier gas through a 1/16" diameter stainless steel tube into a cold trap. Subsequently, that trap was warmed and the released water vapor was used as a carrier gas to pump the noble gases into a zone with dried 3 Å molecular sieve. Noble gases were then purified by two Ti sponge getters, and sequentially allowed entering a Helix SFT™ Noble Gas Mass Spectrometer for He and Ne measurements and an Argus VI™ Noble Gas Mass Spectrometer for Ar, Kr and Xe measurements using a dual chamber cryo-separator. Release temperatures for He, Ne, Ar, Kr, and Xe from the cryo-separator were 40 K, 84 K, 210 K, 245 K, and 290 K, respectively. Further details can be found elsewhere (Wen et al., 2015).

Before each sample analysis, a calibrated amount of air standard and a procedural blank were performed exactly in the same manner as the sample measurement. The blank correction was applied to all measured peaks.

3. Rationale for this study and approach undertaken

Noble gas signatures in ice have been the object of previous studies. These signatures include studies to determine diffusivity and solubility coefficients (Haas et al., 1971; Satoh et al., 1994, 1996), to derive paleoclimate information (Severinghaus et al., 2003), to understand ice formation mechanisms (Namiot and Bukhgalter, 1965; Top et al., 1988; Hood et al., 1998; Malone et al., 2010; Hall et al., 2017) and, more recently, to trace the origin of ice (Utting et al., 2016). Nevertheless, up to now, no study investigated noble gases in snow itself. In order to characterize noble gases in snow, a new experimental design has been developed.

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