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Using oxygen isotopes to establish freshwater sources in Bedford Basin, Nova Scotia, a Northwestern Atlantic fjord

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ABSTRACT

A weekly time-series of oxygen isotope (δ^{18}O) measurements was collected over a 16-month period from near-surface (1 m) and near-bottom (60 m) waters of Bedford Basin, a coastal fjord adjacent to the Scotian Shelf, off eastern Canada. The time-series was complemented with δ^{18}O measurements of local precipitation (rain and snow), river, and wastewater runoff. The isotopic composition of precipitation displayed strong seasonality with an average (volume-weighted) δ^{18}O value of −5.39‰ (±0.96) for summer and a depleted value of −10.37‰ (±2.96) over winter. Winter precipitation exhibited more depleted and variable δ^{18}O of solid precipitation relative to rainfall. The annual, amount-weighted average δ^{18}O of Sackville River discharge (−6.49‰ ± 0.82) was not statistically different from precipitation (−7.24‰ ± 0.92), but exhibited less seasonal variation. Freshwater end-members (zero-salinity intercepts) estimated from annual and seasonal regressions of δ^{18}O versus salinity (S) for Bedford Basin near-surface samples were consistent with the δ^{18}O of summer precipitation and the annual, amount-weighted average for the Sackville River. However, the isotopically depleted signature of winter precipitation was not observed clearly in near-surface waters of Bedford Basin, which might reflect isotope enrichment during sublimation from accumulated snowfall prior to melting and discharge, or retention and mixing within the drainage basin. In near bottom waters, most of the δ^{18}O-S variation (average freshwater end-member: 7.47‰ ± 2.17) could be explained by vertical mixing with near-surface waters (average freshwater end-member: −6.23‰ ± 0.34) and hence with locally-derived freshwater. However the near-bottom δ^{18}O-S variation suggested an additional contribution of a freshwater end-member with a δ^{18}O of −15.55‰ ± 2.3, consistent with a remotely-derived freshwater end-member identified previously for the Scotian Shelf. Residuals from a long-term regression of δ^{18}O-S were generally within the range expected due to analytical uncertainty (±0.05); however near-surface waters exhibited seasonal variability of small amplitude, which was consistent with the timing and δ^{18}O variability of local freshwater inputs.

1. Introduction

The freshwater balance of the northern North Atlantic Ocean is changing as a result of changing Arctic sea-ice cover and increased melting of glacial ice (e.g., on Greenland; Bamber et al., 2012). Much of this Arctic-derived freshwater is transported southwards along the east coast of North America, as far as Cape Hatteras, by a 5000 km long boundary current (Chapman and Beardsley, 1989). This long-range influence of Arctic freshwater potentially impacts alongshore nutrient transport and stratification-dependent biological production over continental shelves. The alongshore continuity of this boundary current was first identified on the basis of an analysis of oxygen isotopes of water (Fairbanks, 1982).

The oxygen isotope composition of H_2O molecules has been used widely as a tracer of the hydrological cycle (Craig, 1961), ocean freshwater sources (e.g., Craig and Gordon, 1965; Fairbanks, 1982; Khatiwala et al., 1999), and sources of deep water masses (Bauch et al., 1995). Craig and Gordon (1965) first used δ^{18}O as an oceanographic tracer and established that, when paired with salinity (S), it can be used to characterize mixing of water masses and to distinguish different sources of freshwater (e.g., precipitation, river discharge, meltwater, etc.).

For the North Atlantic as a whole, freshwater originating from

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Arctic outflow dominates, with a δ^{18}O end-member of −21‰ (Khatiwala et al., 1999). Oxygen isotope analysis of the waters over the Scotian Shelf and Scotian Slope, off the east coast of Canada, revealed that the main freshwater input has its origin at high-latitudes (Fairbanks, 1982; Khatiwala et al., 1999). This Arctic freshwater source is complemented in spring and summer by the outflow of the Gulf of St. Lawrence (Shadwick and Thomas, 2011). Khatiwala et al. (1999) noted that the ^{18}O-salinity signature and resulting freshwater end-member of waters over the Scotian Shelf (−15.55‰; Fairbanks, 1982) are influenced strongly by sea-ice formation and brine rejection over the Labrador Shelf and by the heavier freshwater end-member derived from the St. Lawrence River (−10.3‰). The two main sources of freshwater on the Scotian Shelf, high-latitude (i.e. Arctic) runoff and St. Lawrence River water (SLRW), contribute freshwater in an approximately 2:1 ratio (Khatiwala et al., 1999).

A number of previous studies have used the δ^{18}O-S relationship within estuaries and fjords (Martin and Letolle, 1979; Austin and Inall, 2002), typically focussing on distinguishing various sources and sinks (precipitation, evaporation, river discharge, groundwater input, ice-melt) (e.g., Azetsu-Scott and Tan, 1997; Corlis et al., 2003; MacLachlan et al., 2007; Stalker et al., 2009; Turk et al., 2016; Whitney et al., 2017). The δ^{18}O-S relationship allows characterization of freshwater inputs with distinct δ^{18}O signatures in an estuary. However, few studies have examined the seasonality of the δ^{18}O-S relationship.

Here we present results from a 16-month, weekly time-series of δ^{18}O collected from Bedford Basin, a coastal fjord adjacent to the Scotian Shelf, together with measurements of local river water, wastewater, and precipitation. The data provide insight into the distribution and fate of freshwater sources as well as a reference point against which to evaluate longer-term changes in freshwater inputs, both local and remote, that might arise due to climate change.

2. Study site

2.1. Scotian Shelf

The Scotian Shelf is a 700 km long region of the continental shelf off Nova Scotia, varying in width from 120 to 240 km, and covering 120,000 km^2 with an average depth of 90 m. This region is bound by the Laurentian Channel to the northeast and by the Northeast Channel and the Gulf of Maine to the southwest (Shadwick and Thomas, 2011) (Fig. 1). Three major water masses contribute to Scotian Shelf waters: (1) Warm Slope Water (WSW), a mixture of Labrador Slope Water (LSW) and Gulf Stream (GS) water, (2) Labrador Shelf Water (LShW), derived from the inner branch of the Labrador Current, and (3) St. Lawrence Estuary Water (SLEW), which is influenced by the St. Lawrence River (Khatiwala et al., 1999; Shadwick and Thomas, 2011).

2.2. Bedford Basin

Bedford Basin is a fjord (volume: 5.6 × 10^8 m^3, area: 17 km^2, max. depth: 71 m) situated on the Atlantic coast of Nova Scotia, Canada, and separated from the adjacent Scotian Shelf by a 10 km long channel that is less than 400 m wide at its narrowest point, with a sill depth of ~20 m (Petrie and Yeats, 1990; Gregory et al., 1993; Li and Harrison, 2008) (Fig. 1; 2a).

2.2.1. Freshwater inputs & circulation

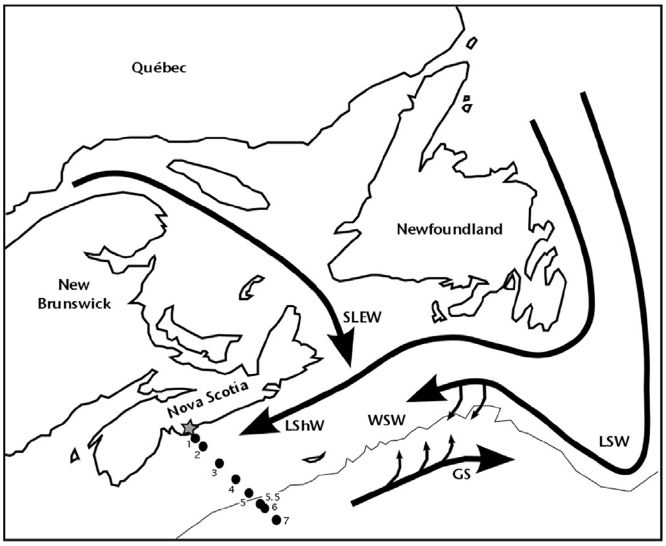

Fig. 1. General movement of water masses along the Scotian Shelf (Khatiwala et al., 1999; Shadwick and Thomas, 2011). SLEW: St. Lawrence Estuary Water, LShW: Labrador Shelf Water, WSW: Warm Slope Water, LSW: Labrador Slope Water, GS: Gulf Stream. Halifax Line stations are indicated along with station numbers (1–7) and the grey star identifies Bedford Basin. The bedrock geology surrounding Bedford Basin includes granite but is composed largely of very fine-to medium-grained metasandstone and slate, with relatively low groundwater permeability and yield (Kennedy and Drage, 2009).

The local input of fresh water to Bedford Basin is small relative to exchanges with the adjacent Scotian Shelf, with a tidal inflow to freshwater input ratio of 109.38 for Bedford Basin (Gregory et al., 1993). Near-surface salinity within Bedford Basin is typically in the range of 29–30.5 and hence lower than surface salinities of 30–31.5 measured at HL-2 (see Fig. 1). The annual average discharge of fresh water into Halifax Harbour is 15.7 m^3/s (Buckley and Winters, 1992) and is supplied from a watershed with area of 281 km^2 (Li and Harrison, 2008). The largest single source is the Sackville River, which enters Bedford Basin at its northwestward end and has an average discharge of 5.41 m^3/s (Buckley and Winters, 1992), ranging from 2 m^3/s to 9 m^3/s in the summer (July–September) and spring (March and April) respectively (Fournier, 1990). The remaining freshwater discharge is via a number of small streams and sewers (Buckley and Winters, 1992) with c. 16% of the total delivered by the Halifax wastewater treatment system, whose water originates from two lakes, Pockwock Lake and Lake Major, as well as a number of smaller reservoirs (Fader and Buckley, 1995). Direct precipitation onto Bedford Basin contributes an additional 0.8 m^3/s (Kerrigan, 2015). The contribution of submarine groundwater discharge (SGD) to overall freshwater input is not known. However given the low permeability bedrock (Fig. 1), a groundwater recharge ratio for the catchment area of 0.12 (Gavin Kennedy, Nova Scotia Department of Natural Resources, personal communication, July 2017), and the likelihood that much of the recharge is delivered to Bedford Basin via rivers and streams, we consider additional input by SGD to be small for this water body.

Bedford Basin is stratified by temperature seasonally, and by salinity almost year-round due to local freshwater inputs (Li and Harrison, 2008). In some years, near-bottom waters can become hypoxic ($[O_2] < 20$ µmol kg^{-1}) by late fall and early winter (Hargrave et al., 1976; Punshon and Moore, 2004). However, bottom waters are replenished periodically, either as a result of (1) wind-induced vertical mixing during periods of weaker stratification in winter (Platt et al., 1972; Punshon and Moore, 2004), or (2) lateral mixing with saltier waters from offshore, typically during spring and fall, due to strong alongshore winds and large tides or storm events (Burt et al., 2013). These lateral intrusions can result from a build-up of coastal sea level, and bring dense offshore continental shelf water with higher O_2 levels over the sill in the Narrows (20 m) and into deep (70 m) Bedford Basin (Burt et al., 2013). Temperature and salinity as well as O_2 data tend to change abruptly with these intrusion events (AMEC Earth and Environmental, 2011; Burt et al., 2013), supporting a shallow offshore intrusion source, consistent with lateral intrusion mechanisms described by Platt (1975).

The overall circulation within Bedford Basin is estuarine with lower-density upper water flowing out towards the Scotian Shelf, and deeper saline water flowing occasionally into the basin over the sill (Fig. 2b). The mean tidal range is 1.5 m and the flushing time of near-surface waters ranges from 10 (Gregory et al., 1993) to 40 days (Shan and Sheng, 2012), while the flushing time for the entire basin was determined to be ~90 days.

3. Methods

3.1. Sample collection

3.1.1. Scotian Shelf sample collection

Samples for $\delta^{18}O$ and salinity were collected offshore of Halifax along the “Halifax Line” (Therriault et al., 1998, Fig. 1) during cruises of the CCGS Hudson in October 2008 and April 2009 (Shadwick and Thomas, 2011). Seawater samples were collected at each site and sample depth for isotopic analyses ($\delta^{18}O$) from 20 L Niskin bottles, mounted on a General Oceanic 24-bottle rosette equipped with a CTD (SeaBird).

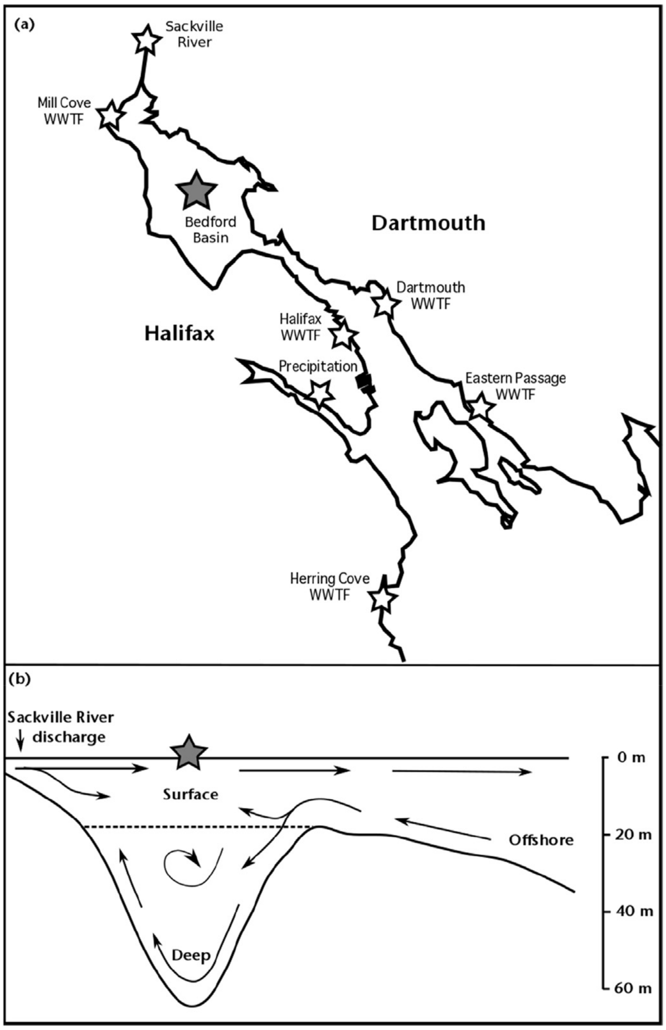

Fig. 2. (a) Sample sites for this study, and their position within Halifax Harbour. The grey star denotes the “Compass Buoy” station (see text) where most sampling took place. (b) A cross-section schematic of the two-layered flow in Halifax Harbour, depicting dense offshore water inflow, and freshened surface water outflow, adapted from Fader and Miller (2008).

3.1.2. Bedford Basin sample collection

Sampling for oxygen isotopes of H_2O was supported by the Bedford Basin Monitoring Program (BBMP), which has maintained weekly sampling in Bedford Basin (Compass Buoy Station: 44°41′37″ N, 63°38′25″ W; Fig. 2a) at four depths (1, 5, 10, and 60 m) since 1992 (Burt et al., 2013; Li, 2014). Oxygen isotope samples were collected at 1 and 60 m to represent surface and deep water respectively. Following collection, 4 L Niskin bottle samples were sub-sampled into 250 ml (for salinity) and 60 ml (for $\delta^{18}O$) Boston Round bottles with Poly-Seal-Lined caps wrapped with electrical tape to prevent evaporation during storage. $\delta^{18}O$ samples were stored at 4 °C until analysis (typically between 1 and 4 weeks), while salinity samples were stored at room temperature (c. 20 °C) and analyzed every 3–4 months.

3.1.3. Sackville River sample collection

Samples from the Sackville River were collected with a bucket lowered from a pedestrian bridge, 1.2 km from the river mouth, and the bucket contents were transferred directly into 60 ml ($\delta^{18}O$) Boston Round glass bottles. Samples were collected monthly, except for January and February of 2013 when the river was frozen.

3.1.4. Wastewater sample collection

Wastewater samples were collected on April 3, 2014 from the five major wastewater treatment facilities (WWTFs) in Halifax: Herring Cove, Halifax, Mill Cove, Dartmouth, and Eastern Passage. Collected samples represent the outflowing, treated wastewater that enters Halifax Harbour. Samples were collected in either Nalgene or glass bottles before being transferred to Dalhousie University, where they were stored in 60 ml Boston Round glass bottles before analysis took place.

3.1.5. Precipitation sample collection

Samples of precipitation were collected from June 2012 to October 2013 from the roof of the Oceanography Department at Dalhousie University (43°38′10″ N, 63°35′38″ W) away from any potential sources of contamination. Rainfall samples were collected using a 500 ml Pyrex bottle fitted with a glass funnel (135 mm diameter). The bottle was typically collected between 8:00 and 10:00 Monday to Friday following a precipitation event. On occasion, samples were collected later in the day following the completion of an event, in order to collect a single sample representative of the event. During major, longer, rainfalls samples were collected throughout to avoid the risk of evaporation or overflow. No samples were collected between 17:00 Friday and 8:00 Monday. Following sample collection, a 60 ml Boston Round glass bottle was filled, capped, taped, and stored at 4 °C. The volume of the remaining sample was measured and the contents transferred into a large glass bottle, containing all of the precipitation samples collected during that month to provide a “monthly-integrated” sample.

Samples of solid precipitation (snow, ice, and hail) were collected using a bucket, weighed down with rocks and lined with plastic. Since snow collection is influenced strongly by wind, some samples could not be volume-weighted accurately. This occurred on three occasions in February 2013, when large snowdrifts were present and snow sample amounts were not representative of the total volume that fell.

Snow samples were collected prior to melting and transferred into 1 L Nalgene bottles. These bottles were stored at 4 °C to allow the snow to melt gradually, and covered to prevent isotopic fractionation during evaporation (Cooper, 1998). Once melted, the water volume was recorded and a 60 ml Boston Round bottle was filled for isotopic analysis. Any remaining meltwater was added to the total monthly precipitation, or “monthly-integrated”, bottle (with the exception of samples that could not be volume-weighted).

3.2. *Volume-weighting of precipitation samples*

Volume-weighted averages were calculated for monthly and yearly precipitation, as both the number of precipitation events and the volume of precipitation that fell in a single event were variable (from 50 to 1200 ml). Volume-weighted averages ($\overline{x}$) of δ^{18}O for years, months and seasons, were calculated as:

$$\overline{x} = \frac{\sum_{i=1}^{N}(V_i \cdot x_i)}{\sum_{i=1}^{N}(V_i)}$$

where the total volume (ml) of precipitation measured for each event was defined as “V” and δ^{18}O measurements were defined as “x”. These calculated volume-weighted monthly averages based on measurements from individual precipitation events were compared with the values measured from the monthly-integrated water samples. Based on linear regression, the volume-weighted averages did not differ significantly from the values measured from the monthly-integrated samples for the 16 months of this study (r^2 value of 0.998). The calculated volume-weighted averages are used in the remainder of this paper. Additional sampling tests, and estimates of small uncertainties or possible bias arising from sampling procedures are discussed in Kerrigan (2015).

3.3. *δ^{18}O and salinity measurements*

Seawater samples collected in 2008 and 2009 over the Scotian Shelf, on the Halifax Line (Fig. 1), were analyzed separately from all other samples. These δ^{18}O samples were processed at the University of Ottawa using an isotope ratio mass spectrometer (IRMS) and salinity data were taken from the CTD. Based on an inter-comparison study by Walker et al. (2015), these earlier isotope data, measured with an IRMS, can be compared directly with our analyses, which uses WS-CRDS (wavelength scanned cavity ring-down spectroscopy).

All other measurements of the oxygen isotope ratio of water (δ^{18}O) were completed using a Picarro L2130-i WS-CRDS isotopic water analyzer. This was equipped with an HTC-xt Leap Pal Technologies autosampler and an A0211 high precision water vaporizer. Full details of the system used in our laboratory are reported in Walker et al. (2015). In-depth descriptions of the standardization and testing of the instrument are presented in Kerrigan (2015). Oxygen isotope measurements are reported in delta (δ) notation in units of permil (‰) and compared to a known standard, VSMOW2 (Vienna Standard Mean Ocean Water 2; IAEA, 2017), so that:

$$\delta^{18}O = \frac{(^{18}O/^{16}O)_{sample} - (^{18}O/^{16}O)_{VSMOW2}}{(^{18}O/^{16}O)_{VSMOW2}} \cdot 1000$$

Walker et al. (2015) determined a within-lab analytical precision of 0.03‰, which is better than the manufacturer's stated precision of 0.05‰, while inter-laboratory differences define an accuracy of 0.1‰ (with the potential to approach 0.05‰). All uncertainties presented in this manuscript are recorded as 95% confidence intervals, and will be written in brackets along with the N value of the averages/zero-salinity intercept, e.g., −10.37‰ (±2.96, n = 34).

Salinity samples collected in Bedford Basin were analyzed at the Bedford Institute of Oceanography using a Guildline Autosal 8400-B salinometer. The error associated with this instrument is 0.002 psμ and IAPSO Standard Seawater was used as a standard reference material. Salinity is reported on the practical salinity scale.

4. Results & discussion

4.1. *Variability of δ^{18}O for precipitation, river, and wastewater inputs*

4.1.1. *Seasonal differences in the δ^{18}O of local precipitation*

The volume-weighted monthly averages of δ^{18}O in precipitation (Fig. 3a) revealed clear seasonal variability, with isotopically enriched precipitation falling during the summer months (defined here as June, July, and August), compared to winter (defined here as December, January, and February). The volume-weighted average for an entire year (June 2012–July 2013) was −7.24 (±0.92‰, n = 60).

The seasonal difference of the δ^{18}O of precipitation between winter and summer months was confirmed using a Wilcoxon-Mann-Whitney (WMW) rank sum test (p value = 0.00231). The data were used to define two, volume-weighted, end-members for precipitation falling within the catchment area of Bedford Basin: a winter end-member of −10.37‰ (±2.96, n = 11) and a summer end-member of −5.39‰ (±0.96, n = 13). This seasonal difference was influenced by wintertime occurrence of solid precipitation (i.e. snow, hail, and graupel), which carried a more depleted isotopic signature. When divided into two categories, rain and snow (the latter included all forms of solid precipitation), a WMW rank sum test confirmed a significant difference (p value < 0.05) between the isotopic composition of rain (−8.10‰ ± 2.28, n = 10) and snow (−14.81‰ ± 3.29, n = 13) when both types of precipitation were falling concurrently (i.e. within the same period from January–April 2013, p value = 0.004) (Fig. 3b). In addition to the strong and variable isotopic depletion associated with snowfall, there was also marked seasonality of rainfall isotopic composition. Fig. 3b also shows a significant difference (p value = 0.016) between the isotopic composition of winter (December, January, and February) and summer (June, July, and August 2013) rainfall, with average δ^{18}O values of −7.90‰ (±2.00, n = 8) and −5.01‰ (±0.99, n = 13) respectively.

The isotopic depletion of solid precipitation, compared with winter rainfall, is likely the result of equilibrium fractionation within clouds. Isotopic fractionation effects occur with all forms of precipitation, however the isotopic composition of solid precipitation can be “frozen in”, while raindrops can undergo continuous molecular exchange as they fall towards the ground, and as such can be altered through exchange with ambient moisture (Gat, 2010).

4.1.2. *Temporal variability in the δ^{18}O of river runoff and wastewater inputs*

Relative to precipitation, seasonal variability of smaller amplitude was observed in δ^{18}O measurements from the Sackville River (Fig. 3a). A WMW rank sum test implied that precipitation and Sackville River oxygen isotope values were not statistically different from each other over the year as a whole (p value of 0.3895), however as noted above, no data from the Sackville River could be obtained during January and February of 2013.

The average of all Sackville River δ^{18}O measurements was −6.55‰ ± 1.32, n = 16. In order to obtain a discharge-weighted mean for an annual cycle, monthly average values (with the exception of January and February) were weighted by the average

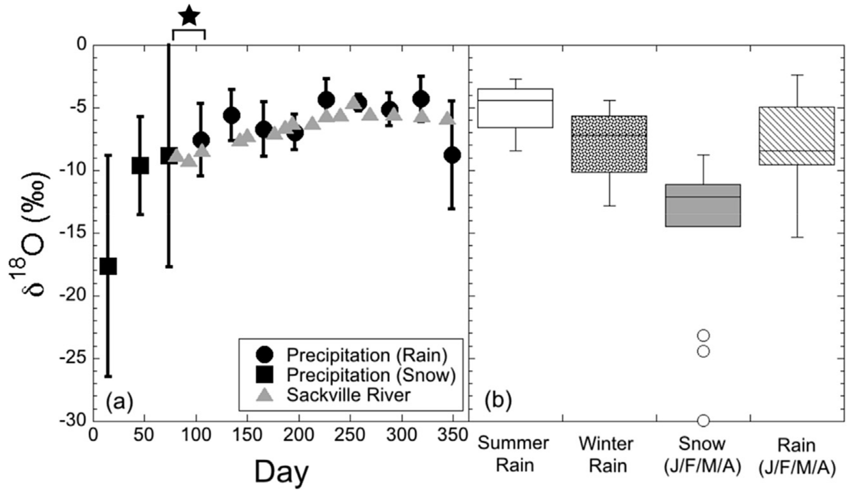

Fig. 3. (a) Volume-weighted monthly average $\delta^{18}O$ of precipitation (black) and individual Sackville River (grey) $\delta^{18}O$ measurements over a year. Months where both snow and rain were sampled are indicated with a box. This plot only includes results from precipitation samples that could be volume-weighted. The black star identifies the time period when the spring melt occurred in 2013 and 2014, based on discharge rates measured at the mouth of the Sackville River (Environment Canada, 2017). (b) Box-whisker plot, where the horizontal line represents the median, showing oxygen isotope composition of summer rainfall (June, July, and August 2013), winter rainfall (December 2012, January, and February 2013), snow (all snow that fell during this study; collected in January (J), February (F), March (M), and April (A)), and rain that fell concurrently with snow (i.e. rain collected over the four months during which snow fell).

monthly river discharge for the period summer 2012 to 2013 (Environment Canada, 2017). This gave a discharge-weighted mean of −6.49 ± 0.82‰, n = 10.

Wastewater samples were collected only once, on April 3, 2014, from five major wastewater treatment facilities (WWTFs) that drain into Halifax Harbour (Fig. 2a). The $\delta^{18}O$ of these inputs (Table 1) reveal some differences between themselves and with the Sackville River which may have been related to: (1) the occurrence of a winter storm on April 2, 2014, and (2) differences in post-precipitation residence times for fresh water supplying the Sackville River and the various WWTFs. We expect input from WWTFs to be more sensitive to short-term variations in precipitation than the Sackville River, which has a large drainage basin including several lakes.

4.2. Freshwater end-member variability within Bedford Basin

4.2.1. Seasonal differences of freshwater end-members for Bedford Basin surface waters

Near-surface waters within Bedford Basin represent a mixture of a large reservoir of seawater from the Scotian Shelf with locally-introduced freshwater, and have a residence time of 10–40 days (Gregory et al., 1993; Shan and Sheng, 2012). We therefore expected the zero-salinity intercept (freshwater end-member) of regressions of $\delta^{18}O$-S to reflect seasonal variation of the isotopic signature of the local freshwater inputs (Fig. 4). Values of zero-salinity intercepts for annual and seasonal groupings of near-surface samples are presented in Table 2 along with the winter and summer precipitation end-members, and annual average amount-weighted $\delta^{18}O$ for precipitation (volume-weighted) and the Sackville River (discharge-weighted).

Table 1
$\delta^{18}O$ of wastewater treatment facility (WWTF) outfalls and the Sackville River. All samples were collected on April 3, 2014.

Sample Site	$\delta^{18}O$ (‰)
Herring Cove WWTF	−8.44
Halifax WWTF	−7.83
Mill Cove WWTF	−8.07
Dartmouth WWTF	−8.37
Eastern Passage WWTF	−8.16
Sackville River	−9.15

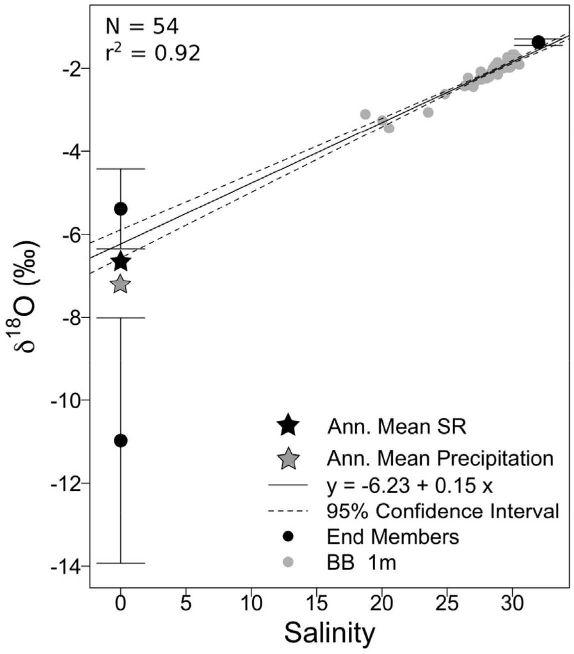

Fig. 4. $\delta^{18}O$-S relationship at 1 m in Bedford Basin for all samples collected from June 2012 to October 2013. Dark circles represent summer precipitation (0, −5.39‰), winter precipitation (0, −10.97‰), and offshore end-members (31.99, −1.37‰), along with their associated error, and stars represent the annual average amount-weighted $\delta^{18}O$ of the Sackville River (SR) (−6.49 ± 0.82‰) and precipitation (−7.24 ± 0.92‰).

Near-surface, zero-salinity intercepts calculated for both summer 2012 and 2013 were isotopically lighter than the summer precipitation end-member, but the differences were not statistically significant (Table 2). In contrast, there was no clear reflection in the wintertime, near-surface, zero-salinity intercepts of the lighter isotopic composition of winter precipitation. The zero-salinity intercept of near-surface samples collected over winter

Table 2
Zero-salinity intercepts determined using the δ^{18}O-S relationship of seasonal groupings of near-surface measurements from Bedford Basin. Amount-weighted summer and winter precipitation end-members, and average annual values for the Sackville River (discharge-weighted) and precipitation (volume-weighted) are also included. 95% CIs are included for all values presented.

	N	Zero-Salinity Intercept (‰)	r^2	p-value
Summer 2012	8	−7.09 ± 1.67	0.90	0.00030
Fall	12	−5.27 ± 0.53	0.95	$7.91e^{-8}$
Winter	8	−6.81 ± 2.76	0.76	0.0047
Spring	10	−6.84 ± 1.05	0.93	$5.47e^{-6}$
Summer 2013	11	−6.54 ± 0.34	0.96	$9.67e^{-8}$
Average (All Data) (July 2012–Oct 2013)	54	−6.23 ± 0.34	0.92	$<2.2e^{-16}$
Annual Average (July 2012–June 2013)	40	−6.54 ± 0.34	0.90	$<2.2e^{-16}$
	N	Amount-Weighted δ^{18}O End-Member (‰)		
Summer Precipitation	13	−5.39 ± 0.96		
Winter Precipitation	11	−10.97 ± 2.96		
	N	Amount-Weighted Annual Average δ^{18}O (‰)		
Sackville River (May 2012–June 2013)	10	−6.49 ± 0.82		
Precipitation (July 2012–June 2013)	60	−7.24 ± 0.92		

(−6.81‰; ±2.76, n = 8) was not significantly different from intercept values estimated from summer data. However the large uncertainty of the near-surface winter intercept value prevented it from being distinguished statistically from either winter or summer precipitation end-members (Table 2). The zero-salinity intercept (−6.23‰ ± 0.34, n = 54) calculated for near-surface (1 m) samples collected over the entire sampling period (June 2012 to October 2013) was consistent with δ^{18}O values measured in summertime precipitation (−5.39‰; ±0.96, n = 13), but not with the average of wintertime precipitation (−10.97‰; ±2.96, n = 11). On the other hand, the zero-salinity intercepts also aligned closely with the annual average Sackville River δ^{18}O (Table 2), which exhibited less seasonal variation than precipitation.

4.2.2. Isotope transport, fractionation, and exchange within the drainage basin

The zero-salinity intercepts and their relation to seasonal precipitation suggest either that: (a) the wintertime precipitation (which includes isotopically light snow) undergoes isotopic modification before being discharged to Bedford Basin (e.g., with the spring melt; Fig. 3a); or (b) the isotopically light wintertime precipitation is mixed with “heavier” water within the drainage basin prior to discharge.

Snowfall, in particular, can be retained within the drainage basin prior to melt and discharge. During this period, the associated light isotopes can be lost preferentially via sublimation and/or evaporation, leaving the remaining snow cover and eventual runoff from spring ice-melt enriched in heavier isotopes relative to initial precipitation. Prior studies have shown that the isotopic composition of a snowpack, which usually includes snow from multiple snowfalls, is more variable than that of snowmelt (Taylor et al., 2001), and can become significantly heavier than the original snowfalls due to water re-mobilization and evaporation/sublimation as snow cover accumulates. These effects can be large, with reported shifts of up to 10‰ for δ^{18}O and 50‰ for δ^{2}H (Cooper, 1998). Hence snowmelt that ultimately flows into Bedford Basin may be more isotopically enriched than the “winter precipitation” end-member estimated from our direct sampling of snowfall. In addition to delayed discharge and preferential loss of light isotopes, the signature of wintertime precipitation is likely to be mixed, within the drainage basin, with water originating from different seasons. The observed seasonal variability of Sackville River isotopic composition, which is also “damped” relative to precipitation, illustrates the combined effect of these processes operating within the drainage basin (Fig. 3a).

On the annual timescale, the most notable result is that the zero-salinity intercept determined from Bedford Basin surface water is fully consistent with the annual amount-weighted averages of δ^{18}O for precipitation and the Sackville River (Table 2). Each of these annual averages represents a different stage of the hydrologic cycle within the drainage basin. Although there is a hint of isotopic enrichment as precipitation is transferred via the Sackville River to Bedford Basin, the overall difference (c. 1‰) is not statistically significant (Table 2). Other studies (e.g., Gremillion and Wanielista, 2000) have observed progressive isotopic enrichment (c. 1‰) due to evaporation within drainage basins. It is likely that this drainage basin is too small, or the seasonal variability too large, for such an effect to be detected.

4.2.3. Freshwater end-members for Bedford Basin's deeper waters

The deeper waters of Bedford Basin (60 m) have higher salinities, reflecting their offshore origin via lateral intrusions, and have a much more restricted salinity range, reflecting limited mixing with locally supplied freshwater. The limited salinity range hinders estimation of zero-salinity intercepts based on regressions of δ^{18}O and salinity. The overall relationship between δ^{18}O and salinity at 60 m is, consequently, only weakly determined (Fig. 5a; r^2: 0.36, p-value: $1.52e^{-6}$), and the uncertainty of zero-salinity intercepts estimated from seasonal data at 60 m is 1–2 orders of magnitude larger than for the near-surface samples. These large uncertainties make it impossible to examine the seasonal variation in zero-salinity intercepts as was done for surface waters. However, by using all available data in a regression, an effective δ^{18}O freshwater end-member of −7.47‰ ± 2.17 was estimated (Fig. 5a). This value is indistinguishable from the average δ^{18}O freshwater end-member determined for near-surface waters (−6.23‰ ± 0.34, n = 54). In turn, this suggests that the primary driver of salinity variations in the deeper waters of Bedford Basin is vertical mixing with near-surface waters, which carry a δ^{18}O signature set by local freshwater inputs.

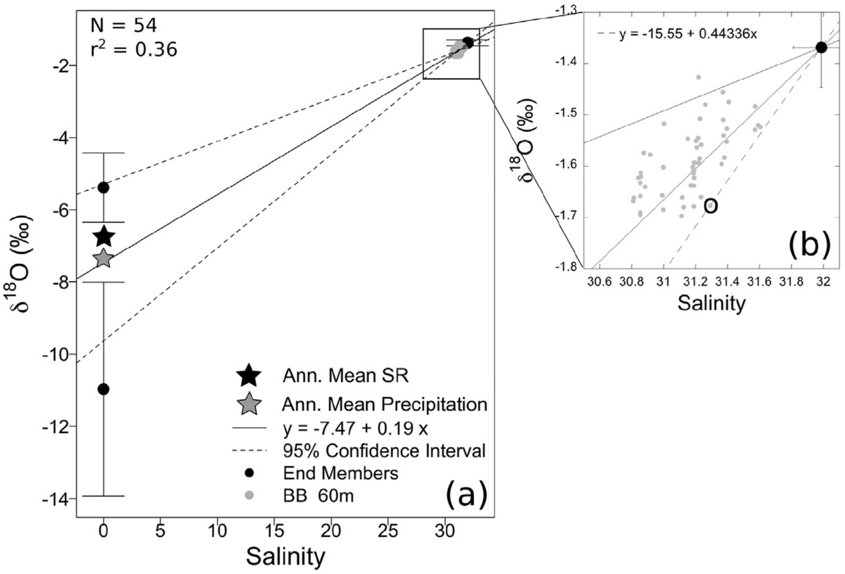

Fig. 5. (a) δ^{18}O-S relationship and mixing triangle at 60 m in Bedford Basin from June 2012 to October 2013. Dark circles represent summer precipitation (0, −5.39‰), winter precipitation (0, −10.97‰), and offshore end-members (31.99, −1.37‰), along with their associated error, and stars represent the annual average amount-weighted δ^{18}O of the Sackville River (SR) (−6.49‰ ± 0.82) and precipitation (−7.24‰ ± 0.92). (b) Expanded view, showing the mixing line (y = −15.55 + 0.443x) determined using the offshore end-member and the sample (circled) that falls the furthest below the mixing triangle.

While this mixing process explains the bulk of the δ^{18}O-S variability in deep water, not all of the observed variability can be explained by local inputs. A mixing triangle can be produced based on local freshwater inputs, using summer and winter precipitation end-members, along with a saline, offshore input. The summer and winter precipitation values are used to define extreme ranges for the potential contribution of locally-derived freshwater. The offshore end-member (S, δ^{18}O: 31.99 ± 0.18, −1.37‰ ± 0.08, n = 70) was established by averaging samples collected in the top 60 m of the first five Halifax Line stations over the Scotian Shelf (HL-1 to HL-5; Fig. 1). These samples were selected to represent the water on the Scotian Shelf that enters Halifax Harbour: including LShW and SLEW, which travel alongshore, close to the surface, but eliminating WSW, which is found only in the deeper waters on the shelf and on the Scotian Slope, and which does not enter Halifax Harbour. Chapman et al. (1986) defined Scotian Shelf water end-members (S, δ^{18}O) in the spring of 1981 (32.35, −1.09‰) and the fall of 1981 (31.92, −1.26‰). These values compare well with the Scotian Shelf end-member used here. Additional details and rationale for the choice of the offshore end-member are presented in Kerrigan (2015). The end-members are presented in Figs. 4 and 5a, for 1 m and 60 m Bedford Basin samples respectively.

Inspection of Fig. 5b shows that a number of samples measured at 60 m in Bedford Basin fell below the mixing triangle, suggesting that an additional, more isotopically depleted, freshwater end-member contributes, at least occasionally, to the deeper waters of Bedford Basin. It was noted earlier that Scotian Shelf waters, which enter Halifax Harbour and spill over the sill into Bedford Basin, have a δ^{18}O signature that reflects a freshwater component considerably more isotopically depleted than local freshwater (e.g., Arctic river water: −21‰; St. Lawrence River Water: −10.3‰; Khatiwala et al., 1999). We have used the sample that falls the furthest outside the mixing triangle (see Fig. 5b) to estimate the isotopic composition of an additional freshwater end-member, presumably supplied from offshore, which is required to explain the observed data distribution at 60 m depth.

The freshwater end-member value for this additional source was calculated with a mixing line connecting the offshore end-member value with the measurement within Bedford Basin near-bottom waters that fell the furthest below the mixing triangle (the black circle in Fig. 5b). The analytical uncertainty associated with δ^{18}O measurements (±0.05‰) is used to calculate the uncertainty of this additional freshwater end-member (±2.3‰). The value obtained (−15.55‰ ± 2.3) is identical to the Scotian Shelf freshwater end-member of −15.55‰ identified by Fairbanks (1982) and similar to the zero-salinity intercept calculated from a regression analysis of Halifax Line samples collected over the Scotian Shelf (HL-1 to HL-5), −16.09‰ (±0.77, n = 72).

4.3. Residual analysis

Temporal variability was investigated in more detail by examining patterns of residuals of individual δ^{18}O measurements from long-term δ^{18}O-S relationships in Bedford Basin. We especially sought residuals larger than the analytical uncertainty, which might be indicative of temporal variations of the δ^{18}O value of freshwater inputs.

The distribution of residuals (observed − predicted) from linear regressions of δ^{18}O-S, established using data from the entire time-series, are presented in Fig. 6 as a function of sampling date: one for near-surface waters (Fig. 6a; $\delta^{18}O = 0.15S - 6.23$; $r^2 = 0.92$) and one for near-bottom waters (60 m) (Fig. 6b; $\delta^{18}O = 0.19S - 7.47$; $r^2 = 0.36$). Also plotted is the analytical uncertainty of the δ^{18}O measurements (±0.05‰; dashed lines, Fig. 6).

For near-surface waters (Fig. 6a), 57% of the measurements fall within the range expected for analytical uncertainty around the annual average regression equation. From October 2012 to January 2013, the measured values fall consistently above the regression line and are more isotopically enriched than values predicted by the regression equation, with the residual magnitude exceeding the analytical uncertainty (Fig. 6a; > +0.05). From May to June 2013, there are a number of samples with measured values more isotopically depleted than the regression equation predicts, and again the residual magnitude exceeds the analytical uncertainty (Fig. 6b; > −0.05). The sign of temporal variations of residuals and their seasonal timing are consistent with local freshwater inputs identified in section 4.1.1; for example, the timing of spring melt which would deliver isotopically depleted winter runoff to the basin (see

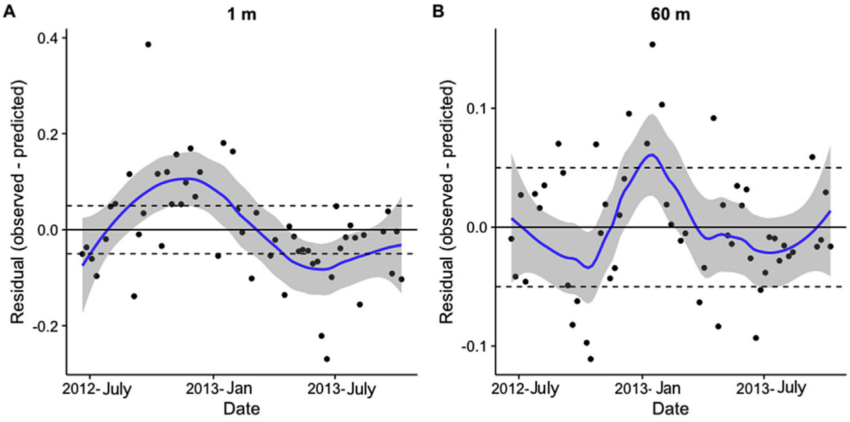

Fig. 6. Residuals (observed ($\delta^{18}O$) − predicted ($\delta^{18}O_o$)) from long-term (June 2012 to October 2013) average $\delta^{18}O$-S regressions of measurements made at: (a) 1 m and (b) 60 m in Bedford Basin. The dotted lines illustrate the range of variation around the regression line that would be expected for an estimated analytical uncertainty of ±0.05‰. The blue line is an applied LOESS smoothing curve, with a span of 0.5, with the grey shading identifying the confidence interval. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Fig. 3a). However, it is most notable that the temporal pattern of the residuals is barely distinguishable above the variability associated with analytical error (Fig. 6).

In the near-bottom waters (60 m; Fig. 6b), almost all (70%) of the residuals fall within the range expected due to analytical uncertainty, although from December 2012 to January 2013, the observed values appear to be more enriched (heavier) than predicted (Fig. 6b; > +0.05).

Overall, the very limited number of residuals that fall outside the range expected due to analytical uncertainty suggests that most of the variability of $\delta^{18}O$ in Bedford Basin can be explained by the mixing of a single, local freshwater end-member with saltier water derived from the Scotian Shelf. In near-surface waters, the impact of seasonal variation in the local freshwater end-member is detectable (Fig. 6a), whereas in deeper waters, seasonal variability of $\delta^{18}O$ residuals is even less marked. An enrichment in the measured values occurs in both near-surface and deep (60 m) waters over the fall and winter of 2012/2013. This enrichment begins in the near-surface waters in October 2012, but cannot be seen until December 2012 in the near-bottom waters (Fig. 6). These variations are at the limit of detection, and hence their significance is questionable. However the sequence and timing may suggest enrichment in $\delta^{18}O$ at both depths during fall and winter, in response to the local influx of isotopically enriched (heavier) freshwater at the surface, coupled with increased vertical mixing between surface and deep waters of Bedford Basin during winter in association with stronger winds and, especially, weaker stratification (Platt et al., 1972; Punshon and Moore, 2004). The lack of a corresponding signature of seasonally-introduced lighter isotopes entering near-bottom waters during summer may reflect the reduced vertical mixing associated with seasonal stratification. Continued time-series sampling over multiple years would be required to resolve these apparent seasonal patterns and test these ideas.

5. Conclusions

This time-series study of Bedford Basin has related seasonal variability in the oxygen isotope composition of precipitation, with variability in riverine inputs and the resulting freshwater end-member composition of surface and deep waters of a coastal fjord. On an annual basis, there was internal consistency, within uncertainty levels, between amount-weighted, annual averages of $\delta^{18}O$ in precipitation and river water and the end-member freshwater composition estimated from a $\delta^{18}O$-S regression of near-surface samples in Bedford Basin.

Although there was marked seasonal variability in the $\delta^{18}O$ of precipitation, especially associated with isotopically light snowfall in winter, the winter precipitation signal was not reflected clearly in the near-surface waters of Bedford Basin. This is likely the result of fractionation and exchange processes operating within the drainage basin prior to discharge to Bedford Basin, including sublimation of light isotopes from the snowpack, and mixing with freshwater derived from different seasons. Within Bedford Basin itself, the bulk of the data in near-surface waters are consistent with mixing of a single, effective, “local” freshwater source (with an average end-member $\delta^{18}O$ of −6.23‰, ±0.34) with an offshore-derived, saline Scotian Shelf end-member (with a $\delta^{18}O$ of −1.37‰ ± 0.08 at a salinity of 31.99 ± 0.18). Seasonal reductions of salinity in near-bottom waters (60 m) are consistent with mixing with this same, locally-derived freshwater end-member via vertical mixing. While this local freshwater source explains most of the variability of $\delta^{18}O$ in the near-bottom water, some of the observed variability requires the contribution of an additional freshwater end-member, consistent with end-members identified previously for the Scotian Shelf.

A detailed temporal analysis of residuals from a long-term average $\delta^{18}O$-S regression equation, show that most of the residuals lie within the range expected on the basis of analytical error. Surface waters show some evidence for a systematic pattern of residuals, which corresponds to the seasonal variation of $\delta^{18}O$ in local freshwater inputs. The residuals at 60 m suggest a barely detectable signal of mixing with locally-derived freshwater in winter, but the summertime isotopic signal is not detectable, likely due to reduced vertical mixing associated with summer stratification.

Overall, this study has shown that time-series measurements in Bedford Basin are consistent with the integrated impact of seasonal variations in the $\delta^{18}O$ of local precipitation as modulated and modified by hydrologic processes operating within the drainage basin. The short-term variation of the $\delta^{18}O$-S relationship within the Bedford Basin are damped relative to source water variations, so that the time-series measurements within the fjord provide integrated measures of both the local freshwater inputs and offshore, saline end-member. This study suggests that long-term measurement of $\delta^{18}O$-S relationships within Bedford Basin have the potential to provide a useful integrated measure of changes in hydrological processes operating at the drainage-basin scale over interannual and longer timescales.

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