Using ^{226}Ra and ^{228}Ra isotopes to distinguish water mass distribution in the Canadian Arctic Archipelago

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Abstract:

As a shelf dominated basin, the Arctic Ocean and its biogeochemistry are heavily influenced by continental and riverine sources. Radium isotopes (^{226}Ra, ^{228}Ra, ^{224}Ra, ^{223}Ra), are transferred from the sediments to seawater, making them ideal tracers of sediment-water exchange processes and ocean mixing. As the two longest-lived isotopes of the Radium Quartet, ^{226}Ra and ^{228}Ra are the two longer-lived isotopes of the Radium Quartet (^{226}Ra, $t_{1/2}$=1600y and ^{228}Ra, $t_{1/2}$=5.8y). Because of their long half-lives they can provide insight into the water mass compositions, distribution patterns, as well as mixing processes and the associated timescales throughout the Canadian Arctic Archipelago (CAA). The wide range of ^{226}Ra and, ^{228}Ra activities, as well asand of the ^{228}Ra/^{226}Ra ratios, measured in water samples collected during the 2015 GEOTRACES cruise, complemented by additional chemical tracers (dissolved inorganic carbon (DIC), total alkalinity (AT), barium (Ba), and the stable oxygen isotope composition of water (δ^{18}O)), highlight the dominant biogeochemical, hydrographic and bathymetric features of the CAA. Bathymetric features, such as the continental shelf and shallow coastal sills, are critical in modulating circulation patterns within the CAA, including the bulk flow of Pacific waters and the inhibited eastward flow of denser Atlantic waters through the CAA. Using a Principal Component Analysis, we unravel the dominant mechanisms and the apparent water mass end-members that shape the tracer distributions. We identify two distinct water masses located above and below the upper halocline layer throughout the CAA, as well as distinctly differentiate surface waters in the eastern and western CAA. Furthermore, we highlightidentify water exchange across 80°W, inferring a draw of Atlantic water, originating from Baffin Bay, into the CAA. In other words, this impliesThis underscoresillustrates the presence of an Atlantic water U-turn located at Barrow Strait, where the same water mass is seen along the northernmost edge at 80°W as well as along the south-eastern -most confines of Lancaster Sound. Overall, this study provides a stepping stone for future research initiatives

within the Canadian Arctic Archipelago, revealing how quantifying disparities in the distributions of ~~in~~ radioactive isotopes can provide valuable information on water mass distributions, flow patterns and mixing, ~~the potential effects of climate change~~ within vulnerable areas such as the CAA.

I: Introduction

I.I. General Background

Over the past 30 years, major research initiatives have been undertaken within the Arctic, highlighting this region's global importance and vulnerability to climate change (Prinsenberg and Bennett, 1987; Shadwick et al., 2013). One of the primary causes of this vulnerability is a modification of the regional hydrographic regime, characterized by cool, CO_2-charged (less alkaline) Pacific waters, that enter the Arctic Ocean via the Bering Strait, flowing along the southern parts of the Canadian Arctic Archipelago (CAA) and being dispersed into Baffin Bay. Previous studies have shown that these eastward flowing waters contribute significantly to carbon sequestration as well as instigate deep-water formation in the North Atlantic (e.g., Aagaard and Carmack, 1989; Burt et al., 2016a; Curry et al., 2011; Hamilton and Wu, 2013; Holland et al., 2001; Ingram and Prinsenberg, 1998; Rahmstorf, 2002; Shadwick et al., 2011a).

Although the various water masses delivered to Baffin Bay play a role in establishing and maintaining the global thermohaline circulation, little is known about the distribution, composition, and modes of delivery of water through the Canadian Archipelago. This study contributes to the knowledge base of circulation patterns in the CAA by using the radioactive radium isotopes ^{228}Ra and ^{226}Ra as well as dissolved inorganic carbon (DIC), total alkalinity (AT), barium (Ba), and the stable oxygen isotope composition of water ($\delta^{18}O$) as tracers of water mass distribution, mixing, and composition throughout the region. Moreover, we hope that this study will provide a foundation~~platform~~ for further investigations of how changes in environmental conditions within this vulnerable area will affect the distribution of these tracers, as well as biogeochemical cycles and circulation in the CAA.

I.II. Oceanographic Setting

Approximately 30-50% of the Arctic Ocean surface area (totaling to $9.5 \times 10^6 km^2$) is dominated by polar continental shelves (Coachman and Aagaard, 1974; Jakobsson, 2002; Rutgers van der Loeff et al., 1995; Shadwick et al., 2011b; Walsh, 1991; Xing et al., 2003). The CAA, a region of branching channels and straits that extends from approximately 120°W to 80°W is located in this shelf-dominated region (Fig. 1). Spanning only 65km across at its widest, this narrow, polar network provides a critical connection between the Pacific and Atlantic Oceans and facilitates the export of approximately one third of the Arctic Ocean’s outflowing water (Coachman and Aagaard, 1974; Hamilton et al., 2013; Hamilton and Wu, 2013).

Previous research has recognized the partition~~ed~~ing of the water column in the CAA into three salinity-defined water masses, the deepest and most saline being the Atlantic Layer (ATL, S_p >33.1), followed by the Pacific Upper Halocline Layer (UHL; 31 < $S_{P\text{~~p~~}}$ < 33.1), and finally the least saline and uppermost being the Polar Mixed Layer (PML; $S_{P\text{~~p~~}}$ <31) (e.g., Aagaard et al., 1981; Aagaard and Carmack, 1994; Bauch et al., 1995; Mathis et al., 2005; Shadwick et al., 2011a). All three water masses have been identified at both the eastern and western boundaries of the CAA whereas only the upper two layers (PML and UHL) are found throughout. The presence of a 200 m shoal at Barrow Strait (Fig. 1), that bridges the western and eastern regions, prevents the Deep ATL water mass from flowing eastward through the CAA (Jones, 2003; Macdonald et al., 1987; Newton and Coachman, 1974). As a result, the bulk eastward transport is composed of the cool, CO_2-charged (less alkaline) Pacific and fresh surface waters that flow from the Canada Basin to Baffin Bay through the CAA (Hamilton and Wu, 2013; Prinsenberg et al., 2009; Wang et al., 2012; Xing et al., 2003).

In addition to the bulk eastward transport through the CAA, the northern regions of the CAA host an occasional westward flowing counter-current during late summer (Peterson et al., 2012; Prinsenberg & Bennett, 1987; Prinsenberg et al., 2009; Rudels, 1985). This suggests that there may be the intrusion of Atlantic waters originating from Baffin Bay, moving into the CAA along the northern edge from the east, and possibly creating a “U-turn”~~, rerouting~~ as the westward current reroutes back into Baffin Bay along the southern edge. The importance of this “U-turn” will be discussed later in the results section.

I.III. Some considerations about the two long-lived Radium Isotopes

^{226}Ra and ^{228}Ra are the two longer-lived isotopes of the Radium Quartet (^{226}Ra, $t_{1/2}$=1600 y and ^{228}Ra, $t_{1/2}$=5.8 y) and~~. They~~ are ~~found~~ often present at readily detectable activities that are largely unperturbed by biological activity in seawater. ^{226}Ra and ^{228}Ra~~,~~ are thus often ~~allowing them to be used as nearly~~considered as conservative radioactive tracers (Charette et al., 2015a, 2016; International Atomic Energy Agency (IAEA), 2010; Moore et al., 1980). ~~Additionally, b~~Both long-lived Ra isotopes are formed from the decay of different Thorium (Th) isotopes ~~in terrestrial soils and marine sediments~~ (^{226}Ra is the daughter of ^{230}Th, whereas~~ile~~ ^{228}Ra is the daughter of ^{232}Th) in terrestrial soils and marine sediments~~ in sediments~~. Subsequently, ~~T~~they~~ and~~ are ~~subsequently~~ distributed to the ocean through riverine inputs, porewater advection, and diffusion across the sediment-water interface~~, primarily along coastlines or the bottom boundary layer~~ (Charette et al., 2015a). Since ^{228}Ra is more rapidly regenerated within sediments and its half-life is relatively short in comparison to ^{226}Ra~~er than ^{226}Ra~~, its activity in~~ is closely associated with~~ coastal and continental shelf regions decreases rapidly away from the source towards the open ocean. Thus, ^{228}Ra~~where its abundance decreases rapidly away from its source towards the open waters, thus generally~~ tracks~~ing~~ the sediment-ocean or shelf-ocean transition (van Beek et al., 2007; Burt et al., 2016b; Kadko and Muench, 2005; Kawakami and Kusakabe, 2008; Moore et al., 1980, 2008; Rutgers van der Loeff et al., 1995). For the purpose of this study, we assume that ^{228}Ra has no pelagic source. As~~WhileWhileLikewise,~~ ^{226}Ra is ~~also~~ released from the sediment and disperses into the water column through advective and diffusive mixing (Charette et al., 2015a; International Atomic Energy Agency (IAEA), 2010; Rutgers van der Loeff and Moore, 1999).~~,~~ ~~but given~~ its longer half-life allows it~~for, ^{226}Ra~~ to be~~can be~~ distributed over great distances, often decaying within the oceanic water column (Charette et al., 2015a; International Atomic Energy Agency (IAEA), 1988). A slight enrichment~~can be seen~~ in Pacific Ocean deep waters, relative to deep waters of the Atlantic Ocean~~,~~ is primarily attributed to ^{226}Ra uptake in the co-precipitation of barite ($BaSO_4$) or its uptake~~n the production of~~ by biological silicate or calcium tests (van Beek et al., 2007; Charette et al., 2015b; Moore and Dymond, 1991). With this exception~~in mind~~, ^{226}Ra displays~~reveals~~ a "nearly" conservative distribution in the oceans, thus facilitating its use as a long-term pelagic-based tracer of water masses and of shelf inputs ~~With the exception of a slight enrichment in Pacific Ocean deep waters relative to the deep waters of the Atlantic Ocean~~ (Broecker et al., 1967; Charette et al., 2015a; Chung, 1980).~~, ^{226}Ra reveals a nearly conservative distribution in the oceans facilitating its use as a long-term pelagic-based tracer of water masses and of shelf inputs~~. These characteristics allow the two long-lived Ra isotopes to be used as radioactive geochemical tracers to distinguish ~~water-mass~~water mass sources and their distribution patterns~~, in our case~~ within the CAA.

II. Methods

II.I. Sample Collection

During the summer of 2015 Canadian GEOTRACES cruise, 64 water samples were collected throughout the Canadian Arctic Archipelago aboard the icebreaker CCGS Amundsen at 17 different stations as a subset of the overall biogeochemical sampling (Fig. 1). Samples for dissolved inorganic carbon (DIC), total alkalinity (AT), barium (Ba), the stable oxygen isotope composition of water ($\delta^{18}O$), and Ra isotopes were collected at various depths from the surface to 1000m on the up-cast of a rosette system equipped with (24) 12-L Niskin bottles. Surface samples (2-12m) for Ra were collected using an onboard pump collecting ship-side. In addition, temperature and salinity (S_P) measurements were recorded on the downcast by a Sea-Bird SBE 9 (Seasave V 7.23.2) CTD throughout the water column. The CTD salinity-probe measurements were calibrated post-cruise using a Guidline salinometer in the home laboratory against taken discrete samples taken directly from the Niskin bottles into 250 mL screw-cap HDPE bottles. DIC and AT samples were collected directly from the Niskin bottles into 250mL or 500mL borosilicate glass bottles to which 100µL of a saturated $HgCl_2$ solution was added before being sealed with ground-glass stoppers, Apiezon® Type-M silicon grease and elastic closures (Burt et al., 2016a). The bottles were then stored in the dark at room temperature or 4°C until they could be processed on board. A VINDTA 3C (Versatile Instrument for the Determination of Titration Alkalinity, Marianda) was used to firstly analyze the DIC DIC samples for DIC by coulormetric titrations, and thefor secondly determine AT by potentiometric titrations (Shadwick et al., 2011a). A calibration of the instrument was performed against certified reference materials (CRM) provided by Andrew Dickson (Scripps Institution of Oceanography) and the reproducibility of the DIC and AT measurements was better than 0.1%.

Each Ra sample (105-215L) was sequentially pre-filtered through 10µm and 1µm filters, either directly using the ship's pump, or a high-volume pump connected to the Niskin bottles. The Ra isotopes were then pre-concentrated by elution through manganese dioxide (MnO_2)-coated acrylic fiber cartridges at a constant flow rate of 1 L min^{-1} (Charette et al., 2001; Moore and Reid, 1973; Rutgers van der Loeff and Moore, 1999). To verify the extraction efficiency of the MnO_2 fiber cartridge, a second fiber-filter was occasionally mounted in series. ^{224}Ra was then determined using a Radium Delayed Coincidence Counter (RaDeCC) (Moore, 2008), thatwhich had been calibrated against an IAEA (International Atomic Energy Agency) distributed reference material. The detection limit was estimated to 3 atoms L^{-1} (0.05dpm $100L^{-1}$) for ^{224}Ra (see for details Moore, 2008; Moore and Arnold, 1996). No ^{224}Ra activity could be detected in any of the second cartridges mounted in series, confirming 100XX%

extraction efficiency. ~~Samples~~ The Mn-fibers were then shipped to the Woods Hole Oceanographic Institution to be ashed at 820°C for 16h, homogenized and transferred to counting vials (Charette et al., 2001). Well-type gamma spectrometers (Canberra and Ortec high purity germanium) were used to quantify ^{226}Ra (via ^{214}Pb @ 352 keV) and ^{228}Ra (via ^{228}Ac @ 338 and 911 keV) (International Atomic Energy Agency (IAEA), 2010). Each detector was calibrated with Mn-fiber ash spiked with ~~a~~ NIST-certified reference material #4967A (^{226}Ra) or a gravimetrically-prepared $Th(NO_3)_4$ solution with the ^{228}Ra daughter in secular equilibrium with its parent ^{232}Th. Detection limits, determined using the Currie Hypothesis test (De Geer, 2004), were determined to be 0.2 dpm for both ^{226}Ra and ^{228}Ra (Gonneea et al., 2013), which is equivalent to ~0.15 dpm ~~100 L~~ $100L^{-1}$ for a typical 130 L sample.

Barium (Ba) concentrations were determined in water transferred directly from the Niskin bottles to 30mL HDPE plastic bottles containing 15µL of concentrated ultrapure hydrochloric acid (Thomas et al., 2011). Each subsample was then analyzed by Isotope Dilution using Sector Field Inductively Coupled Plasma Mass Spectrometry (SF-ICP-MS, Element 2, Thermo Finnigan) in Brussels. The instrument was run in the low mass resolution mode m/Δm =300. Limit of detection and limit of quantification based on blank analyses were: 0.06 and 0.20 nM, respectively (LOD = 3 X s.d. blank; LOQ =10 X s.d. blank). Reproducibility of multiple measurements of reference materials (SLRS5; SLRS3; OMP) was ≤ 2.5%. Details of the instrument's operational conditions are given by Thomas et al. (2011). The barite saturation state (Q_i) is the ratio of the aqueous barium and sulfate ion activity product ($Q_{(BaSO4,\ aq)}$) to the barite solubility product (K_{Sp}):

$$\textit{Saturation State } BaSO_4(Q_i) := \frac{Q_{BaSO_4 aq}}{K_{Sp(Barite)}} \qquad \text{(eq. 1)}$$

As described in greater detail by Thomas et al. ~~.~~(2011), Q_i ~~has been~~was computed according to~~after~~ Monnin (1999) and Hoppema et al. (2010).

Samples destined for measurements of the stable oxygen isotope composition of seawater ($\delta^{18}O$) were taken directly from the Niskin bottles into 13mL screw-~~-~~cap plastic test tubes (Lansard et al., 2012). The samples were analyzed at the GEOTOP-UQAM stable isotope laboratory using the CO_2 equilibrium method of Epstein and Mayeda (1953) on a Micromass Isoprime universal triple collector isotope ratio mass spectrometer in dual inlet mode (Mucci et al., 2018)~~.~~ at the GEOTOP-UQAM Light Stable Isotope Geochemistry Laboratory. The data were normalized against three internal reference waters, themselves calibrated against Vienna Standard Mean Ocean Water (V-SMOW) and Vienna Standard Light Arctic Precipitation (V-SLAP). Data are reported on the δ-scale in ‰ with respect to V-SMOW, and the average relative standard deviation on replicate measurements is better than 0.05‰.

II.II. Principal Component Analysis

~~The~~ Principal Component Analyses (PCA) were performed to quantitatively determine the correlation between variables as well as the affinity between each of the samples to arbitrary components, while reducing the effects of random variation by using a correlation matrix (Gunasekaran and Kasirajan, 2017; Jolliffe and Cadima, 2016; Pearson, 1901; Peres-Neto et al., 2003). ~~PCA is measured in Eigenvalues and Eigenvectors, quantitative measures of the relative variation of variables along the axes as well as the loadings (or coefficients) of each variable to the associated axes, respectively. Furthermore, PCA creates "best fit" linear relationships between points within arbitrary space, providing a useful statistical tool to distinguish associated trends, reduce dimensionality, and uncover relationships between related variables within a three-dimensional space. or~~ In this study, associated or derived variables such as the radium isotopic ratios were excluded from the PCA due to the congruency with other incorporated variables. Prior to statistical analysis, the variables from each station and depth were transformed to fit a near-normal distribution and normalized to satisfy the parameters of the analysis. Interpolations of the $\delta^{18}O$ and Ba data were made with respect to salinity, as samples were not collected at every depth at each station. The interpolations were verified relative to the original data by means of linear regression and comparison of slopes. Only three surface data samples were interpolated for Ba samples, alleach from within Baffin Bay. After interpolation and normalization, each sample was categorized by depth: Surface, Middle Depth, Deep Archipelago, or Deep Atlantic, ranging from 0-20m, 21-80m, 81-500m and >500m, respectively.

In addition, quantitative analyses of the PCA results were conducted by a broken stick analysis as a means to distinguish the loading significance of each variable. To this end, eigenvectors were scaled to V-vectors (product of eigenvector multiplied by the square root of the specific eigenvalue) and V^2 vectors (V-vectors^2) (Jackson, 2004; Peres-Neto et al., 2003). End--members were calculated for each of the variables significantly loading on PC1 from the derived partial values (eEigenvector for associated variable multiplied by the PC score for that sample). Each partial value was then de-normalized and back-transformed, thus deriving a refined rendition of the original data set (Appendix 1). Lastly, linear regressions of each variable, with the exception of ^{228}Ra, against the practical salinity were plotted to express robust end-member relationships from within the previously categorized salinity-defined water masses present throughout the CAA. For ^{228}Ra, the end-member was derived from linear regression to the practical salinity where S=25. We report the respective end-members as "apparent end-members", as they resemble the mean end-member properties in the CAA. For example, an apparent freshwater end-member at null practical salinity (S_P=0) would be composed of various individual river and meteoric water end-members, in consideration of their relative weights in this composite. As ^{228}Ra originates from shelf sediments, which in our study are primarily located in waters at the lower salinity ($S_P \approx 25$) range of the CAA, the the ^{228}Ra end-member was derived from the linear regression to S_P = 25 (eg. Rutgers van der Loeff et al., 2003).

III. Results and Discussion

III.I. Water Mass Properties

Surface values of S_p, density, DIC, AT, and $\delta^{18}O$ were found to increase from west to east through the CAA (Figs. 1, 2a, c, d, f, g, Appendix 2). This trend was extended to the temperature profiles taken throughout the CAA, with the exception of station CAA5, which was found to closely resemble the temperature profile of CAA3 (Fig. 2b, Appendix 2). Prinsenberg and Bennett (1987) reported similar trends ~~in results from~~employing samples collected in 1982 across Barrow Strait, a sill less than 200m deep located roughly between 105°W to 90°W, where analogous transects for salinity and temperature were recorded throughout the surface layer (Fig. 1). This is both the widest and shallowest section of the CAA. It is responsible for restricting the eastward flow of Deep ATL waters found in the Western ~~Canadian Basin~~Canada Basin and inhibiting high salinity (S_p > 33.1) ATL water within Baffin Bay from venturing westward (Hamilton and Wu, 2013; Jones, 2003; Prinsenberg, 1982; Prinsenberg and Bennett, 1987; Shadwick et al., 2011a; Yamamoto-Kawai et al., 2010).

In c~~C~~ontrast~~ing these trends~~, dissolved Ba, ^{226}Ra, and ^{228}Ra ~~concentrations~~ ~~have been found to~~ decreased eastward both at the surface as well as at the mid-depth maximum. This ~~, which~~ is assumed to reflect~~suspected to a result~~ from ~~of~~ the elevated flow rates, increasing distance from their source within the CAA, and proximity to the Ba- and ^{228}Ra-depleted ATL waters in Baffin Bay (Figs. 2e, 3, Appendix 2 and 3) (Thomas et al., 2011). Most often~~In general~~, the $^{228}Ra/^{226}Ra$ ~~isotopic~~ ratio ~~(Fig. 3c)~~ decreases with depth (Fig. 3c), but occasionally follows a more complex spatial pattern, which will be ~~unraveled at the end of this paper~~discussed later.

~~The property-property diagrams of~~~~P~~Positive relationships between DIC and AT vs. S_p ~~display strong positive relationships in surface waters~~ (~~see~~ Figs. 4 and 5a for regression intercepts and error~~with $S_{p=0}$ intercepts of 371 µmol DIC kg^{-1} and 492 µmol AT kg^{-1}, respectively~~)~~. This~~ ~~implies~~ indicate the ~~addition~~ importance of freshwater inputs ~~(low in TA and DIC)~~ ~~by means of~~from sea-ice melt (SIM) and Meteoric Water (MW, surface runoff and direct precipitation). These freshwater additions contribute ~~, can be~~ ~~thus attributed to the PML~~to low ~~T~~AT and DIC found in the surface PML waters of the stations west of 96.5°W (Figs. 1, 2, 4, 5c). Whereas both DIC and AT concentrations increase along the surface from west to east throughout the CAA, the hThe highest DIC concentrations were observed at the pycnocline of the western most station (CB4) (Fig. 2, and 4,Fig. 5a, and Appendix 2). This maximum in metabolic (respiratory) DIC decreasesd slightly eastward due to the increasing contribution of low-DIC ATL waters (Fig. 5a) (Shadwick et al., 2011a). Similar results were observed for AT, although ATAT, void oflacking the metabolic maximum witnessed within the DIC samples, concentrations were also found toalso increased both linearly with depth and eastward but without (Figs. 2d, g, 4). This is explained by the concomitant increase in AT and S_p values, rather than a decreasepreciation associated with metabolic activity, thus distinguishing AT from DIC (Burt et al., 2016a; Shadwick et al., 2011a; Thomas et al., 2011).

Despite the intrusion of deep Atlantic Ocean waters throughout the CAA (Jones, 2003; Newton and Coachman, 1974), CB4 (in the Canada Basin) displays different ATL, UHL and PML water-masswater mass characteristics than those observed at stations within the CAA (Appendix 2 and 3). Hence, for the remainder of this study, we will omitignore data from CB4 in our discussion of the circulation in the CAA, although we will return to the role and positioning of CB4 in relation to the CAA waters at a later stage of the paper, particularly in relation to the $^{228}Ra/^{226}Ra$ ratio.

In order to identify water-masswater mass distributions and mixing regimes within the CAA, DIC was normalized to a constant salinity (DIC_{norm}) (eq. 2). DIC_{norm} is a powerful tool to gain a better understanding of water mass characteristics and distributions by eliminating This approach accountscontrols for the influence of fresh water inputs, thereby and thus highlighting alternative possible non-conservative behaviorbehaviourcontrolling parameters such asrelated to biological processes at the time scale of mixing (Friis et al., 2003; Shadwick et al., 2011b).

$$DIC_{norm} = ((DIC_{measured} - DIC_{S=0}) * S_{measured}^{-1}) * S_{reference} + DIC_{S=0} \quad \text{(eq. 2)}$$

~~The distribution of~~ DIC$_{norm}$ ~~displays an~~decreases eastward ~~decrease~~ in surface waters along the eastward bulk flow throughout the CAA, consistent with observations that surface DIC values were lowest in the w~~W~~estern samples (Figs. 1, 2g, h, 5b, c). The reversal in trend reflects the decrease in accumulated respiratory DIC as waters flow ~~longitudinally~~ eastward through the CAA. The presence of two distinct~~, non-mixing~~ ~~water-mass~~water masses was~~ere~~ highlighted ~~through~~by the DIC$_{norm}$–S$_p$ relationship, distinguishing the surface (PML) from subsurface samples (Figs. 1 and 5c), within each of which mixing processes control the DIC$_{norm}$ distribution. The two water masses themselves, however, hardly mix. ~~These results confirm the previous distinction proposed,~~ ~~s~~A similar water mass grouping has been proposed, based on ~~the~~ Ba distributions~~ of Ba, between~~ in waters above and below the UHL ~~across the pycnocline~~ (Figs. 2, 5b, c) (Shadwick et al., 2011a; Thomas et al., 2011).

III.II. The use of Radium Isotopes as Water Mass Tracers

Radium isotopes, specifically ^{226}Ra, ^{228}Ra and their ratio (^{228}Ra/^{226}Ra), were used as proxies to reconstruct the water mass distribution throughout the CAA. Like the chemical constituents DIC, AT, δ^{18}O and Ba, as well as the stable oxygen isotopic composition of waters, Ra isotope activity and ratios were found to vary between stations, with depth, as well as across water masses (Appendix 3). The highest ^{228}Ra activities were observed at the surface, particularly at the shallow stations 312 and 314, located in the center of the CAA, ~~and thus more strongly influenced by~~indicating~~reinforcing~~ the ~~sedimentary~~ ^{228}Ra ~~release~~ source in coastal sediments (Figs. 1, 6). Lower ^{228}Ra activities were found at higher salinities and depths, comparable to values reported by Burt et al. (2016b) in the North Sea, where elevated ^{228}Ra activities were present within the shallow, lower salinity waters. Like the DIC$_{norm}$-S$_p$ relationship, the ^{228}Ra- S$_p$ relationship (Figs. 5b, c, 6) reveals two distinct water masses that distinguish~~separate~~ the surface (PML) from subsurface waters. For the surface sample grouping, a negative slope (^{228}Ra-S_P) was obtained (slope= -2.467x), whereas for the deeper samples a less negative slope was found (slope= -0.4854x). The more negative slope, associated with the surface samples collected throughout the CAA, indicates that the system is heavily influenced by the influx of ^{228}Ra from the CAA shelf sediments~~.~~ (Fig. 6). In contrast, the slope derived from the ^{228}Ra activities recorded in the deeper waters of the CAA may imply that these samples originate from an open ocean setting, with minimal (or much less) contact with continental shelf or coastline sediments over the past few decades. As noted earlier (Figs. 3, 6b), the ^{228}Ra activities decrease from west to east through the CAA in both the surface and deep samples. These values are interpreted as reflecting the mixing of Pacific waters with Atlantic (Baffin Bay) waters east of Barrow Strait. We provide a more detailed analysis of the ^{228}Ra activity distribution pattern below (section III.III.I.II).

III.III. Characterizing Water Masses and Isolating End-Members through Principal Component Analysis (PCA)~~.~~

Further investigation of the dominant ~~water-mass~~water mass patterns was undertaken through Principal Component Analyses (PCA). The first and second principal components (PCs) accounted for 59.1% and 17.5% (total 76.6%), respectively, of the variability in the data (Fig. 7, Table 1). The third PC accounted for a further 13.2% of the variability (89.8% in total). The fourth and fifth PCs together accounted for less than 10% of the variability and were not included in subsequent analyses. PC1, in turn, was inverted to establish apparent end-members of the source waters found in the CAA (section III.III.II.I, see methods II.II).

III.III.I Qualitative Analysis of PCA

III.III.I.I Surface Water Mass Distinction

The first PC (PC1) loaded very heavily on salinity, AT, and $\delta^{18}O$, accounting for 94–97% of the variability in each (see Table 1). It also loaded heavily on DIC and ^{228}Ra (67% and 66% of variability) and less heavily on Ba (37%). The latter five parameters were all inversely correlated with salinity (Fig. 7). The second PC (PC2) loaded heavily on temperature (83% of variability) and relatively weakly on DIC (a further 22% of variability for a total of 89% between PC1 and PC2) and Ba (34%, for a total of 71% between PC1 and PC2). There is a correlation between temperature and DIC ~~were directly correlated~~ (Fig. 7), so the component of DIC accounted for by PC2 cannot be ascribed to temperature-dependent solubility. The third PC (PC3), which only accounted for 13.2% of the variability in the data, loaded on ^{226}Ra (74% of its variability) and was the only PC that did so.

The ordination of samples on PC1 and PC2 shows a strong separation between surface and mid-depth samples vs. deep samples (Fig. 7), reflecting the consistent differences in their parameter values (Figs. 2-6). Variability within surface and subsurface layers was examined by re-running the analysis, using only these data, to minimize the influence by the systematically-different deep-water data on their ordination. The restricted analysis retained most of the parameter relationships observed in the full PCA (Fig. 7, 8). The first PC explained slightly more of the variability (63.5% vs. 59.15%), while the second PC explained slightly less (14.7% vs. 17.5%), for a total of 77.2% vs. 76.7%. There was strong loading of salinity, AT, δ^{18}O, DIC and ^{228}Ra on PC1, with the latter four being inversely correlated with salinity. In contrast, ~~though,~~ temperature was strongly correlated with salinity rather than orthogonal to it and Ba was strongly loaded on PC2.

The re-ordination of the surface and mid-depth data indicates a strong geographic separation of the samples on PC1 (Fig. 8), which is also evidenced by temperature-salinity and ^{228}Ra/^{226}Ra-DIC plots (Fig. 9). The first surface group comprises samples from the eastern edge of the CAA, under the influence of Atlantic waters, which enter the CAA via Baffin Bay and Lancaster Sound. The second group comprises the PML-influenced surface samples from the two southern interior CAA stations

(312 and 314) and the northwestern CAA stations (CAA4-CAA7), and the mid-water samples (also from s~~S~~tations 312 and 314). It is worth noting that the outer-most surface west samples, likely best visible in the bottom left quadrant of Fig. 9b, ~~are sampled~~came from station CB4. This attribution will be explained later with reference to the apparent end-member properties.

III.III.I.II Distinction of Deep-Water Masses and Indication of Flow

The ordination in the initial PCA is analogous to a T-S diagram, given that PC1 loads on salinity and its covariates and PC2 loads most heavily on temperature. There are very strong similarities between deep-water samples collected in Baffin Bay (Fig. 7; Deep ATL). The deep-water samples within the CAA are ordinated along a gradient between the Deep ATL samples and an end-member that would have negative factor loadings on PC1 and PC2. This would likely be Pacific water. The two deep samples from the westernmost station, CB4, are anomalous (Fig. 7). Their ordination suggests that they are an end-member for the Deep ATL water, ~~this is due to~~likely Deep ATL waters that flow west past Svalbard, before crossing the Lomonosov Ridge and accumulating in the Canada Basin (Coachman and Barnes, 1963; Newton and Coachman, 1974).

Paradoxically, samples collected at Station CAA3 are found at both ends of this trend, having both the highest and lowest similarity to samples collected in the Deep ATL ~~samples of samples~~ from within the CAA. The three deep-water samples collected at CAA5 are intermediate between the Deep ATL and deep CAA3 waters~~ compared to the remainder of the CAA~~. The very strong similarity between the deep-water samples collected at CAA3 and those from Baffin Bay indicate~~s~~ that they are ATL water that recirculated counter-clockwise around Baffin Bay, ~~and~~ combined with Arctic outflow through Nares Strait~~-~~ (Bâcle et al., 2002; Curry et al., 2011; Lobb et al., 2003). A third PCA was performed excluding the alkalinity (that ~~which~~ clearly expresses the bulk eastward flow~~dilution of freshwater~~ following the ~~dominant bulk eastward flow~~), to visualize the transition of Deep ATL water as it mixes with the UHL in the CAA (Fig. 10). Results of this analysis reveal that the Deep Arch water mass at stations CAA1, CAA3 and CAA5 are more closely linked to the Deep ATL group, implying that they are in fact part of the Deep ATL water mass (Fig. 10). This suggests that there is an intrusion of ATL water along the northern edge of the CAA. This westward flow with a speed of 2.2 cm/s was observed by Prinsenberg and colleagues (2009) and is weaker than the dominant eastward current flow (15.3 cm/s). This mild inflow of water along the northern edge of the Archipelago is then assumed~~suspected~~ to be redirected and exits back to Baffin Bay through the southern Archipelago station (CAA3).

There is further support from observations of dissolved Ba and the barite saturation states (Q_i) along the north-to-south transect across the eastern Lancaster Sound (CAA1, CAA2, 323, 324, CAA3) (Fig. 11a, b). An increase in Ba and Q_i was observed from north to south at the surface as well as at depth. Lower Ba concentrations and barite saturation states have been observed in Baffin Bay waters, which are fed by the West Greenland Current. In contrast, the continentally-impacted waters of~~from~~ the CAA are characterized by significantly higher values for the two Ba properties, such that the origin of the waters from the Atlantic and~~or~~ the CAA can clearly be discriminated~~ clearly~~ (Thomas et al., 2011). Furthermore, substantially lower values of the ^{228}Ra/^{226}Ra ratio at depth (Fig. 11c) indicate the inflow of Atlantic water on the northern side of Lancaster Sound as well as its outflow along its southern side. Compatibly~~ongruently~~, t~~T~~he ~~same~~ pattern is again revealed by the~~ by~~~~with~~ ^{226}Ra/Ba (Fig. 11d), which is dominated by the ^{226}Ra variability (Fig 13b,c). The lowest ^{226}Ra/Ba value reflects the inflow of low ^{226}Ra waters from the Atlantic Ocean (Fig. 11d), as the observed ^{226}Ra activities (8-9 dpm 100L^{-1}, Fig. 3a) are in the same range as those measured in the surface waters of the Atlantic Ocean (Le Roy et al., 2018), part of which feed into the West Greenland C~~c~~urrent. Since ^{226}Ra activities reveal a much larger north-to-south gradient across Lancaster Sound than Ba does (Figs. 2e, 3a), the~~ seemingly opposing~~ discrepancy in ~~strength of~~ the gradients shown in Figs. 11c and 11d is~~are~~ dominated by changes in ^{226}Ra. The waters transiting through the CAA and exiting on its southern side are enriched with both

^{228}Ra and ^{226}Ra as they interact with the shallow sediments. The differential~~Differences in the two isotopes'~~ half-lives of the two isotopes generate~~ underlie~~ the gradient in the ratio (Figs. 11c, 12) generally revealing higher ^{228}Ra fractions the more recent the contact of that particular water mass with shelf sediments. The Ba and Ra data are consistent in revealing the~~indicating~~ bi-directional flow that link~~ings~~s the northern and southern stations along 80°W. This can be attributed to the counter-clockwise, cyclonic circulation found throughout Baffin Bay (Figs. 11,~~ and~~ 13, and 14).

A closer look at the ^{226}Ra-Ba relationship reveals the association between ~~water-mass~~water masses observed in the CAA and those of the surrounding oceans, in particular~~ to~~ the Atlantic Ocean (Fig. 13). ^{226}Ra and Ba appear to vary without an apparent clear relationship. The highest surface values of the ^{226}Ra/Ba~~ ratio~~ are observed in the interior of the CAA, wh~~ile~~ereas the lowest are found in the Canada Basin and the eastern side of the CAA (Fig. 13d). The direct relationship between ^{226}Ra activities and Ba concentrations shows that the Arctic samples with a $S_P > 34$, i.e., waters of Atlantic origin, fall along the relationship established by Le Roy et al. (2018) for the Atlantic, or reported by van Beek et al. (2007) for the Sargasso Sea~~.~~ (Fig. 13a). This relationship, in turn, is similar to the one for the world ocean established from the GEOSECS data-base (Le Roy et al., 2018). This implies that the deep Lancaster Sound samples, as well as the deep Canada Basin samples, can similarly be linked to an Atlantic origin. The remaining CAA samples, with a $S_P < 34$ display a clear deviation from this relationship and towards higher Ba values (Fig. 13b), which can be attributed to the high Ba runoff from rivers draining into the CAA (Guay and Falkner, 1997a). The open-ocean ^{226}Ra/Ba ~~ratio~~ has been reported to be relatively constant at about 2.2-2.5 dpm (µ~~10^{-6}~~mol^{-1}), with elevated values observed only near deep-ocean sediments (van Beek et al., 2007; Le Roy et al., 2018). In contrast, the CAA data show a wider range of ^{226}Ra/Ba values, which appear to be strongly controlled by the ^{226}Ra activity, rather than variability of the Ba concentration (Fig. 13b,c). The ^{226}Ra/Ba~~ ratio~~ in water masses of Atlantic origin ($S_P > 34$) are offset toward higher ratios for a given ^{226}Ra activity, a consequence of the relatively higher Ba content of the water masses transiting through the CAA. The highest $^{226}Ra/Ba$ surface values were observed in the interior of the CAA, whereas the lowest ones were measured in the Canada Basin and the eastern side of the CAA (Fig. 13d).

III.III.II. Interpretations of PC1 and PC2

III.III.II.I.: Principal Component One: Advection / Land-Ocean Transition

PC1 was found to correlate significantly with S_p, DIC, AT, $\delta^{18}O$, ^{228}Ra and Ba, suggesting that this axis represents the land-ocean gradient, i.e., the advective (estuarine) mixing regime of fresh and salt water (Table 1, Fig. 7). This interpretation is consistent with our previous attribution of the longitudinal, eastward increase in S_p, DIC, and AT of surface waters through the CAA to a decreasing coastal influence (Figs. 3, 4, 5).

Here, the ^{228}Ra activity has to be viewed from a somewhat different perspective, as the sedimentary/shelf sources also reside in the low salinity range of our samples (S_p~ 25-30), but does not align with any riverine source (e.g., Rutgers van der Loeff et al., 2003, see below). Therefore, in regards to the PC1 axis, ^{228}Ra relative to ^{226}Ra, represents the coastal, shelf to open ocean transition, decreasing in activity laterally as waters primarily follow the bulk eastward flow and are transported away from the sedimentary source within the CAA (Figs. 12, 13, see also Charette et al., 2016). Accordingly, the fresher ^{228}Ra end-member was defined with S_P = 25.

The loading of PC1 on $\delta^{18}O$ and Ba is also high, particularly for $\delta^{18}O$. These variables allow us to discriminate between freshwater sources (MW vs. SIM) while also demonstrating a clear mixing gradient along the land-ocean transition (Guay et al., 2009; Macdonald et al., 1999; Yamamoto-Kawai et al., 2010).

We exploit the relationships of salinity to the individual properties derived from PC1 in order to define the freshwater and marine (saline) end-members (Table 2). Again, these end-members should be considered as "apparent"~~ end-members~~, as these represent observed mean end-member properties, and not end-member water masses, for example, end-members of individual rivers. "Apparent" end-members for each of the significant loading variables associated with PC1 were calculated (Table 2). It should be noted that ^{226}Ra was included in the PC1 analysis even though it primarily weighs on the PC3 axis (73.8%), not the PC1 axis. This is because salinity is not a significant loading factor on the PC3 axis (Table 1). Therefore, with the exception of PC3, ^{226}Ra ~~next ~~most closely associates with the PC1 axis (18.6%), thus allowing for PC1 to be used to establish the ^{226}Ra "mixing" ~~endmember~~end-member (Table 1).

The computed "apparent" end-members sit along the mixing curve, many located "halfway" between the SIM and MW values reported in the literature (e.g., Cooper et al., 2005; Guay and Falkner, 1998; Macdonald et al., 1989; Shadwick et al., 2011a; Thomas et al., 2011). These "apparent" freshwater end-members (S_P=0) thus reflect~~mirror~~ the combination of freshwater sources. Since both the MW and SIM are equally represented in the "apparent" DIC, AT and Ba end-members, it can be assumed that the freshwater end-member is located within the western portion of the CAA, as the freshwater contribution to the eastern ATL water mass is dominated by SIM, with little to no MW (Shadwick et al., 2011a, 2011b). The "apparent" end-member surface values for $\delta^{18}O$ were found to closely resemble MW (rather than the larger SIM values), with MW being, in essence, the dominant source of freshwater (Table 2) (e.g., Thomas et al., 2011). The end-members associated with the UHL and ATL were also found to be very similar to those reported in the literature, especially AT, Ba, and ~~$\delta^{18}O$ (Guay and Falkner, 1997a, 1997b; Macdonald et al., 1989; Shadwick et al., 2011a; Thomas et al., 2011; Yamamoto-Kawai et al., 2010).~~ $\delta^{18}O$ (Guay and Falkner, 1997a, 1997b; Lansard et al., 2012; Macdonald et al., 1989; Shadwick et al., 2011a; Thomas et al., 2011; Yamamoto-Kawai et al., 2010). The "apparent" DIC end-members for these water masses do not closely concur with literature values, nor do those produced here (Fig. 5a), as the DIC end-member associated with the UHL is expected to be larger than that of the ATL ~~(e.g., Shadwick et al., 2011a).~~ (e.g., Lansard et al., 2012; Shadwick et al., 2011a). We argue that this is due to the impact of biological processes, which cannot be resolved solely from mixing-conservative properties. These characteristics will be discussed within the context of PC2 in section III.III.II.II. Furthermore, this result may be due to the normalization required for the PCA linearization of the Deep ATL and Arch samples, thus diminishing the characteristic DIC maximum at the pycnocline. Lastly, we propose apparent end-members for ^{226}Ra and ^{228}Ra in the CAA. The highest ^{228}Ra and ^{226}Ra activities were found in the fresh(er) sources attributed to the surface samples collected in the western CAA (Fig~~ure~~. 6). The S_P = 0 end-member for ^{226}Ra (25.2 dpm 100L^{-1}) is consistent with the effective ^{226}Ra end-member for the Mackenzie River (26.1 dpm 100L^{-1}; Kipp et al., 2019), which contributes to the freshwater budget of the CAA. Smith et al. (2003) reported a Beaufort shelf end-member with a ^{228}Ra activity of 12 dpm 100L^{-1} at S_P= 2 and a ^{228}Ra/^{226}Ra of ~1. Therefore, with a shelf apparent ~~endmember~~end-member for ^{228}Ra of 22.4 dpm 100L^{-1} at S_P= 25 (Table 2) and a ^{228}Ra/^{226}Ra of ~2, the shelf sediment influence of ^{228}Ra in the CAA is ~~quite~~ conclusive.

Similar apparent end-member ^{226}Ra activities were observed within the open waters of the UHL and ATL, while substantially lower ^{228}Ra activities were recorded in the ATL (Table 2). Rutgers van der Loeff et al. (2003) reported ~~a~~ high-salinity ^{226}Ra end-members in a similar range (~6-9 dpm 100L^{-1}) to the apparent ones reported here, wh~~ile~~ereas the apparent ^{228}Ra end-members obtained in our study~~here~~ are clearly lower than those reported by Rutgers van der Loeff et al. (2003, ~3.2-15.4 dpm 100L^{-1}). An obvious explanation for this discrepancy may be the circulation history of the respective water masses, as the Atlantic end-member reaches the~~in the~~ CAA much later than the Eurasian sector of the Arctic Ocean. Thus the ~~most likely has a~~ longer circulation history of the ATL waters observed in the CAA allows for a substantial decay of ^{228}Ra compared to the ATL waters observed ~~than on~~on the Eurasian side ~~due to both the decay and accumulation (from shelf interaction) of ^{228}Ra~~. Furthermore, the salinity of the samples reported in this study are higher than the samples measured by Rutgers van der Loeff et al. (2003), implying the presence of a stronger ATL component in our samples. The differences between the high salinity~~,~~ apparent~~,~~ ^{226}Ra and ^{228}Ra end-members~~,~~ might reflect their vastly different half-lives, allowing for an appreciable ~~tangible~~ decay of ^{228}Ra at oceanic transport timescales in contrast to the "nearly conservative" ^{226}Ra. Coinciding with the previous result, higher variability in ^{228}Ra was seen throughout the water column, whereas ^{226}Ra activities varied only slightly. Overall, the identification of Ra end-members in the region highlights the Ra sources and transport pathways throughout this complex coastal/shelf environment.

We use the derived apparent end-member properties of ^{228}Ra and ^{226}Ra to gain further insight into the distributions of the two isotopes and thus the flow pattern within the CAA. The ^{228}Ra/^{226}Ra was computed as a function of salinity from the apparent end-members and compared to the relationship between the ^{228}Ra/^{226}Ra and δ^{18}O (Fig. 12a, b). The apparent ^{228}Ra/^{226}Ra over S_P mixing ratio appears as if the ratio was only affected by conservative mixing of the two respective end-members (Table 2). When relating this ideal behaviour to the ratios observed in our study, three main groups of samples can be identified. A: the higher salinity (S_P>32, δ^{18}O>~-3) samples, that more or less fall together with the mixing relationship. B: the samples characterized by substantially higher ^{228}Ra/^{226}Ra (~27<S_P<30, ~-5<δ^{18}O<-3), and C: the second group of low-salinity samples with ~S_P<31 (δ^{18}O<~-3), characterized by a substantially lower ^{228}Ra/^{226}Ra isotopic ratio. The spatial distribution (Fig. 12c) of these sample groups unravels processes that shape the Ra distributions, that, at the first view, did not seem to fit into the broader scheme described in Fig. 9. Samples with higher ^{228}Ra/~~:~~^{226}Ra ~~ratios~~ than the mixing ratios are located within the CAA, at stations 312 and 314, and at the downstream stations along the southern coast of the N~~n~~orthwest P~~p~~assage, which in turn are under the strong influence of shelf/sediment derived ^{228}Ra accumulation as they flow eastward. This water mass mixes on the southern side of Lancaster Sound with the water from Baffin Bay, yielding a flow pattern highlighted by the higher ^{228}Ra/^{226}Ra ~~ratios~~ in the CAA (Fig. 11c, 12c). The stations with lower Ra isotopic ratios than the mixing ratios are located on the northern part of the Lancaster Sound and connect to the Canada Basin via McClure Strait and Parry Channel (Figs. 1, 12c). The waters at stations with lower Ra isotopic ratios mix with (inflowing) water from Baffin Bay along the northern side of Lancaster Sound (Fig. 11c). The overall lower $^{228}Ra/^{226}Ra$ waters reflect the long-term isolation of CB4 waters from their margin source (e.g., Kipp et al., 2018) such that the ^{228}Ra activities are diminished noticeably by radioactive decay. Consistent with this finding is the clear separation between water of the northern and southern sides of Lancaster Sound, as discussed in Fig. 11. We integrate the observations and findings featured in Figs. 11-13 into a revised scheme, shown in Fig. 14, to reveal the main flow pattern.

This analysis can further be exploited to highlight the release of ^{228}Ra from shallow shelf sediments to waters in the lower salinity range rather than from rivers. Both the $^{228}Ra/^{226}Ra$- S_P and $\delta^{18}O$ reveal a non-conservative addition of ^{228}Ra to waters in the salinity range of $25<S_P<30$ (Fig. 12). Furthermore, when considering $\delta^{18}O$, conservative mixing from a riverine source can be excluded (see also Burt et al., 2016b; Kipp et al., 2018; Moore, 2000, 2007). On the other hand, the $\delta^{18}O$ values of -3 to- -4 ‰ imply that the ^{228}Ra source is under riverine influence, as the $\delta^{18}O$ signature of the sea-ice end-member is generally thought to be approximately -2‰ (e.g., Eicken et al., 2002; Thomas et al., 2011; Yamamoto-Kawai et al., 2009, and references therein, see also Thomas et al., 2011 Fig. 5d).

III.III.II.II: Principal Component Two: Particle-related impacts (nutrient-type behaviour)

The correlation of Ba, DIC and temperature with PC2 is based on the hydrographic peculiarities of the CAA, where temperature displays a "classical", inverse nutrient-type profile (Fig. 2), resulting from the presence of a temperature minimum in the UHL. As is the case for Ba and DIC, nutrient-type profiles are generally shaped by the interaction of biological (production/respiration, adsorption/desorption) processes and gravitational particle settling. Properties revealing such distributions are represented by PC2, with the temperature minimum coinciding with those minima found at the pycnocline~~s~~ depths.

IV. Conclusions

It is our hope that with a better understanding of the distributions of the long-lived Ra isotopes, coupled with other chemical constituents, future initiatives can be supported to investigate the changes in~~impacts influenced by the~~ water mass distribution ~~of~~in ~~climate change within~~ this region. Given the results of the PCA as well as the distribution ~~presence~~ of ^{228}Ra, our data ~~are consistent with~~ reveal the existence of a western flow of water along the n~~N~~ortheastern edge of the CAA. This flow is a component~~pattern coincides with~~ of a cyclonic~~the~~ U-turn of ~~cyclonic~~ Baffin Bay ~~originating~~ water, intruding westward into the CAA before being rerouted~~ing~~ back to the east. The bulk eastward transport of water through the CAA was confirmed, highlighted by the distribution of Ra radioisotopes and chemical constituents in apparent end-members throughout the region. Overall, the results from this study provide the foundation for future GEOTRACES studies and~~or~~ other initiatives that focus on the sensitivity of trace element fluxes to changing environmental conditions by identifying and quantifying anomalies in the distribution of radioactive isotopes in the Canadian Arctic Archipelago. Furthermore, this study provides an additional tool to better understand and ~~the vulnerability of ecozones, such as the Arctic to climate change by~~ characterize~~ing~~ water mass distributions, flow patterns, mixing and their respective time scales in ~~these~~ challenging sampling areas such as the Arctic.

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Figure Captions:

Figure 1: Map of the Canadian Arctic Archipelago (CAA) showing the 17 stations sampled during the 2015 GEOTRACES cruise aboard the CCGS Amundsen (red dots), where the two unlabeled stations along the Eastern CAA cross-~~-~~channel transect are the surface stations 323 and 324, numbers refer to CAA stations (1-7).~~;~~ Nares Strait (NS), Barrow Strait (BS), McClure Strait (McS), Lancaster Sound (LS) and Parry Channel (PC) are denoted, the latter ~~one~~ connecting the CAA from McS via BS to LS. Underway (UW) samples have been taken on the way from Baffin Bay (BB) into Lancaster Sound. Lastly, the blue and grey lines indicate the 200m and 100m isobaths, respectively.

Figure 2: Vertical distributions of practical salinity, temperature, density, AT, Ba, $\delta^{18}O$, DIC and normalized DIC (DIC_{norm}) ~~observed at~~ observed at stations CAA1 to CAA7~~7~~ ~~stations~~ throughout the CAA.

Figure 3: ^{226}Ra (a), ^{228}Ra (b) and $^{228}Ra/^{226}Ra$ (c) profiles ranging from surface to depth for stations CAA1 to ~~-~~CAA7 (where pink dots indicate station CAA2 which was only sampled at the surface and bottom) throughout the Canadian Arctic Archipelago.

Figure 4: Total Alkalinity (AT) versus salinity measured throughout the CAA with coloured depth (a) and longitude (b). A linear regression analysis yields AT = 52.7*S_P+492.25 (R^2=0.915).

Figure 5: Dissolved Inorganic Carbon (DIC) (a) and salinity-normalized Dissolved Inorganic Carbon (DIC_{norm}) (b) were plotted against salinity (S_p), with colours indicating depth (m) (a, b) and longitude (c). The DIC vs. SP regression yields DIC = 8.33*S_P+1835.0 (R^2= 0.116). ~~, t~~The DIC_{norm} vs. S_p plots were fitted with a piecewise regression analysis representing the surface, y=-19.7~~661~~x+2680.9

(R^2=0.533) and at depth; y=-34.2~~186~~x+3282.2 (R^2=0.765). In plots ~~(~~b & c~~)~~ CB4 was excluded from the piecewise regression (represented by unfilled, crossed out grey circles), whereas~~ile~~ stations 312 and 314 surface samples were excluded entirely. The black diamonds identify the average Atlantic deep-water samples from stations CAA1, CAA2 and CAA3.

Figure 6: ^{228}Ra (dpm/100L) plotted against practical salinity with colour indicating depth (a) and longitude (b) fitted with a piecewise regression excluding the deep stations of the Canada Basin (grey circle~~diamond~~s) and yielding f(x) = -2.47~~666~~x + 86.4~~377~~ (R^2 = 0.284~~35~~) for the surface trend (0-80m) and f(x) = -0.485~~4~~x + 21.1~~27~~ (R^2 = 0.072~~2~~) for the deep trend (>80m). The average of Atlantic deep waters sampled from stations CAA1, CAA2 and CAA3 is defined by a black diamond.

Figure 7: Eight-variable Principal Component Analysis (PCA) of PC1 and PC2 for 64 samples from 17 stations throughout the Canadian Arctic Archipelago, distinguished by depth groupings; Surface (0-20m; purple), Mid (20-80m; blue), Deep ~~Archipelago~~ (Deep Arch, 80-500m; red) Archipelago and Deep Atlantic (Deep ATL, >500m; green). The sample collected at station CB4 at 200m depth was excluded from this plot. The ellipses represent 95% confidence intervals associated with each water mass grouping.

Figure 8: Eight-variable Principal Component Analysis of surface samples (0-20m;) east (green, east of 85°W) and west (blue, west of 85°W) and mid-Depth (20-80m; red) samples collected throughout the Canadian Arctic Archipelago analyzing PC1 and PC2 for 27 samples from 17 stations, with the exception of the surface sample collected at station CB4 that~~which~~ was excluded from this plot. The ellipses represent 95% confidence intervals associated with each water mass grouping.

Figure 9: Temperature-Salinity plots with colours indicating depth (a) and longitude (b) as well as the Radium Isotopic Ratio(^{228}Ra/^{226}Ra)-DIC plots with colour indicating depth (c) and Salinity (d), ~~furthermore~~ highlighting three water masses throughout the CAA, the two surface water masses the Western Surface (Surf W) and Eastern Surface (Surf E) waters and one water mass (Atlantic) at depth **(Deep).** In (c) and (d) station CB4 has been denoted with a circle and cross.

Figure 10: Principal Component Analysis (PCA) of PC1 and PC2 for 64 samples from 17 stations throughout the Canadian Arctic Archipelago, composed of the seven normalized variables **Salinity** (S_P), Temperature (T), DIC, Ba, $\delta^{18}O$, ^{228}Ra, and ^{226}Ra, excluding AT. They are distinguished by depth groupings; Surface (0-20m; purple), Mid (20-80m; blue), Deep ~~Archipelago~~ (Deep Arch, 80-500m; red) Archipelago and Deep Atlantic (Deep ATL, >500m; green).

Figure 11: Cross section at stations CAA1-3, 323 and 324 for dissolved Ba (a) and barite saturation state (Q_i) (b), as well as $^{228}Ra/^{226}Ra$ (c) and $^{226}Ra/Ba$ (dpm µmol^{-1}) (d). The low values of both properties indicate the presence of Atlantic water (see Thomas et al., 2011).

Figure 12: Relationship between the $^{228}Ra/^{226}Ra$ as derived from the apparent end-members vs. salinity (a) and $\delta^{18}O$ (b). In insert in (a), the S_P vs. $\delta^{18}O$ relationship. Increasing fractions of sea-ice would cause a near-horizontal shift in that relationship (sea-ice $\delta^{18}O$ end-member = -2‰), whereas meteoric water would cause a “diagonal” shift (meteoric water $\delta^{18}O$ end-member = -20‰); see also Thomas et al., 2011 for more details. $^{228}Ra/^{226}Ra$ in surface samples across the CAA depict~~ing~~ the different flow pass via the CAA/northwest passage, and via McClure Strait, Parry Channel and Lancaster Sound (c). For reasons of clarity, only the surface samples have been shown in (c). The colour coding groups the ~~,~~ ~~black, white and gray stars group~~ samples into water masses with high salinity and low isotopic ratio (ATL), with low salinity and high isotopic ratio (shelf waters near the ^{228}Ra source), and lastly with low salinity and low isotopic ratio (waters in direction of CB4), respectively.

Figure 13: Relationships between ^{226}Ra and Ba (a), and between the $^{226}Ra/Ba$ and ^{226}Ra and Ba concentrations (b), respectively (c). The red symbols indicate samples with $S_P>34$ (Atlantic origin). In (b) the linear regressions yield for samples with $S_P>34$ f(x) = 0.20x + 0.25, R^2=0.99, and $S_P<34$ f(x) =

0.18x + 0.05, R^2=0.90. Within (a) and (b), open circles are drawn from van Beek et al., (2007), whereas ile the dashed line in (a) is redrawn from Le Roy et al., (2018). Panel (d) depicts the spatial distribution of the $^{226}Ra/Ba$ in surface waters across the CAA.

Figure 14: Sketch of proposed surface flow pattern as identified in the current study where dotted lines indicated reduced certainty of trends.

Appendices:

Appendix 1: Equations used to normalize (*X_0*) the data distribution for Temperature, Salinity, Dissolved Inorganic Carbon (DIC), $\delta^{18}O$, ^{226}Ra, ^{228}Ra and Ba collected throughout the CAA 2015 GEOTRACES cruise in preparation for Principal Component Analyses.

Appendix 2: DIC, AT, $\delta^{18}O$, and Ba plotted against depth for each station including CAA1, CAA2, CAA3, CAA4, CAA5. CAA6, CB4, 312, and 314, where profiles were taken throughout the 2015 GEOTRACES cruise in the Canadian Arctic Archipelago. Colours indicate the water masses present at the sampled depth; red is the Polar Mixed Layer (PML), yellow is the Upper Halocline Layer (UHL), and blue is the Atlantic Layer (ATL).

Appendix 3: ^{226}Ra, ^{228}Ra, and Ra isotopic ratio ($^{228}Ra/^{226}Ra$) plotted against depth for each station, including CAA1, CAA2, CAA3, CAA4, CAA5, CAA6, and CAA7, where profiles were taken throughout the 2015 GEOTRACES cruise in the Canadian Arctic Archipelago. Colours of depth indicate water masses at the sampled depths; red is the Polar Mixed Layer (PML), yellow is the Upper Halocline Layer (UHL), and blue is the Atlantic Layer (ATL).

Response to Interactive Comments from: "Using 226Ra and 228Ra isotopes to distinguish water mass distribution in the Canadian Arctic Archipelago" by Chantal Mears et al. Michiel Rutgers van der Loeff (Referee).

(Please note, the Referees comments are italicized and our responses are un-italicized.)

We would like to thank Michiel Rutgers van der Loeff for taking the time to comment in great detail on our paper.

The paper presents and discusses the results of a study of long-lived radium isotopes in the Canadian Archipelago as part of a larger GEOTRACES study conducted 2015- 2016 in the Arctic Ocean. The study is supported by parallel analyses of Ba and of the carbonate system (DIC, AT). The chemical and hydrographical data are subjected to a principal component analysis and the results are used to derive apparent water mass endmembers. The general features are well explained by 228Ra releases from (especially southern) shelf regions, a separation of water masses by the pycnocline (at about 100m depth), an eastward outflow of water from the Canada Basin over the sill in Barrow Strait (less than 200m deep), and an anticlockwise recirculation of water from the Baffin Bay through the Lancaster Sound.

Graphs are very clear but a few are in my view not necessary for the message. Unless a clearer reason is given for the need for the normalization of DIC, Figs. 5 b and c can be omitted. Figs. 13 b and c do not add to the information given in Fig. 13a

We thank the referee for this comment. Indeed, Figs 5b,c and 13b,c appear "underrepresented" in our discussion. Still, in our opinion these Fig 5b/c, i.e. DIC_{norm}, provides valuable understanding to the watermass distribution, as Fig 13b,c do by identifying species controlling the ^{226}Ra/Ba ratio. In any revised version we will aim to clarify and underpin the use to these panels. In particular the Figs 13b,c highlight the difference between the high-Ba CAA waters and the ones from the Atlantic, characterized by relatively lower Ba values. This discrimination appears to cause an offset in the ^{226}Ra/Ba relative to a given ^{226}Ra.

This paper adds an important chapter to the distribution and behavior of Radium isotopes and their use as water mass tracers in the Arctic Ocean. It deserves publication in Biogeosciences after a minor revision. It is generally well written, but there are several paragraphs that should be clarified.

Specific comments: 101: "westward" or eastward?

Westward is intended here, but the sentence has been rephrased to "This suggests that there may be the intrusion of Atlantic waters originating from Baffin Bay, moving into the CAA along the northern edge from the east, and possibly creating a "U-turn" as the westward current reroutes back into Baffin Bay along the southern edge."

122: "nearly conservative". You might mention that there is a certain uptake (with Ba) by primary production

We agree with the referee and thus have changed this sentence to include Ra's role with biological uptake and accumulation at depth to "A slight enrichment can be seen in Pacific Ocean deep waters relative to deep water of the Atlantic Ocean, primarily due to ^{226}Ra uptake by biology for the production of silicate or calcium tests or in barite ($BaSO_4$) co-precipitation (van Beek et al., 2007; Charette et al., 2015b; Moore and Dymond, 1991). With this exception in mind, ^{226}Ra reveals a "nearly" conservative distribution in the oceans, thus facilitating its use as a long-term pelagic-based tracer of water masses and of shelf inputs (Broecker et al., 1967; Charette et al., 2015a; Chung, 1980),

135: why is salinity called Sp? Why not just S as usual (and as used in eq 2)?

In our understanding S_P appears to be the new convention to refer to salinity.

218: I understand that 228Ra was also regressed against salinity to derive the endmember value at S=25.

We take note of the referees comment here, and thus have added "Lastly, linear regressions of each variable, with the exception of ^{228}Ra, against the practical salinity were plotted to express robust end-member relationships from within the previously categorized salinity-defined water masses present throughout the CAA. For ^{228}Ra, the end-member was derived from linear regression to the practical salinity where S_P=25, as it source is in shelf sediments, of which location is in the S_P range of 25."

233: "similar results". It is not clear whether these authors found similar east-west gradients or similar exceptions to that rule as the example of CAA3 and CAA5.

To clarify the referees comment, the line has been adapted to "Prinsenberg and Bennett (1987) reported similar trend in results from samples collected in 1982 across Barrow Strait, a sill less than 200m deep located roughly between 105°W to 90°W, where analogous transects for salinity and temperature were recorded throughout the surface layer (Fig. 1)."

251: I do not find CB4 results in Fig 2

This station was purposefully left out of the profile comparison in Fig 2, as it's values differ greatly than those found within the Archipelago. Profiles of CB4 are provided in the Appendix, which has now references in line 251.

246-256: this paragraph is too condensed to be understandable. I would like to see a bit clearer formulations.

We agree with the Referee, the paragraph has been edited in hope to clarify the results: "The property-property diagrams of DIC and AT vs. S display strong positive relationships in surface waters (see Fig. 4 and 5a for regression intercepts and error). The surface waters of the stations west of 96.5°W thus appear impacted by sea-ice melt (SIM) and Meteoric Water (MW) (Figs. 1, 2, 4, 5c). Highest DIC concentrations were observed at the pycnocline of the western most station (CB4) (Fig. 2, Fig. 5a and Appendix 2). This maximum in metabolic (respiratory) DIC decreases slightly eastward due to the increasing contribution of low-DIC ATL waters (Fig. 5a) (Shadwick et al., 2011a). AT concentrations were found to increase more linearly with depth, void of the metabolic maximum witnessed within the DIC samples (Figs. 2d, g, 4). This is explained by the concomitant increase in AT and S values rather than metabolic activity, thus distinguishing AT from DIC (Burt et al., 2016a; Shadwick et al., 2011a; Thomas et al., 2011)."

246-247: For the intercept values of AT and DIC versus salinity, refer to Figs. 4 and 5a. I understand that the intercept in DIC is used later for the normalization of DIC. That makes it important to show the regression lines and indicate the error of the intercept values.

We agree with the referee and intercept values were removed from the text and referred to within the figures. Regression lines and error will be added to Figure 4 and 5a.

248: "thus attributed" I don't understand what is meant here

As discussed above, we have rewritten this section.

253-255: "similar results"? With respect to longitude yes, but without the metabolic maximum

As discussed above, we have rewritten this section.

254: "both"?

As discussed above, we have rewritten this section.

255: do you mean that at the depth of the pycnocline both S and AT but not DIC are increasing eastward?

As discussed above, we have rewritten this section.

Lines 263-274; Normalization of DIC: I understand from Friis et al. (2003) that normalization is meant to remove the salinity dependence of DIC, which is apparently not the result here (compare Figs. 5 a,b). The rationale of this normalization is not well explained. The difference between surface and subsurface water masses is clearly seen in Fig. 5a (especially when in panel a the symbols belonging to anomalous station CB4 are marked as they are in panel b and c) and is not made clearer by the normalization procedure depicted in Figs 5b and c.

The normalization depicts here the clearly separated surface and deeper water masses. Both water bodies reveal individually conservative mixing gradients, but hardly, if any, mixing between them.

272: "consistent" is misplaced here. The inverse longitudinal trend is noted in the next sentence, but it is not explained why the accumulated DIC decreases "as waters flow longitudinally eastward through the CAA". When surface water flows eastward, DIC increases, S increases and DICnorm decreases. Is this the non-conservative behavior meant in line 266 to be shown by the normalization (evasion of CO2?) or just an effect of mixing with high-S/low-DICnorm water from Baffin Bay?

It is consistent in respect to the higher presence of meteoric waters in the western parts, which dilute the DIC of the PML on the one hand. The higher DICnorm reflects the high DIC concentration of the meteoric water, which is largely absent in the eastern parts. In the subsurface waters the observed decrease in DICnorm reflects the mixing of the Pacific waters (of western origin) with high respiratory DIC with the Atlantic waters (of eastern origin) which carry a much lower respiratory DIC signal.

363: why does the similarity of Stas CAA1, 3 and 5 to the Deep ATL group suggest a westward intrusion along the northern edge when Stas 1 and 5 are along the northern edge but Sta 3 is along the southern edge?

We take into account the referees comment here and will aim to better explain the "U-turn" hypothesized, "Results of this analysis reveal that the Deep Arch stations CAA1, CAA3 and CAA5 are more closely linked to the Deep ATL group, implying that they are in fact part of the Deep ATL water mass (Fig. 10). This suggests that there is an intrusion of ATL water along the northern edge of the CAA. This westward flow with a speed of 2.2 cm/s was observed by Prinsenberg and colleagues (2009) and is weaker than the dominant eastward current flow (15.3 cm/s). This mild inflow of water along the northern edge of the Archipelago is then suspected to be redirected and exits back to Baffin Bay through the southern Archipelago station (CAA3)."

380: Explain why you show in Fig. 11d the 226Ra/Ba ratio and not 226Ra itself. Why is the scale in Fig. 11c inverted? That is a bit confusing.

Because of its long have life ^{226}Ra on its own does not provide a strong signal, in particular not in the deeper waters (Fig. 3). As ^{228}Ra is much more responsive to decay on the timescales relevant for this study the ^{228}Ra/^{226}Ra ratio appears more appropriate to reveal systemic processes. We appreciate the note on the colour scale, this appears to be an error on our side.

Lines 386 and further; Figure 13: It is very useful to plot 226Ra vs Ba and to map the 226Ra/Ba ratio. But what is the advantage of plotting the ratio against each of its components 226Ra and Ba (Figs 13 b,c)? In 13a, what is the broken line? If this is the relationship found by Le Roy (and/or van Beek), then mention it. If not, then include it. What are the circles? They are missing in panel13c. The red symbols (S>34) in Fig. 13a appear to show that 226Ra is independent on Ba, in contrast to the findings of LeRoy and van Beek.

We apologize for this oversight: In Fig 13a,b the open circles are the data from van Beek et al., the dashed line in Fig 13a is redrawn from LeRoy et al. The regressions in Fig 13b are from our own data as specified in the caption. We will add this to the caption.

In line 391 you may mean that the average position of the red symbols (~10 dpm/100L; 44 nM) falls along that line, but their 226Ra variation is not associated with a corresponding variation in Ba.

Unfortunately, it is not fully clear to us, what the referee intends to state here? It was our intend to show that ^{226}Ra and Ba reveal a somewhat “noisy” relationship within the CAA (Fig 13a). However, if you normalized the ^{226}Ra to Ba, i.e., use the ^{226}Ra/Ba ratio, it becomes evident that this ratio ins controlled by the variability in ^{226}Ra and a “Ba-offset” originating from the CAA. This offset is not visible in the Atlantic data, neither in ours, nor in the ones plotted from literature.

396: Please give a reference for the high Ba runoff of rivers draining into the CAA

Yes we agree with the referee and this will be added to the paper (Gauy&Falkner 1998).

Lines 476-509; Figure 12: What is the meaning of the broken line in 12a? Is that the relationship based on apparent endmembers? Perhaps this is related to the expressions “higher” and “lower . . . than the mixing ratios” in lines 487-494. Please clarify. The symbols in Figs.12 a,b appear to have no other background than their position relative to each other on this graph. It is then more appropriate to use an enveloping ellipse as in Fig 9 rather than different symbols. It might be interesting to identify the outliers geographically (in Fig 12 c). Legend: flow paths? The figure is discussed again in lines 503-509, but that formulation must be improved.

We appreciate this comment. In essence, all information is given, but not well enough presented. The dashed line has been compiled from the data given in table 2, and the geographical attribution is shown by the shading of the symbols, but we have fallen short in spelling that out. We will improve the wording of that section. In order to improve clarity we also now have colour-coded the panels 12a and 12b to allow for a geographical attribution. An insert for the δ18O-S_P relationship has been embedded as requested by referee 2. We hope that the improved figure underlines our line of argument

Line 502: you mean: the 228Ra/226Ra -δ18O relationship (Fig. 12b). If you want to show nonconservative behavior from this relationship, shouldn't you draw an endmember mixing line as in panel a?

The issue with compiling the $\delta^{18}O$ mixing line is that the two freshwater endmembers (sea-ice and meteoric water) reveal different endmember characteristics. As discussed our analysis (Tab. 2) gives "only" an apparent, ie. mean endmember, which however is in reality a composite (see for example Thomas et al., 2011 and elsewhere). Instead, we intend to use that plot to unravel the characteristics of the two low-S_P groups of $^{228}Ra/^{226}Ra$ samples, one with less negative $\delta^{18}O$, pointing to sea-ice melt and long history and distance from the ^{228}Ra source, and one imprinted by more negative $\delta^{18}O$ (meteoric water) from within the CAA with young history and proximity to the ^{228}Ra source. As indicated in the preceding section we will attempt to improve the respective wording.

Line 503-505: I guess you mean that conservative mixing of 228Ra or 228Ra/226Ra with a pure riverine source can be excluded, but that will hold for a plot against salinity as well as for a plot against 18O.

As indicated in the preceding section we will attempt to improve the respective wording.

FIGURES: Fig. 7: make sure that the names of all the parameters are readable. "18O" is covered.

We acknowledge the referees comment here and have made the parameter titles within Fig 7 visible.

Fig 14: Is this surface flow? If not specify at what depth.

We agree with the referees comment and the figure caption for Figure 14 was changed accordingly to "**Figure 14:** Sketch of proposed surface flow pattern as identified in the current study."

Interactive comment on "Using 226Ra and 228Ra isotopes to distinguish water mass distribution in the Canadian Arctic Archipelago" by Chantal Mears et al. Michael E. Böttcher (Referee)

We would like to thank Michael E. Böttcher for taking the time to review our manuscript. Any comments provided by the Referee will be placed in italics while answers will be un-italicized.

The submitted manuscript on "Using 226Ra and 228Ra isotopes to distinguish water mass distribution in the Canadian Arctic Archipelago" by Mears, Thomas, Henderson, Charette, MacIntyre, Dehairs, Monnin, and Mucci, presents a detailed data set for a hydrographic and chemical characterization of water masses in the Canadian Arctic Archipelago that was derived during the GEOTRACE program: Oxygen isotopes (18O/16O) of water, radium isotopes, parameters of the dissolved carbonate system (AT, DIC), and dissolved Ba. The data are used to separate water masses and mixing properties, and define Ra sources. A thermodynamic analysis is used to defined the saturation states of the aqueous solution wrt. barite, and all data are furthermore analyzed by a principle component analysis (PCA).

I will not repeat the arguments already provided in the detailed review by Michiel Rutgers van de Loeff, which I agree upon, and only concentrate on further minor aspects.

Overall, the data data set is original, impressive, and the conclusions derived from the investigation are well argued, including the presenting figures.

Detailed comments:

- I suggest to add a covariation diagram and further discussion of d18O-H2O values versus salinity.

This has been shown and discussed in Thomas et al., 2011 their Fig. 5d. Please see also our comment in response to referee 1's comment re. line 502. We have added a respective panel as insert to Fig. 12c.

- Fig.2: Its: d18O.

We agree with the Referee, this was an over look on our part and has been changed within the figure.

- Table 2: Units are missing.

We agree with the Referee, this was an oversight on our part and has been changed within the table.

Reference list: L624: What kind of publication is this?

-e.g., lines 618, 631, 664, 710, 728 etc.: Please delete all informations about 'access dates' and if an article was read on-line from the reference list. Serious scientific journals guarantee that the scientific content published after acceptance is the same off and on-line, and keeps its content in all details over time. This may be different for other on-line sources, that are often questionable permanent references.

Thank you Referee, the proper citation has been placed for L624 and the access dates have been deleted from the reference list.

Michael Ernst Böttcher, April 17th, 2020

Interactive comment on "Using 226Ra and 228Ra isotopes to distinguish water mass distribution in the Canadian Arctic Archipelago" by Chantal Mears et al. Amber Annett (Referee) a.l.annett@soton.ac.uk Received and published: 23 April 2020

Thank to Amber Annett for taking the time to review our manuscript. The Referees comments are italicized while our answers are unitalicized.

Mears and colleagues present an extensive dataset from the Arctic, spanning the Canada Basin to Baffin Bay via the Canadian Arctic Archeipelago, with the aim of characterizing water mass end-members, evolution of properties and transport. The study employs a range of parameters, including DIC, Ba, oxygen and Ra isotopes to probe the influence of freshwater and shelf interaction, detecting westward flow along the northern flank of Lancaster Sound.

Overall, the context and aims are well established. The introduction provides necessary background to the regional oceanography and methodological approaches. Methods are appropriate, adequately detailed and the results clearly support the conclusions drawn and are well situated within the existing literature. I agree with the comments already provided by reviewers Rutgers van der Loeff and Böttcher, including that the manuscript represents an addition worthy of publication in Biogeosciences after minor revisions. My additional comments are that the presentation of some results could be improved to make the interpretations more *immediately visible from the figures (specified below), and that providing some additional quantitative details in the implications section could enhance the impact of this manuscript.*

Line 101: The final sentence of this paragraph is not needed, and references to what will be discussed or unravelled later in the manuscript should be minimized where possible.

Upon review we agree with the Referee and the last sentence of the paragraph "The importance of this "U-turn" will be discussed later in the results section", has been removed.

Line 373: Not substantially lower, unless you define the depths – Fig 11c indicates higher 228/226Ra ratios at 100m and 400m, with lower ratios only present at 0, 200 and 700m.

We agree with the Referee here, providing a depth is both necessary and adds clarity, the statement has then been changed to " Furthermore, substantially lower values of the $^{228}Ra/^{226}Ra$ ratio at depth (Fig. 11c) indicate the inflow of Atlantic water on the northern side of Lancaster Sound as well as its outflow along its southern side".

Line 380: the opposing gradients, do you mean the different strength of the gradient?

We agree with the referees comment, this makes the discuss difficult to follow and has thus be changed to "Since ^{226}Ra activities reveal a much larger north-to-south gradient across Lancaster Sound than Ba does (Figs. 2e, 3a), the discrepancy in strength of the gradients shown in Figs. 11c and 11d are dominated by changes in ^{226}Ra."

Line 374 says "the same pattern"

We agree with the referee here and have changed the wording to better complement the statement to "The corresponding pattern is revealed with $^{226}Ra/Ba$"

Line 467: clarify the impact of longer circulation history (it could be interpreted as 228Ra decay, or accumulation of 228Ra from shelf inputs)

In essence it should be both. Data from van der Loeff (2003) are from the Eurasian Shelf thus close to a ^{228}Ra source. The Atlantic waters in the CAA have been without shelf contact for longer times allowing for tangible decay of ^{228}Ra. We will amend wording here.

Line 296: Mention of CB4 could be integrated more clearly into this paragraph explicitly linking it to the southern flank of LS. Further, what level of confidence do you have in where the inflow turns back around or ceases to influence water properties? The westward flow is not shown reaching CAA6 in Fig 14, but the north/south gradient persists between CAA6 and CAA7 in Figs 12c and 13c - if there is uncertainty in how far this water reaches you could include a dashed arrow?

Thank you for this hint. This pattern should be attributed to the intrusion of the Northwest Passage waters, which we have drawn solely at one point. We will amend the figure accordingly.

Line 525: "rerouted" rather than rerouting

We agree with the Referee, and this has been changed within the manuscript.

Line 531: This section feels a bit unsupported; some quantification effort would deliver meaningful context for using these findings as a tool to probe impacts or vulnerability to climate change, and increase the impact of the manuscript. Some suggestions: based on 228Ra decay, what is the minimum time scale of the eastward transport of water between CAA1 and CAA6? This must make some assumptions (e.g. no additional inputs) but would provide a minimum time scale; is it rapid or slow? What are the temperature differences between east and westward flowing waters? Where will increased heat be delivered – pumped into LS or out into Baffin Bay? Does any historical data support a strengthening or weakening of this u-turn route and what does/would that mean for transport of heat (or nutrients, or any other parameter).

Thank you, we will amend this sentence to have a stronger relation to our paper.

Fig 2: If full CTD cast data is available, it would be preferable to show this rather than only the points for which DIC/Ra/Ba samples were collected, in order to situate samples relative to pycnoclines, thermal minima, etc.

We will inquire at the GEOTRACES data center about this proposition.

Fig 4: Pink/red colours are difficult or impossible to distinguish for a few of the points. Stick to a different palette for clarity?

We agree with the referee's an attempt will be made to change the colour bar to be discernable.

Fig 6 legend: Should this read Grey circles? If there are diamonds, they're not visible. Repeat here what the division is between "surface" and "deep" waters.

This is correct, it should have read grey circles and the characterization of surface or deep water masses has been repeated.

Fig 8: refer to section of text where the east/west boundary is stated (can it be shown on a map as well?)

We agree with the Referee, that it is added information and clarity to define what surface east and west are, and thus the definition has been added to the figure title of Figure 8. An additional map has not been made as there is a 10° longitudinal gap between stations west and east and therefore with reference to Figure 1, it is easily discernable.

Fig 9: Same comment regarding colour bar as Fig 4. Could the CB4 sample be designated with a different symbol so that it can be picked out easily on c and d?

We agree with the referee, an attempt will be made to highlight CB4.

Fig 11 & 12: Colour bar for 228/226Ra should also go from blue (low) to red (high) - otherwise the rationale for the inversion must be presented.

As responded to referee 1, this appears to be an error on our side. Thank you.

Fig 12: This figure panel needs overlap with the black/grey/white symbols of 12a and 12b, it's currently not possible to see where each group was collected from on the map. Also there's plenty of space on the map, please label 312 and 314 as readers may not remember all the station designations.

Thank you, we will colour-code symbols in Fig 12a,b. Addressing hereby the related concern of referee 1 as well.

Fig 13: Include legend on the figure showing what red symbols denote
This has already been indicated in the caption, but we will add information to the panel.

Interactive comment on "Using 226Ra and 228Ra isotopes to distinguish water mass distribution in the Canadian Arctic Archipelago" by Chantal Mears et al. Manmohan Sarin (Editor) sarin@prl.res.in Received and published: 14 May 2020

We would like to extend thanks to the editor, Manmohan Sarin for taking the time to provide comments. Below italicized are the editors comments, while un-italicized are our response.

This is a potentially interesting study on use of Radium isotopes to distinguish water mass distribution in the Canadian Arctic Archipelago. Further to the comments made by three reviewers, authors' may like to provide some quantitative information (in the abstract) on the concentrations of radium isotopes and 228-Ra/226-Ra ratios measured in different water masses. There is no single result/number provided in the abstract.

Thank you, we will provide such information in the revised abstract, where appropriate.

Lines 48-51, in abstract, can be moved to conclusion/implication section.

This line is restated in the conclusion section. But draws importance to the relevance of the study and other studies comparable to ours.

Authors' may like to reassess the regression parameters (slope, intercept and R2) stated in figure captions for 5 and 6. Are these numbers significant and meaningful to 3rd and 4th decimal units? For example, in Figure 6, regression analysis is given as: y = -2.4666x + 86.377 (R2 = 0.2835) for the surface trend and y = -0.4854x +21.127 (R2 = 0.0722) for the deep trend. The slope, intercept and R2 values for linear regression analysis are rather absurd (3rd/4th decimal) considering the analytical uncertainties in the measurements of radium isotopes in individual water samples.

We agree with the Editor, these numbers digits are exaggerated and will be shorted for conciseness.

Line 109: For the benefit of a general reader, it may be relevant to name the parent isotopes of Radium, 230-Th (226-Ra) and 232-Th (228-Ra).

We agree with the editor, it could be helpful for the reader to have the Thorium parent isotope and thus the sentence has added this information in, "Additionally, both long-lived Ra isotopes are formed from the decay of different Thorium (Th) isotopes (^{226}Ra is the daughter of ^{230Th}, while ^{228}Ra is the daughter of ^{232}Th) in sediments and are distributed to the ocean through porewater advection and diffusion across the sediment-water interface, primarily along coastlines or the bottom boundary layer (Charette et al., 2015)."

Line 161: Was there any attempt to simultaneously use 295 keV peak to quantify 226- Ra?

We have not attempted to use the 295KeV peak to quantify ^{226}Ra, as the uncertainty of the ^{226}Ra data would have been improved only incrementally, deemed insignificant for the purpose of the study.