



## Using noble gas ratios to determine the origin of ground ice



Nicholas Utting<sup>a,b,\*</sup>, Bernard Lauriol<sup>c</sup>, Denis Lacelle<sup>c</sup>, Ian Clark<sup>a</sup>

<sup>a</sup> Department of Earth Science, University of Ottawa, 25 Templeton Street, Ottawa, ON K2N 1N5, Canada

<sup>b</sup> Faculty of Science, Concordia University of Edmonton, 7128 Ada Boulevard, Edmonton AB T5B 4E4, Canada

<sup>c</sup> Department of Geography, University of Ottawa, 60 University, Ottawa, ON K1N 6N5, Canada

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### ABSTRACT

Argon, krypton and xenon have different solubilities in water, meaning their ratios in water are different from those in atmospheric air. This characteristic is used in a novel method to distinguish between ice bodies which originate from the compaction of snow (i.e. buried snow banks, glacial ice) vs. ice which forms from the freezing of groundwater (i.e. pingo ice). Ice which forms from the compaction of snow has gas ratios similar to atmospheric air, while ice which forms from the freezing of liquid water is expected to have gas ratios similar to air-equilibrated water. This analysis has been conducted using a spike dilution noble gas line with gas extraction conducted on-line. Samples were mixed with an aliquot of rare noble gases while being melted, then extracted gases are purified and cryogenically separated. Samples have been analysed from glacial ice, buried snow bank ice, intrusive ice, wedge ice, cave ice and two unknown ice bodies. Ice bodies which have formed from different processes have different gas ratios relative to their formation processes.

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### Introduction

In formerly glaciated permafrost regions, bodies of massive ground ice and icy sediments are commonly found in the headwall of thermokarst slumps and along eroding river banks and coastal cliffs (Mackay, 1966; Lewkowicz, 1987; Burn and Lewkowicz, 1990). The origin of these massive ground ice bodies has been attributed either to a glacial origin (i.e., firnified or basal ice), buried surface ice (i.e., lake ice, river ice) or intrasedimental ice of a segregation / intrusive origin (Mackay, 1971; Rampton, 1982; Kaplyanskaya and Tarnogradskiy, 1986; Astakhov and Isayeva, 1988; Harry et al., 1988; St-Onge and McMartin, 1999; Murton et al., 2005; Froese et al., 2008; Waller et al., 2009). Determining the ice emplacement mechanisms (e.g. glacial vs. periglacial processes) is fundamental to unlocking the paleoenvironment and paleohydrology of a particular location. For example the discovery of buried glacier ice may help constrain the past extent of ice sheets (e.g. Lacelle et al., 2007) or reveal the preservation of permafrost during interglacial periods (Froese et al., 2008).

With the exception of wedge ice, typically with a V-shape profile (French, 1996), identifying the origin of massive ground ice exposed in permanently frozen sedimentary units using morphology alone is difficult. Other characteristics have been used to interpret ice origin including stratigraphic context, sediment content, bubble content and ice crystallography (Pollard and French, 1985). Although these features are important for interpreting ice origin, they can sometimes be ambiguous.

For example, both basal glacier ice and segregated ice may contain significant amounts of interbedded stratified sediment (Klassen and Shilts, 1987; Waller et al., 2009); buried snowbank and segregated ice may display anhedral equigranular crystals (Pollard and French, 1985); the orientation of air bubbles in buried snow bank ice and ice formed by freezing of liquid water may both contain vertically oriented bubble trains (Schumskii, 1964; Killawee et al., 1998). In response to the ambiguities in using stratigraphic, sedimentary and crystallographic methods, geochemical-based approaches have been used (eg. Craig, 1961; Cardyn et al., 2007). The stable O-D isotopes of water have diagnostic fractionation processes during freezing of liquid water. If ice forms from an accumulation of snow and is transformed to ice, the isotopic values ( $\delta^{18}\text{O}$  and  $\delta\text{D}$ ) should fall along the local meteoric water line (Craig, 1961). Comparatively, ice which forms from the freezing of liquid water will display a lower regression slope value relative to the local meteoric water line (Jouzel and Souchez, 1982; Mackay and Dallimore, 1992). The limitation of this technique is that depending on the fractionation factor between water and ice, the regression slope of the ice can be similar to that of local meteoric water lines that have values less than 7 (Lacelle and Clark, 2011). As such, these analyses should be supplemented by other methods when attempting to identify ice origin.

Recent studies have measured ratios of  $\text{O}_2$ ,  $\text{N}_2$  and Ar gases occluded within bodies of massive ground ice to infer their origin (Cardyn et al., 2007; Lacelle et al., 2007; St-Jean et al., 2011). Cardyn et al. (2007) first used molar gas ratios ( $\text{N}_2/\text{Ar}$  and  $\text{O}_2/\text{Ar}$ ) to differentiate ground ice of glacial origin from ice formed by freezing of liquid water. This is based on the premise that glacial ice should display molar gas ratios similar to those of atmospheric air incorporated during occlusion and transformation of the firn to ice, whereas the ratios in subsurface ice

\* Corresponding author at: Faculty of Science, Concordia University of Edmonton, 7128 Ada Boulevard, Edmonton Alberta T5B 4E4, Canada.

E-mail address: [Nicholas.utting@gmail.com](mailto:Nicholas.utting@gmail.com) (N. Utting).

bodies originating from the freezing of water will deviate from atmospheric values due to the solubility of gases in water. However the freezing process does not leave gas concentrations unchanged in ice. The fractionation of gases during freezing has been studied in the context of the formation of sea ice. A laboratory study demonstrated that during the formation of frazil and columnar ice fractionation occurred (Top et al., 1988). Ice was enriched in helium (26%), and depleted in neon (−10%), argon (−45%), krypton (−60%) and xenon (−50%). Fractionations similar to laboratory values have been observed in natural ice bodies (Malone et al., 2010).

While the analysis of occluded O<sub>2</sub>, N<sub>2</sub> and Ar gases in subsurface ice can be used in determining the origin of ground ice, it can be complicated by the biogeochemical reactivity of certain gases (i.e. N<sub>2</sub>, O<sub>2</sub>, CO<sub>2</sub>, CH<sub>4</sub>; e.g. Lacelle et al., 2011). For example, Lacelle et al. (2011) presented evidence that O<sub>2</sub> and CO<sub>2</sub> gases in bodies of massive ground ice were modified by heterotrophic bacteria living within the ice matrix, thus making these gases useful biomarkers. The modification of occluded gases by biological activity has also been observed in polar ice. The basal ice layers of GRIP and GISP2 ice cores in Greenland showed high CO<sub>2</sub> and CH<sub>4</sub> concentrations that were related to high microbial counts in basal layers of ice; as such the elevated CO<sub>2</sub> and CH<sub>4</sub> gas concentrations are due to in situ microbial activities of methanogens (Price and Sowers, 2004; Tung et al., 2005). In a study of N<sub>2</sub>O concentration in portions of the Vostok ice core (Antarctica), Sowers (2001) found a 30% excess at a depth corresponding to the penultimate glacial maximum (135 ka BP). The N<sub>2</sub>O excess layer correlated with high bacteria counts and dust concentration, which Sowers (2001) suggested to be the result of in situ production by nitrifying bacteria. As a result, the concentrations of O<sub>2</sub>, N<sub>2</sub>, CO<sub>2</sub> and CH<sub>4</sub> can potentially be affected by biological reactions, compromising the analysis of these gases as an approach to distinguish firnified ice (glaciers, snowbanks) from ice derived from the direct freezing of water.

In an effort to derive an approach that would eliminate modification of gas concentration by biogeochemical processes, this study examines the use of noble gases to identify the origin of tabular massive ground ice bodies. Noble gases are inert and affected only by physical processes. Some previous studies have analysed noble gas concentrations in glacier ice to identify glacier melt layers from firn layers (Severinghaus et al., 2003; Severinghaus and Battle, 2006; Ahn et al., 2008; Orsi et al., 2015). In this study, the difference in solubilities of Ar, Kr and Xe is exploited to determine the origin of massive ground ice bodies in permafrost from various sites in the Canadian Arctic. To verify this method, ground ice of 'known origin' interpreted by other methods was also analysed. The method was found to work most effectively in assessing ice genesis and mechanisms of emplacement when complemented by other tools for characterizing ground ice.

### Principles in using noble gases to determine ground ice types

Noble gases (He, Ne, Ar, Kr, Xe) are inert, making up less than 1% of the atmosphere composition. Since He and Ne may be mobile in ice

(Huber et al., 2006), this study focuses on Ar, Kr and Xe as a means of differentiating ground ice bodies. Argon makes up 0.93% of the atmosphere, whereas Kr is 0.000114% and Xe is 0.0000086% (Andrews, 1992). Although these concentrations have changed over geologic history, they have remained relatively constant over the last 1 Ma (Severinghaus and Battle, 2006).

The concentration of Ar, Kr and Xe gases dissolved in water is dependent on their concentration in the atmosphere, water salinity and temperature. The concentration of a given gas in surface water equilibrated with atmospheric air can be determined using Eq. (1).

$$C_{(cc_{stp}/cc)} = B_{(cc_{stp}/cc/atm)} \cdot P_{(atm)} \quad (1)$$

In Eq. (1), C is the concentration of gas in water, B is the Bunsen coefficient (at a given temperature) and P is the partial pressure of the gas of interest in the atmosphere. Table 1 shows the concentrations of Ar, Kr and Xe in atmospheric air, the Bunsen coefficients at different water temperatures and the concentration in water at these temperatures and the associated gas ratios in  $\delta$  notation. In  $\delta$  notation, gas ratios are compared to the ratio in atmospheric air. Gas concentrations are compared with xenon as it is expected to be the least mobile in ice bodies, therefore the most conservative tracer. The formula for calculating  $\delta$  (Ar/Xe) is shown in Eq. (2) (Kr may replace Ar).

$$\delta \left( \frac{Ar}{Xe} \right)_{Sample} = \left( \frac{Sample \left( \frac{Ar}{Xe} \right)}{Atmosphere \left( \frac{Ar}{Xe} \right)} - 1 \right) \cdot 100 \quad (2)$$

Using this notation, the value of  $\delta$ (Ar/Xe) and  $\delta$ (Kr/Xe) for air is 0.0% and for air-equilibrated water at 0°C is −75.5% for  $\delta$ (Ar/Xe) and −50.7% for  $\delta$ (Kr/Xe). There is a 3% difference between gas ratios in air equilibrated water at 10°C and 0°C (Table 1). This difference in  $\delta$ (Ar/Xe) and  $\delta$ (Kr/Xe) between atmosphere and air equilibrated water can be exploited in determining the origin of ground ice bodies. For example, if a ground ice body originates from the firnification of snow, as with glacial ice and buried snow pack, the gas ratios in that ice will be similar to the atmospheric values. Conversely, ground ice that forms from the freezing of water is expected to have noble gas ratios similar to those dissolved in water at recharge. O<sub>2</sub>/Ar and N<sub>2</sub>/Ar ratios are presented using the same notation where atmospheric air has a value of 0% for both  $\delta$ (O<sub>2</sub>/Ar) and  $\delta$ (N<sub>2</sub>/Ar) and air equilibrated water has a  $\delta$ (O<sub>2</sub>/Ar) of −7.1% and  $\delta$ (N<sub>2</sub>/Ar) of −57.5%.

Noble gases are inert and cannot be altered by biological or chemical reactions as observed for O<sub>2</sub>, N<sub>2</sub> and CO<sub>2</sub> (e.g. Sowers, 2001; Tung et al., 2005; Lacelle et al., 2011). However, the concentrations of noble gases can be affected by physical process during the transformation of firn ice into occluded glacier ice (Paterson, 1994). In firn, gas fractionation can occur below the convection zone in the diffusive zone and the non-diffusive zone of occlusion (Schwander, 1989). In firn, physical fractionation may occur with the heavier gases settling to the bottom

**Table 1**  
Concentrations of gases in atmosphere and water at 0°C, 1°C, 2°C, 5°C and 10°C and associated Bunsen coefficients and  $\delta$  values. (Bunsen coefficient as per: Weiss, 1970; Weiss, 1978; Clever, 1979, Concentrations in air equilibrated water calculated using Eq. (1)).

	Bunsen coefficient (cc/cc/atm)			Concentration (cc <sub>stp</sub> /cc)			$\delta$ (Ar/Xe) %	$\delta$ (Kr/Xe) %
	Ar	Kr	Xe	Ar (x 10 <sup>−4</sup> )	Kr (x 10 <sup>−8</sup> )	Xe (x 10 <sup>−8</sup> )		
Atmosphere				93.4	114.0	8.6	0.0	0.0
Concentration in water								
0°C	0.054	0.11	0.22	5.0	12.5	1.91	−75.9	−50.7
1°C	0.052	0.11	0.21	4.9	12.1	1.84	−75.6	−50.3
2°C	0.051	0.10	0.21	4.8	11.7	1.77	−75.3	−50.0
5°C	0.047	0.09	0.18	4.4	10.7	1.58	−74.3	−48.9
10°C	0.042	0.08	0.15	3.9	9.2	1.32	−72.7	−47.2

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