1	Particulate trace element distributions along the Canadian Arctic CEOTRACES section: shelf water interactions, advective transport and contrasting
2	biological production
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11	Key Points:
12	• Advective transport shapes the distributions of lithogenic-derived particulate trace elements such
13	as pAl, pV, pFe in the deep Canadian Arctic basins.
14	• Manganese oxides, likely related to bacterially-mediated authigenic precipitation, dominate bulk
15	distributions in the Canada Basin (CB) and Baffin Bay (BB).
16	• A strong productivity gradient in the Canadian Arctic Ocean (CB << BB = Labrador Sea)
17	controls particulate phosphorus, organic carbon and nitrogen cycling.
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26 Abstract

27 Marine particles are important regulators of the biogeochemistry of many trace elements and isotopes 28 in the ocean, and as such, there has been an increasing motivation to unravel the processes which control 29 their cycling. Here, we present vertical distributions of total particulate trace elements (Al, V, Fe, Mn and 30 P) and particulate organic carbon and particulate nitrogen (POC and pN) collected during the GEOTRACES 31 Canadian cruise in 2015 in the Canada Basin (CB), the Canadian Arctic Archipelago (CAA), Baffin Bay 32 (BB) and the Labrador Sea (LS), where particulate trace metal data are scarce. While particulate trace 33 elements are generally affected by one-dimensional dynamics in ocean waters (e.g. deposition, 34 scavenging/remineralization, and sinking), lateral transport of lithogenic-derived particles (pAl, pV and pFe) plays a dominant role in shaping their distributions in the deep CB, BB and LS basins. Higher 35 36 concentrations of the aforementioned particulate trace elements are measured along the flow path of 37 boundary currents and in near-bottom waters. Unlike pAl, pV and pFe, primarily controlled by lithogenic 38 sources, bulk pMn distributions in our study regions (with the exception of LS) are dominated by authigenic Mn^{+3/4} oxides, with distinctively high concentrations in CB and BB subsurface halocline waters, as well as 39 40 in the deepest samples in BB. Enhanced bacterially-mediated Mn⁺² oxide formation is anticipated to occur 41 in these halocline waters which have the potential to sustain large populations of Mn oxidizing bacteria as 42 result of the close sediment-water interactions and distinct environmental conditions of these water masses. 43 Overall, the highest concentrations of pP, POC and pN occurred in surface and near-surface waters (> 100 44 m) at the same density as a chlorophyll-a peak and transmissivity drop, with a clear west-east increasing 45 concentration gradient from the CB to LS. In summary, biogeochemical cycles of particulate elements in the Canadian Arctic Ocean are controlled by enhanced lateral transport and sediment resuspension (pAl, 46 47 pFe and pV), authigenic formation of Mn and biological production (pP, POC and pN).

Keywords: Advective transport, Lithogenic sources, Particle dynamics, Sediment resuspension, Trace metal
biogeochemistry, Canadian Arctic Ocean, Redfield stoichiometry, GEOTRACES.

50 **1 Introduction**

51 Marine particles –operationally defined as material collected onto 0.2-0.4 µm pore size filters– are 52 important trace metal reservoirs, largely exceeding the dissolved pool in shelf regions, which strongly 53 modulate oceanic biogeochemistry by controlling the residence time of most elements (Lam and Bishop, 54 2008; Jeandel et al., 2015; Ohnemus and Lam, 2015; Homoky et al., 2016; Jeandel, 2016). Particulate 55 material is a heterogeneous phase, with diverse sources and complex composition. Lithogenic particles, 56 derived from weathering and erosion of the continental crust, are primarily delivered to ocean waters by 57 means of atmospheric deposition (Mahowald et al., 2018), river discharge (Aguilar-Islas et al., 2013; 58 Jeandel and Oelkers, 2015; Charette et al., 2016; Jeandel, 2016; Colombo et al., 2019a) and sea ice melt 59 (Measures, 1999; Tovar-Sánchez et al., 2010; Aguilar-Islas et al., 2013; Giesbrecht et al., 2013). Mid-depth 60 lateral transport of continental margin sediments and near-bottom sediment resuspension events are also 61 important sources of lithogenic particulate trace elements (e.g. Al, Ti, V, Fe) to the water column (Lam and 62 Bishop, 2008; Jeandel et al., 2015; Ohnemus and Lam, 2015; Charette et al., 2016; Jeandel, 2016; Gourain 63 et al., 2019; Morton et al., 2019). In addition to lithogenic sources, vast quantities of biogenic marine 64 particles are produced in surface waters through the assimilation of dissolved carbon, macronutrients (e.g. silicic acid, nitrate, phosphate) and micronutrients (e.g. Mn, Fe, Co, Ni, Cu, Zn, Cd) by phytoplankton 65 (Yigiterhan et al., 2011; Jeandel et al., 2015; Lam et al., 2015a; Twining et al., 2015). Moreover, authigenic 66 67 particles such as Fe and Mn oxides, are generated at the ocean floor above reducing sediment conditions 68 and in the water column where dissolved species of redox-sensitive elements are oxidized, producing 69 particles enriched in these elements over their crustal abundances (Lam and Bishop, 2008; Yigiterhan et 70 al., 2011; Hansel, 2017; Oldham et al., 2017; Morton et al., 2019; Vieira et al., 2019).

Given the relevance of particles in the cycling of trace elements and isotopes (TEIs), the fact that many particulate TEIs are useful tracers of biogeochemical and physical ocean processes, and, as TEIs dissolve and/or remineralize, they release important micronutrients for phytoplankton (Lam and Bishop, 2008; Jeandel et al., 2011; Anderson et al., 2014; Jeandel and Oelkers, 2015; Cheize et al., 2019), the study of 75 particulate trace element distributions is one of the main objectives of the international GEOTRACES 76 program. In recent years, large oceanographic campaigns, framed within the international GEOTRACES 77 program, have shed light on the biogeochemistry of particulate trace elements on the North Atlantic Zonal 78 Transect (Lam et al., 2015; Ohnemus & Lam, 2015; GA03 GEOTRACES cruise), the Eastern Pacific Zonal 79 Transect (Ohnemus et al., 2017; Lam et al., 2018; Lee et al., 2018; GP16 GEOTRACES cruise), and the 80 North Subarctic Atlantic from the Iberian margin to the Labrador Sea (Gourain et al., 2019; GEOTRACES 81 GA01 cruise). Nonetheless, our knowledge of particulate trace metal distributions in other regions, such as 82 the Arctic Ocean, is still very limited. A handful of studies have reported concentrations of total dissolvable 83 trace metals in the Chukchi Sea and adjacent shelf break areas of the Canada Basin, however, these studies 84 focused on the leachable fraction of the particulate pool (Thuróczy et al., 2011; Cid et al., 2012; Giesbrecht 85 et al., 2013; Kondo et al., 2016; Vieira et al., 2019). To the best of our knowledge, three studies have 86 described the distributions of particulate elements in the Chukchi Sea and Western Arctic Ocean (GN01 87 GEOTRACES cruise; Aguilar-Islas et al., 2013; Whitmore et al., 2019; Xiang and Lam, 2020), and three 88 other studies have reported particulate organic carbon distributions in the Canada Basin (Trimble and 89 Baskaran, 2005; Jackson et al., 2010; Brown et al., 2014).

This study contributes to the growing body of literature by presenting spatial and vertical distributions of total particulate trace elements (pAl, pV, pFe, pMn and pP) and particulate organic carbon and particulate nitrogen (POC and pN) across the deep Canada Basin, Baffin Bay and Labrador Sea, as well as pP, POC and pN distributions in the shallow Canadian Arctic Archipelago. With the aid of ancillary data, modeled tidal stresses and particle tracking simulations, we unravel the sources and processes shaping particulate distributions in the unique Canadian Arctic Ocean, a land-influenced environment characterized by extensive continental margins and shelves.

97 2 Study Area and Hydrography

98 The Canada Basin is strongly salinity stratified, with a fresh polar mixed layer (PML), seasonally 99 modified by freeze-thaw cycles of sea ice and snow, and a multilayered halocline (~30-400 m) which 100 insulates the PML from the underlying saltier and warmer Atlantic Layer (AL). The halocline assembly 101 consists of the Alaskan Coastal Water (ACW), with a potential temperature (θ) of about ~0 °C and a salinity 102 (S) range of 30 < S < 32, a relatively saltier (32 < S < 33) summer Bering Sea Water (sBSW) occupying the 103 bulk of the central Chukchi Sea, and the winter Bering Sea Water (wBSW), distinguished by a weak 104 temperature minimum near S = 33.1, which is advected from Bering Strait and contributes to the middle 105 halocline layer (Steele et al., 2004; McLaughlin et al., 2005; Timmermans et al., 2017; Figure 1 and Figure 106 S1a in the supporting information). Unlike the upper and middle halocline, the lower halocline (LH) 107 consists mostly of Atlantic-origin waters, and is identified by a sharp increase in temperature at salinities 108 between 33.3 and 34.6 (McLaughlin et al., 2005; Shimada et al., 2005; Woodgate & Aagaard, 2005). The 109 AL (~400-1200 m) has two components: the Fram Strait Branch (FSB), distinguished by a temperature 110 maximum and the Barents Sea Branch (BSB), which is deeper and colder (Smethie et al., 2000; McLaughlin 111 et al., 2004; Aksenov et al., 2011). Underlying the AL lies the old, cold and more saline Canada Basin Deep Water (CBDW > 1200 m; Timmermans et al., 2003; Figures 1 and S1a). 112

113 The Canadian Arctic Archipelago (CAA) is a complex network of islands and shallow straits, 114 connecting the Arctic Ocean to Baffin Bay. This shelf dominated region is an important export conduit for 115 fresh and nutrient rich Pacific waters (phosphate, silicic acid and bio-active trace metals) to the North 116 Atlantic, enhancing the productivity downstream (Michel et al., 2006; Beszczynska-Möller et al., 2011; 117 Wang et al., 2012; Hill et al., 2013; Colombo et al., 2021). The CAA links the Arctic Ocean with Baffin 118 Bay by three main pathways: 1- Parry Channel (sill depth: ~120 m), running from M'Clure Strait to 119 Lancaster Sound, where Arctic Water (AW) is exported to Baffin Bay, 2- Nares Strait (sill depth: ~220 m), 120 and 3- Jones Sound (sill depth: ~125 m; Figure 1).



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122 123 Figure 1. Sampled stations for particulate trace elements, organic carbon and nitrogen during the Canadian Arctic GEOTRACES cruises (GN02 and GN03), alongside bathymetry and a schematic of water circulation in the Canada Basin (after 124 125 Aksenov et al., 2011; Kondo et al., 2016) and Baffin Bay and the Labrador Sea (after Yashayaev and Clarke, 2008; Curry et al., 2011; Lozier et al., 2017). ACW: Alaskan Coastal Water, wBSW: winter Bering Sea Water, FSB: Fram Strait Branch, BSB: Barents 126 127 Sea Branch, AW: Arctic Water, EGC: East Greenland Current, WGC: West Greenland Current, WGIW: West Greenland Intermediate Water, BC: Baffin Current, LSW: Labrador Sea Water, NSDW: Northeast Atlantic Deep Water, DSOW: Denmark 128 Strait Overflow Water. Canada Basin stations: CB2-CB4. Canadian Arctic Archipelago stations: CB1 and CAA1-CAA9 (only 129 particulate phosphorus, organic carbon and nitrogen data from the CAA are presented in this study), Baffin Bay stations: BB1-130 BB3, Labrador Sea stations: LS2 and K1. Landmarks and straits of the Canadian Arctic Archipelago are displayed in the inset; MS: 131 M'Clure Strait, VS: Viscount Melville Sound, BS: Barrow Strait, LS: Lancaster Sound NS: Nares Strait, JS: Jones Sound. Parry 132 Channel is the main pathway in central CAA connecting M'Clure Strait with Lancaster Sound.

133 Baffin Bay is connected to the Labrador Sea and the North Atlantic Ocean through Davis Strait

134 (~650 m). The overall circulation in this bay is cyclonic, with a northward flow on the eastern side of Davis

135 Strait, the West Greenland Current (WGC), which consists of subsurface freshwater of Arctic origin (East 136 Greenland Current), and the warm and salty West Greenland Intermediate Water (WGIW: ~300-800 m; 137 $\theta > 1.3$ °C and S > 34.2) of North Atlantic origin (Cuny et al., 2005; Curry et al., 2011; Lozier et al., 2017). 138 This northward inflow is modified during its cyclonic circulation, and the upper layers of the WGC are 139 mixed with Arctic Waters, resulting in the fresh and cold Arctic Water (AW; $\theta < 0$ °C and 32.0 < S < 33.7), 140 which continues its flow to the Labrador Sea as the Baffin Current (Tang et al., 2004; Cuny et al., 2005). 141 Underlying the WGIW, lies the Baffin Bay Deep Water (BBDW), which is characterized by a small change 142 in salinity and a steady decrease in potential temperature (Tang et al., 2004; Figures 1 and S1c).

143 The subsurface circulation in the Labrador Sea is cyclonic, consisting of two components: the WGC 144 and the Labrador Current. The Labrador Current flows southward along the Labrador Shelf and Slope and 145 is the extension of the Baffin Current (BC; Cuny et al., 2002; Fischer et al., 2004). The Labrador Sea Water 146 (LSW) lies between the subsurface water and the deep western boundary current. Two LSW classes were 147 distinguished from the CTD data in 2015: the newly ventilated and fresher LSW formed during the winters 148 of 2014 and 2015 (LSW-2015), and the oldest, saltiest and least oxygenated LSW produced in the winters 149 of 1987-1994 (LSW-87/94; Yashayaev and Loder, 2016). Underlying the LSW-87/94, lies the saline, warm $(\theta \sim 3 \circ C, S \sim 34.92)$ and less oxygenated Northeast Atlantic Deep Water (NSDW), and the deepest, less 150 saline, colder ($\theta \le 2.6$ °C, S ~ 34.9) and more-recently oxygenated Denmark Strait Overflow Water 151 152 (DSOW; Yashayaev et al., 2007; Yashayaev and Loder, 2016; Figures 1 and S1d).

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155 **3 Methods**

156 **3.1 Sample collection**

157 Samples were collected on the CCGS Amundsen as part of GEOTRACES sections GN02 and GN03 158 (10/07/2015-1/10/2015) which covered an area from 56°N to 77°N and 53°W to 150°W in the Canadian 159 Arctic. The sampling was carried out during summer and early fall months, and hence, most stations were 160 ice-free when seawater samples were collected (Colombo et al., 2020). Vertical profiles ranging from 161 approximately 10 to 3500 m depth were obtained at 17 stations: three in the Canada Basin, nine in the CAA 162 (only pP, POC and pN distributions from the CAA are discussed in this paper), three in Baffin Bay and two 163 in the Labrador Sea (Figure 1). A trace-metal clean sampling system consisting of a powder-coated 164 aluminum frame, which held twelve 12 L Teflon-coated GO-FLO bottles (General Oceanics, Miami FL 165 USA) and a SeaBird 911 CTD/SBE 43 oxygen sensor instrument package (Seabird Electronics Inc, 166 Bellevue WA USA), attached by a 4000 m 4-member conducting Vectran cable encased in polyurethane (Cortland Cable Co., Cortland NY USA) was used to collect seawater samples for particulate trace element 167 168 analysis.

169 On-board the ship, these samples were transferred to a trace metal clean sampling van (HEPA filtered 170 environment), where ten liters of unfiltered seawater were collected into pre-cleaned 10 L LDPE cubitainers 171 (Bel Art and Nalgene) by the use of a piece of C-flex tubing (Masterflex) together with a Teflon straw. 172 Seawater was then filtered through a 0.45 µm Supor filter (47 mm diameter) inside a HEPA-filtered clean 173 air bubble. The filtration system included a cubitainer, a spigot, C-flex tubing, a peristaltic pump (Cole-174 Palmer), a 47 mm filter holder (Millipore) with customized screws, waste tubing and a waste container for 175 volume recording. After filtration, filters were dried inside a laminar flow hood, folded in half, and stored 176 in clean poly bags until analysis. Supor filters were always handled using pre-cleaned forceps and clean 177 gloves. The sampling devices, containers and the filtration system were cleaned according to GEOTRACES 178 protocols (Cutter et al., 2010).

Particulate organic carbon and nitrogen (POC and pN) samples were collected using the *CCGS Amundsen* rosette, consisting of 24×12 L Niskin bottles, a Sea-Bird SBE 911 CTD, and fluorometer, transmissometer, colored dissolved organic matter and oxygen sensors. POC and pN samples were collected at the same stations (excepting station CAA9) and depths where particulate trace elements were sampled. On-board the ship, between two to nine liters of seawater were filtered through 0.7 μ m / 25 mm precombusted (450 °C for 4 h) glass fiber (GF) filters (Whatman), which were later stored at -20 °C (Brown et al., 2014).

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3.2 Sample processing and analysis

In order to prevent contamination, the processing and analysis of particulate trace elements was conducted at the University of British Columbia (UBC) in class 1000 laboratories, pressurized with HEPA filtered air, and under class 100 laminar flow fume hoods. All the plasticware used during the sample preparation and analysis were cleaned according to GEOTRACES protocols (Cutter et al., 2010).

191 3.2.1 Particulate trace elements

192 Filters containing the particulate fraction were digested at UBC following the protocol described by 193 Ohnemus et al. (2014). In brief, the organic fraction and the Supor filter matrix were dissolved by reflux 194 heating with sulfuric acid and hydrogen peroxide (H_2SO_4/H_2O_2) , followed by mineral matrix dissolution by 195 reflux heating using a mixture of mineral acids (HNO₃, hydrochloric acid [HCl] and hydrofluoric acid 196 [HF]). After the digestion procedure, the clear residues were resuspended in 1% HNO₃ with 10 ppb of 197 indium as an internal standard; particulate samples were diluted prior to ICP-MS analysis (the particulate 198 digestion protocol is fully described in the supporting information: Supplementary methods). All reagents 199 used in the digestion and subsequent sample preparation (H_2SO_4 , HNO_3 , HCl, HF and H_2O_2) were Optima 200 grade (Fisher Scientific, Ontario, Canada).

Particulate Al, V, Fe, Mn and P were analyzed from a twelve-point calibration curve prepared in 1%
 trace metal grade HNO₃ from 1 ppm certified single element standards. The analyses were conducted by a

203 high resolution Thermo Finnigan Element2 ICP-MS in the Pacific Centre for Isotopic and Geochemical 204 Research (PCIGR), an analytical center at UBC. A medium mass resolution was selected for Fe, V and Mn 205 in order to remove isobaric interferences, and Al and P were analyzed using low mass resolution. During 206 sample analysis, solution blanks (1% HNO₃ Milli-q water with indium) and filter blanks were run to ensure 207 quality throughout the measurements; particulate trace element concentrations reported here were corrected 208 for the analytical blank by subtracting the average solution blank on the corresponding analytical day and 209 from the filter blank measurements. The accuracy and precision of this method was tested by analyzing the 210 certified reference material BCR-414, and GEOTRACES inter-calibration samples collected in the Pacific 211 Ocean. The BCR-414, as well as GEOTRACES inter-calibration samples underwent the same digestion 212 and analytical method used to analyze the Canadian Arctic GEOTRACES samples. Results from these 213 analyses (mean, standard deviation and relative standard deviation values) as well as, solution and filter 214 blank concentrations are listed in Table S1. Measured values in this study are in good agreement with 215 consensus values, yielding recoveries ranging from 76 to 123% (104±11%) for Al, V, Fe, Mn and P (Table 216 S1).

217 *3.2.2 Particulate organic carbon and particulate nitrogen (POC and pN)*

Frozen filters containing POC and pN were oven dried at 60 °C for 24 h at UBC, and then acid fumed with concentrated HCl for three days and dried at 50 °C for 24 h to remove particulate inorganic carbon (Brown et al., 2014). POC and pN analyses were conducted by the use of a carbon/nitrogen/sulfur vario MICRO cube analyzer (Elementar), and the concentrations reported here were corrected by subtracting the average concentrations of pre-combusted blank GF filters. The detection limits of the analyses -3σ of replicate measurement of pre-combusted blank GF filters– were 0.012 mg for POC (n= 10) and 0.009 mg for pN (n= 8).

3.3 Tidal stress estimates, particle tracking simulations and statistical analysis

Tidal flow over topography can generate strong sediment resuspension events. In order to identify the spatial distribution and the prevalence of these events in Baffin Bay and the Labrador Sea, we mapped tidal stress, calculated by squaring the barotropic tidal speeds. The magnitude of the tidal stress is proportional to the integrated effect of resuspension on an area, i.e. high tidal stress suggests frequent resuspension events (Wang, 2002). The barotropic tidal speeds were extracted from the MOG2D-G hydrodynamic gravity waves model (Carrère and Lyard, 2003) by Epstein (2018).

The paths of the winter Bering Sea water (wBSW) sampled at the three Canada Basin stations were traced from August 2015 backward two years using Ocean Parcels (Lange and Sebille, 2017) with five-day averaged velocity fields from a 1/12 degree coupled ocean-ice model of the Arctic and Northern Hemispheric Atlantic (Hu et al., 2018) configuration of the Nucleus for European Modeling of the Ocean (NEMO; Madec et al., 2017). In order to trace the wBSW, water parcels were released at model depth levels 237 23-30 (i.e. 110-380 m).

Python 3.6.0. programming language and NumPy, Matplotlib, SciPy and pandas libraries were used for statistical analysis and graphic design. Particulate trace element data, as well as POC and pN, are presented as mean ± standard deviation (otherwise noted), and differences between concentration means have been tested using student t-test at a significance level of 5% or lower. A linear least-squares regression fit (slope, intercept, correlation coefficient and p-value) has been applied to evaluate the relationship among different trace elements.

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245 **4 Results**

246 **4.1 Dataset overview**

This study presents total pAl, pV, pFe, pMn, pP, POC and pN distribution in the Canadian Arctic Ocean (CAO), across three deep basins: The Canada Basin, Baffin Bay and the Labrador Sea, as well as pP, POC and pN distributions in the shallow Canadian Arctic Archipelago. These are the first observations collected in the CAO for many of these elements (full dataset presented in this manuscript is included inthe Research Data Electronic Annex).

Among the deep CAO basins, the Canada Basin (CB) and the Labrador Sea (LS) had low concentrations of lithogenic-derived particulate elements, such as pAl, pFe and pV (albeit pV surface and subsurface distributions are not associated with lithogenic phases; discussed below), while the concentrations of these elements were higher in Baffin Bay (BB; Table S2 and Figure 2). However, even for BB, lithogenic particle concentrations in the deep (~1000-3500 m) basins were significantly lower than those measured in the shallow and shelf-dominated Canadian Arctic Archipelago (CAA; Table S2 and Figure 2).

The concentrations of pP, POC and pN in the Canadian Arctic Ocean did not follow the trends described above for lithogenic elements. Concentrations of these biogenic-derived elements were similar across the CAA, BB and LS, while significantly (*p-value* < 0.05) lower values were observed in the CB (Table S2 and Figure 2). The highest values of lithogenic particulate elements (Figure 2, red points) were commonly associated with near bottom waters, while biogenic-derived elements (pP, POC and pN) peak at the surface. The distribution of particulate Mn, unlike the other elements, was similar across the CB, CAA and BB, whereas much lower concentrations were measured in the LS (Table S2 and Figure 2).



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Figure 2. Observed concentration ranges of particulate trace elements (pAl, pV, pFe, pMn and pP) and particulate organic carbon and particulate nitrogen (POC and pN) in the Canada Basin (CB), Baffin Bay (BB) and the Labrador Sea (LS). Particulate Al, V, Fe and Mn data in the Canadian Arctic Archipelago (CAA) were retrieved from Colombo et al. (2021). Boxes mark the 25th and 75th percentiles for all data points, whisker bars represent 10th and 90th percentiles, solid green horizontal lines in each box represent the median, blue triangles represent average values, and red points are the outliers representing data that extend beyond the whiskers.

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4.2 Vertical distributions of particulate elements in the Canadian Arctic Ocean

275 4.2.1 Particulate Al, V, Fe and Mn

276 The Canada Basin: Surface ($\sigma_{\theta} < 24$ and z < 40 m) concentrations of pAl, pV, pFe and pMn slowly 277 decreased with depth down to the top of the winter Bering Sea Water halocline layer (wBSW), where 278 concentrations increased with depth, especially for pV and pMn. At the shelf-break CB2 station, a steady 279 increase of pAl, pV and pFe was observed with depth below the halocline assembly. This increase contrasts 280 with the low and relatively uniform values measured across the Atlantic Layer (AL= FSB+BSB) and 281 Canada Basin Deep Waters (CBDW) at CB3 (Figure 3). Particulate trace metal concentrations in the AL 282 and CBDW at CB4 were significantly higher (*p-value*< 0.001) than those of CB3. Particulate Mn profiles 283 resembled those of pAl, pV and pFe for surface and subsurface waters, but, unlike the previous elements, 284 no differences were found among the three stations below the halocline assembly (Figure 3).

285 *Baffin Bay:* Similar to CB, particulate concentrations in BB surface waters ($\sigma_{\theta} < 26.3$) were uniformly 286 low. This low signature extended down to the top of the Arctic Water (AW), where a subsurface peak was 287 observed, reaching concentrations as high as 34.2 nmol L⁻¹, 57.3 pmol L⁻¹, 7.12 nmol L⁻¹, 2084 pmol L⁻¹ for 288 pAl, pV, pFe and pMn, respectively (Figure 3). Underneath the AW, vertical distributions of pAl, pV and 289 pFe across the West Greenland Intermediate Waters (WGIW) and Baffin Bay Deep Waters (BBDW) 290 exhibited distinct spatial differences. The lowest concentrations of these elements were measured at station 291 BB2, followed by BB3 and BB1 (Figure 3). Clear maxima of pAl, pV and pFe were observed in the deepest 292 samples, close to the sea-floor, where concentrations were two to ten-fold higher than WGIW and BBDW 293 values (Figure 3). For pMn, this spatial difference in deep water (WGIW + BBDW) distributions among 294 BB stations was not as clear as that described for pAl, pV and pFe (BB1>BB3>BB2), albeit two to ten-fold 295 higher maximum concentrations were also found in the deepest samples (Figure 3).

296 The Labrador Sea: The concentrations of pAl, pFe and pMn in surface waters ($\sigma_0 < 27.4$) were lower 297 than those measured in BB, but in the same range as the CB values, with the exception of high surface pV 298 $(45.7\pm6.43 \text{ pmol } \text{L}^{-1})$, and the rather high pAl measured at 9 m depth (55.4 nmol L^{-1} ; Figure 3). 299 Interestingly, subsurface and deep water pFe and pMn concentrations at station LS2 were considerably 300 higher than at K1, which was not the case for pAl (Figure 3). At LS2 station, from ~300-500 m, a subsurface peak with concentrations as high as pAI: 20.4 nmol L⁻¹, pV: 17.5 pmol L⁻¹, pFe: 3.29 nmol L⁻¹, pMn: 301 302 182 pmol L⁻¹ was present. Particulate concentrations increased from the base of Labrador Sea Waters-2015 303 (LSW-2015) towards the sea floor, with the highest values found at the base of the Northeast Atlantic Deep 304 Water (NSDW; Figure 3). At station K1, a shallower subsurface peak of pAl was observed from 305 approximately 100-150 m, while pAl deep water concentrations remained nearly uniform across 306 LSW-2015, LSW-87/94 and NSDW, with much higher concentrations (7-fold higher than LSW and 307 NSDW) in the Denmark Strait Overflow Water (DSOW). Similarly, pFe and pMn did not change 308 significantly across LSW and NSDW, but a sharp increase in their concentrations was measured in DSOW 309 (~ two-fold higher than LSW and NSDW; Figure 3).

310	Despite the spatial variability across the deep CAO basins, as a general trend, pAl and pV
311	concentrations from CB, BB and LS, not accounting for benthic nepheloid layers, are higher than those
312	reported for the East Pacific Zonal transect (GP16), the South Atlantic Ocean (GA06_w), the Eastern
313	Tropical Atlantic Ocean (GA06_w) and the North Atlantic (GA01_e; Fitzsimmons et al., 2017; Milne et
314	al., 2017; Gourain et al., 2019; GEOTRACES Intermediate Data Product Group, 2021). Similarly, pMn
315	concentrations in CB and BB are considerably higher than those of aforementioned basins (~100-300
316	pmol kg ⁻¹), while LS concentrations fall within the range of reported values for other major basins. For pFe,
317	CAO concentrations are within the same range as those reported for the South Atlantic Ocean, the Eastern
318	Tropical Atlantic Ocean and the North Atlantic (~0.5-5 nmol kg ⁻¹), but higher than pFe measured in the
319	East Pacific Zonal transect (<1 nmol kg ⁻¹), where particulate trace metal concentrations are generally low
320	(Fitzsimmons et al., 2017; Milne et al., 2017; Gourain et al., 2019; GEOTRACES Intermediate Data
321	Product Group, 2021).



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323 **Figure 3.** Profiles of particulate Al, V, Fe, Mn, pP, POC, pN versus potential density (σ_{θ}) for the deep stations in the Canada Basin (upper panel: CB2-CB4), Baffin Bay 324 325 (middle panel: BB1-BB3) and the Labrador Sea (lower panel: LS2 and K1). As the deepest samples from Baffin Bay had extremely high concentrations of Al, V, Fe and Mn, these values are shown between parenthesis to emphasize the features of the rest of the profile. Colored bands label important water masses to facilitate the interpretation and discussion 326 of the trace element data following Aksenov et al. (2011), Kondo et al. (2016), Tang et al. (2004), Curry et al. (2011), Yashayaev and Loder (2009 and 2016). wBSW: winter Bering 327 Sea Water ($\sigma_{\theta} = 25.9-27.1$ and $z = \sim 100\ 300\ \text{m}$). LH: Lower Halocline, FSB: Fram Strait Branch, BSB: Barents Sea Branch (FSB+BSB: $\sigma_{\theta} = 27.9-28.0$ and $z = \sim 350-1200\ \text{m}$). CBDW: 328 Canada Basin Deep Water (σ_{θ} > 28.1 and z> 1200 m), AW: Arctic Water (σ_{θ} = 26.3 27.1 and z= ~40-180 m), TrW: transitional water (~180-300 m), WGIW: West Greenland 329 Intermediate Water ($\sigma_{\theta}=27.3-27.6$ and $z=\sim300-800$ m), BBDW: Baffin Bay Deep Water ($\sigma_{\theta}>27.6$ and z>800 m), LSW: Labrador Sea Water ($\sigma_{\theta}=27.70-27.81$; LS2: ~470-1870 m 330 and K1: ~140-2060 m), ISW: Icelandic Slope Water, NSDW: Northeast Atlantic Deep Water (σ_{θ} = 27.81-27.86; LS2: ~1870-2400 m and K1: ~2060-2640 m), DSOW: Denmark 331 Strait Overflow Water (σ_{θ} > 27.86; LS2: >2400 and K1: >2640 m).

332 *4.2.2 Particulate phosphate (pP), organic carbon (POC) and nitrogen (pN)*

The Canada Basin: The lowest pP, POC and pN concentrations of this study were measured in this basin; these elements peaked at the surface (z = ~10) and subsurface ($\sigma_{\theta} = 24.8-25.2$ and z = 60-70 m; Figure 3), where transmissivity diminished and chlorophyll-a (Chl-a) was highest (Figure S2). Below 100 m ($\sigma_{\theta} = ~25.7-26.1$), pP, POC and pN decreased with depth, although the difference in concentrations between surface and deep waters was not as large as that observed in the CAA, BB and LS (Figure 3 and 4).

Baffin Bay: pP, POC and pN were highest in the upper 40 m (σ_{0} < 26.3), with concentrations ranging from 43.7 to 72.1 nmol L⁻¹, 6.29 to 8.57 µmol L⁻¹ and 1.02 to 1.65 µmol L⁻¹, respectively, at BB1 and BB2, and highest at BB3 (pP: 120 nmol L⁻¹, POC: 15.9 µmol L⁻¹, pN: 3.04 µmol L⁻¹; Figure 3). Across AW, POC and pN concentrations were lowest at station BB3, followed by BB1 and BB2 (Figure 3), where a deeper subsurface Chl-a peak and transmissivity drop were recorded (Figure S2). Underneath AW, pP, POC and pN continued to decline with depth through the WGIW (Figure 3)

The Labrador Sea: pP, POC and pN were elevated in surface waters (z< 40 m) and sharply decreased from the subsurface waters to the top of LSW-2015 (Figure 3). Underneath LSW-2015, pP, POC and pN concentrations steadily decreased with depth to the DSOW, where minima in POC and pN concentrations were reached.

The Canadian Arctic Archipelago: As described for the deep basins, pP, POC and pN were highest in surface waters, while much lower concentrations were measured in deeper waters ($\sigma_{\theta} = \sim 26.3-26.8$ and z > 100 m). Marked differences in the distributions of these elements were noticed in the CAA, with considerably lower concentrations measured in the western CAA (CB1 and CAA8 stations) compared to eastern CAA regions (CAA1-CAA7 and CAA9; Figure 4). Surface concentrations of pP, POC and pN in the western CAA ranged between 8.08 to 25.3 nmol L⁻¹, 1.95 to 3.33 µmol L⁻¹ and 0.109 to 0.425 µmol L⁻¹, respectively (Figure 4). On the other hand, in the eastern CAA, surface and subsurface concentrations were

- 355 higher (pP: 31.1-74.9 nmol L⁻¹, POC: 7.86-27.78 μ mol L⁻¹, pN: 0.953-3.304 μ mol L⁻¹), and decreased
- 356 uniformly to the bottom (Figure 4).



Figure 4. Profiles of pP, POC and pN versus potential density (σ_{θ}) in the Canadian Arctic Archipelago (CAA). Note that the vertical axis scales vary.

5 Discussion 359

360

5.1 The importance of advective transport, nepheloid layers and authigenic Mn oxides in modulating the distributions of lithogenic elements and Mn 361

In order to elucidate the sources and dominant phases (i.e. lithogenic, biogenic, authigenic) of 362 363 particulate samples in the Canadian Arctic Ocean (CAO), we normalized our data to pAl, and compared elemental pAl ratios to average upper continental crustal ratios (UCC; Shaw et al., 2008; Rudnick and Gao, 364 365 2013). Particulate Al has been extensively used as a lithogenic tracer due to its high natural abundance in 366 the earth's crust, its similar concentration range in both the UCC and bulk continental crust (the relatively 367 constant ratio of metal to aluminum in crustal rocks) and its scarce anthropogenic sources (Covelli and 368 Fontolan, 1997; Ohnemus and Lam, 2015; Lam et al., 2018; Lee et al., 2018; Gourain et al., 2019).

369 In the CB, BB and LS, particulate concentrations show three distinctive patterns. On one hand, pV and 370 pFe were strongly positively correlated with pAl across many orders of magnitude ($R^2 = 0.83$ and 0.90, 371 respectively), and their ratios were in agreement with those reported for the UCC (Figure 5, upper panel), 372 reflecting the dominance of lithogenic-derived inputs of these elements. On the other hand, pP was instead correlated with POC and pN (section 5.2) and decoupled from pA1 ($R^2 = 0.01$), with ratios that strongly 373 374 deviated from the UCC (Figure 5), which evidence the dominant role of biogenic sources (i.e. 375 phytoplankton biomass). And last, unlike the aforementioned particulate elements, pMn display only a 376 moderate correlation with pAl (R2=0.60; Figure 5, upper panel), with distributions that vary greatly in the 377 lithogenic fraction (from 1 to 100%; Figure 5, lower panel). These results suggest that distinct particle 378 inputs and/or alteration processes modulate pMn in the deep CAO basins.

379 No significant correlations ($R^2 < 0.01$ and *p*-value> 0.05) were found between pAl, pV, pFe and pMn 380 versus pP, which indicate that inputs from phytoplankton do not contribute substantially to variations in 381 these particulate elements across the CAO. Therefore, the biogenic fraction of pFe -the most abundant trace 382 metal in phytoplankton (Ho et al., 2003)- is completely masked by its lithogenic input (Figure 5).



Figure 5. Upper panels: relationship between pV, pFe, pMn and pP versus pAl in the Canada Basin (CB), Baffin Bay (BB) and the Labrador Sea (LS); note the logarithmic scale of the plots. The depth of the samples is indicated with the color scale; data are from all deep basin stations and all depths. Linear regression lines and R² values are displayed in the figure (light gray), as well as average upper continental crustal (UCC) ratios for these elements indicated by the dashed and dash-dotted black lines. UCC particulate elements to Al molar ratios are selected from Shaw et al. (2008), who reported continental surface Precambrian shield composition in Canada (crustal ratio A), and from an updated continental crust composition review (crustal ratio B; Rudnick and Gao 2013). Additionally, the lithogenic fraction (%) of some samples is annotated in the figure to illustrate differential sources for these elements. Samples that lie along UCC lines are assumed to be entirely derived from lithogenic sources (100%). Samples plotting along the crustal ratios are assumed to be entirely derived from lithogenic sources (100%). Samples plotting along the crustal ratios are assumed to be entirely derived from lithogenic sources (100%). Samples plotting along the crustal ratios are assumed to be entirely derived from lithogenic sources (100%). Samples plotting along the crustal ratios are assumed to be entirely derived from lithogenic sources (100%). Samples plotting along the crustal ratios are assumed to be entirely derived from lithogenic sources (100%). Samples plotting along the crustal ratio $\frac{pTE}{pAL}_{uCC}$, where pTE is the lithogenic fraction of a given particulate trace element, and the UCC are the upper continental crust ratios retrieved from Rudnick and Gao (2013), as this work and the upper continental crust ratios retrieved from Rudnick and Gao (2013), as this work are traceded as the lithed and the data determines are traceded as the lithogenic fraction of a given particulate trace element, and the UCC a

392 presented the most updated review of the continental crust composition.

5.1.1 The influence of lateral advection of shelf-derived waters and Mn oxide formation on
surface and subsurface particulate distributions in the CB, BB and the LS

Due to the high latitude and remote location of the CAO, atmospheric fluxes tend to be attenuated in 395 396 this region (Marsay et al., 2018; Shelley et al., 2018). This fact likely explains the relatively low surface concentrations measured in CB, BB and LS (Figure 3), far away from shelf areas where freshwater and 397 398 landfast ice are important sources of particulate elements (Giesbrecht et al., 2013; Kondo et al., 2016; 399 Gourain et al., 2019). Lithogenic inputs dominate the concentrations of pAl and pFe (> 90 %) in surface 400 waters across the entire CAO (Figure 5). Among the deep water basins, the highest pAl and pFe 401 concentrations were measured in BB surface waters (Figure 3), which captures the CAA outflow enriched 402 in pAl and pFe (Colombo et al., 2021). In the CB, the downwelling Beaufort Gyre entrains and accumulates 403 freshwater and sea ice (Proshutinsky et al., 2009). These freshwater inputs, and its associated lithogenic 404 particulate load (Measures, 1999; Giesbrecht et al., 2013), to CB surface waters (S= 25.1–29.2; Figure S1) 405 are expected to drive the higher pAl and pFe values found at the shallowest depths (200 to 400% higher) 406 compared with underlying waters (Figure 3).

407 On the other hand, the lithogenic contribution of pV and pMn in CAO surface waters represents less 408 than 40 and 30% of bulk concentrations. Elevated non-lithogenic contributions of pMn and pV could be 409 linked to enhanced primary production at the surface, which increases the export of biogenic-derived 410 particles (Twining et al., 2015; Lee et al., 2018; Gourain et al., 2019; Whitmore et al., 2019). Nonetheless, 411 strong phytoplankton production of pMn (essential micronutrient) in surface waters is not supported by our 412 dataset given that non-lithogenic pMn fractions do not change significantly across CB and BB (Figures 3 413 and 6), despite the large differences in the biological productivity observed among these two deep basins 414 (CB Chl-a <<< BB Chl-a; Figures S2, S3 and S4). Indeed, the lowest pMn concentrations were measured 415 in LS (Figure 3), where intense phytoplankton spring blooms took place (Figures S3 and S4). Moreover, 416 pMn distributions in the CAO did not follow the trends of pP, POC and pN, whose spatial and vertical 417 distributions are strongly controlled by phytoplankton productivity (section 5.2). Hence, the excess of pMn

over UCC values are likely explained by authigenic formation of Mn^{+3/4} oxides, most probably 418 419 bacterially-mediated Mn^{+2} oxidation/precipitation. Even though the abiotic oxidation of Mn is 420 thermodynamically favored under typical pH and pO_2 conditions in most oceanic waters, this reaction has 421 an extremely low kinetic rate. Bacteria are known to accelerate Mn⁺² oxidation, and therefore are considered 422 the most important drivers of Mn precipitation in aquatic environments (Cowen and Silver, 1984; Cowen 423 and Bruland, 1985; Sunda and Huntsman, 1988; Moffett, 1997; Lam and Bishop, 2008; Yigiterhan et al., 424 2011; Noble et al., 2013; Lee et al., 2018; Wright et al., 2018; Morton et al., 2019). Surface pMn 425 concentrations in the CAO are lowest in the upper water column, despite being largely dominated by 426 non-lithogenic sources (~70-95%; Figures 3 and 6). This low surface pMn concentrations may reflect 427 diminished bacterial Mn oxidizing activity (photoinhibition) and photoreduction of Mn oxides (Mn^{+3}) by 428 sunlight, resulting in lower rates of particulate Mn formation in surface waters (Sunda and Huntsman, 1988; 429 Sunda and Huntsman, 1990; Francis et al., 2001). Particulate V cycling in CAO surface waters is tightly 430 coupled with pMn, as dissolved V (dV) has proven to be easily scavenged onto Mn oxide particles 431 (Whitmore et al., 2019 and references therin). The unexpectedly high pV in surface LS waters $(45.7\pm6.43 \text{ pmol } \text{L}^{-1} \text{ and } \text{Litho}_{\text{PV}} < 7\%; \sim 100 \text{ to } 200 \% \text{ higher than CB and BB surface concentrations;}$ 432 433 Figure 3) where the lowest pMn concentrations were measured, could be attributed to the stripping of dV 434 by the increased particle flux (Whitmore et al., 2019) associated with the aforementioned phytoplankton 435 bloom (Figures S2, S3 and S4).

Beneath surface waters, pAl and pFe concentrations rapidly increase within the winter Bering Sea Water (wBSW) in CB and the Arctic Water (AW) in BB (Figure 3). These subsurface waters are advected from the shallow and shelf-dominated Chukchi Sea and CAA, respectively, carrying a unique biogeochemical signature to the deep basins. The wBSW is distinguished by a weak temperature minimum, high nutrient concentrations, high dissolved nutrient-like trace metals, high colored dissolved organic matter (CDOM), and low oxygen concentrations (Hioki et al., 2014; Kondo et al., 2016; Colombo et al., 2020; Figures S1 and S2). These properties show the signature of the large organic matter remineralization 443 and sedimentary denitrification (low N* values) that occurs over the Bering and Chukchi shelves (Hioki et 444 al., 2014; Granger et al., 2018). The fresh CAA outflow into Baffin Bay is also characterized by a 445 temperature minimum, high dissolved nutrient-like trace metals, high CDOM, and relatively high oxygen 446 concentrations, resulting from the intense mixing and sediment resuspension in the CAA (Colombo et al., 447 2020; Colombo et al., 2021; Figures S1 and S2). The elevated values of pAl and pFe in both wBSW in CB 448 and AW in BB are attributed to advective transport of lithogenic-derived particles (Litho pFe: ~80-100 %; 449 Figure 5) from the shallow Chukchi and CAA shelves, where the incorporation of resuspended sediment 450 into shelf waters has been recently reported (Xiang and Lam, 2020; Colombo et al., 2021). The sediment-451 rich wBSW in CB and AW in BB cascade downslope, and are then transported long distances along their 452 flow paths (e.g. >1400 km from CAA3 and CAA4 in the southern Lancaster Sound, via BB3 on the Baffin 453 Bay slope and out to BB1 in the Davis Strait).

454 In contrast, the much sharper subsurface pMn peak in CB and BB is dominated by the non-lithogenic fraction (wBSW: > 97 % and AW: > 90 %, respectively; Figures 3 and 6). Distinctively high Mn oxide 455 concentration, many times higher than those found in other oceanographic regions, was also described by 456 457 Xiang and Lam (2020) in halocline waters in the Chukchi Abyssal Plain, the Makarov Basin and western 458 Canada Basin. Here, we demonstrate that this Mn oxide signature is still prevalent in the Beaufort Sea (CB 459 stations), and in BB (Figure 3). The pMn peak in wBSW and AW is hypothesized to be produced within 460 the water column (bacterially-mediated Mn oxide precipitation), rather than being an advected sedimentary signature (reduction in the sediment \rightarrow diffusion to the overlying water \rightarrow oxidation \rightarrow precipitation of redox 461 sensitive elements such as Fe and Mn). 462

In the Chukchi Sea, where large pulses of organic matter trigger strong reducing conditions in sediments, large fluxes of dissolved manganese (Mn^{+2}) are generated in porewater and diffused into the overlying seawater (Granger et al., 2018; Vieira et al., 2019; Xiang and Lam, 2020). Given its slow oxidation kinetics and the potential photoinhibition of Mn oxidizing bacteria in the shallow Chukchi shelves (< 50 m), most of Mn^{+2} remains in the dissolved phase for a long time in the halocline waters (wBSW) – leaving Chukchi shelf waters depleted of pMn oxides and highly enriched in dMn (Xiang and Lam, 2020),
which are transported to the slope region where Mn authigenically re-precipitates.

470 In the CAA, characterized by reduced pulses of organic matter and strong mixing regimes, the 471 reductive benthic supply of Mn⁺² and its subsequent precipitation is not anticipated to be significant, and 472 the dominance of Mn oxides in the CAA is related to the advection of wBSW from the CB (Colombo et 473 al., 2021). The strongly reduced authigenic sedimentary formation of Mn oxides in the Chukchi Sea and 474 CAA, and the ubiquity of non-lithogenic pMn across the entire water column in CAO (with the exception 475 of LS), suggest that pMn in wBSW and in AW is a result of increased oxidative Mn precipitation in the 476 water column, as described for other basins (Sunda and Huntsman, 1988; Yigiterhan et al., 2011; Noble et 477 al., 2013; Morton et al., 2019). In effect, the pMn peak in both wBSW in CB and AW in BB is associated 478 with a rapid decrease with depth in dMn concentrations in these basins, where the pMn fraction transitions 479 from less than 20% of total Mn (dMn + pMn) in surface waters (dMn is dominant) to \sim 55-70% in wBSW 480 and AW (Figures S5 and S6).

481 Marine microbial communities, including Mn oxidizing bacteria, are widely distributed in ocean 482 waters, however, their abundance and diversity vary both spatially and vertically based on environmental 483 conditions (e.g. light penetration, temperature, availability of labile organic matter, nutrients and 484 micronutrients such as Fe and Mn; Dick et al., 2006; Zakharova et al., 2010; Zinger et al., 2011). Both, the 485 wBSW and AW have the potential to sustain large populations of Mn oxidizing bacteria given their 486 distinctive environmental conditions. These water masses are relatively deep -where sunlight, and 487 therefore, microbial photoinhibition is attenuated- and have elevated concentrations of dissolved Mn and 488 Fe, advected from the shallow Chukchi and CAA shelves, which would allow Mn oxidizing bacteria to 489 proliferate (Sunda and Huntsman, 1988; Moffett, 1997; Zakharova et al., 2010; Colombo et al., 2020). 490 Another feature that wBSW and AW have in common is their close interaction with the seafloor upstream 491 while they transit shallow shelf environments (Chukchi Sea and the CAA). As result of the interaction with 492 the seafloor, where microbe communities are taxonomically richer and more abundant than in the water

493 column (Feng et al., 2009; Zinger et al., 2011), wBSW and AW are not only enriched in lithogenic particles 494 (as evidenced by pAl and pFe distributions) and CDOM, but they also could potentially entrain sedimentary 495 microbes (Walsh et al., 2016). An analogous process has been described for hydrothermal plumes, which 496 entrain manganese oxidizing bacteria from surficial sediments and transport them long distances; resulting 497 in high rates of bacterially-mediated Mn^{+2} oxidation within these plumes (Dick et al., 2006; Dick et al., 498 2013; Fitzsimmons et al., 2017).

499 We posit two complementary mechanisms which could be driving the sharp non-lithogenic pMn peak 500 in wBSW in the CB interior: 1) advective transport of small Mn oxides particles precipitated over the 501 Chukchi shelf-break and/or slope region 2) enhanced precipitation/concentration effect of dMn supplied 502 from above. Strong bacterially-mediated oxidation occurs in the water column over the shallow Chukchi 503 shelf-break, where the high dMn signature (~ 40-200 nmol kg⁻¹) is rapidly lost (Kondo et al., 2016; Vieira 504 et al., 2019; Jensen et al., 2020). Once the wBSW is advected to the ocean interior, dMn concentrations are 505 further diminished over the Chukchi slope, dropping to 1.34 ± 0.57 nM about 350 km away from Chukchi 506 shelf (Kondo et al., 2016). Beyond the slope towards the CB interior, dMn is conserved, as the dMn 507 concentrations measured in the CB interior $(1.37 \pm 0.41 \text{ nmol kg}^{-1}; \text{ Colombo et al., 2020})$ are nearly the 508 same as those measured ~350 km offshore Chukchi Sea. The rapid loss of dMn over the slope and the 509 bacterially-mediated Mn oxide precipitation is reflected in the sediment record. The nearshore shallow 510 sediments within the Chukchi shelves, contain low and lithogenic derived Mn, whereas in the slope region, 511 the sediments are greatly enriched in non-lithogenic Mn (Macdonald and Gobeil, 2012). Although most of 512 the non-lithogenic Mn particulates originating in the shelf-break and/or slope region are rapidly lost due to 513 sinking, the smaller sized Mn oxides could be entrained and transported long distances to the CB interior. 514 Small microbially catalyzed pMn oxides (Cowen and Silver, 1984; Cowen and Bruland, 1985) have been 515 shown to be transported long distances (4000 km at 0.2-0.5 cm s⁻¹) from hydrothermal vent sources 516 (Fitzsimmons et al., 2017). In addition, no significant pMn settling across isopycnals was observed in the 517 distal hydrothermal plume due to the small (0.5-1 μ m) and low density nature of Mn oxide coatings on

518 bacterial cells (Fitzsimmons et al., 2017; González-Santana et al., 2020). The Stokesian settling velocities 519 of pure birnessite (major manganese-bearing mineral; density= 2900 kg m⁻³) particles with 0.5 to 1 μ m diameters are 4-17 m y⁻¹, however, these values are likely reduced if we account for the lower specific 520 521 gravity of bacterial-Mn oxide capsule complexes. Results from Ocean Parcels particle track simulations 522 (Methods, section 3.3) suggest that it takes from about one to three years for the wBSW to travel from the 523 Chukchi shelves to our sampled stations (CB2: 580 km at 1.8 cm s⁻¹, CB3: 896 km at 2.8 cm s⁻¹, CB4: 410 km at 1.3 cm s⁻¹; speeds are an average estimate based on the distance the parcels travelled in a given time 524 525 period within Ocean Parcels). Given the timescale of wBSW circulation in the CB, and even considering 526 the sinking rates of pure birnessite, advective transport of small-size pMn from the Chukchi shelf-break is 527 a plausible mechanism explaining the wBSW pMn peaks observed in the CB.

528 Enhanced oxidative precipitation of the Mn flux (pMn + dMn) falling through the water column could 529 be a complementary mechanism. Ice-rafted sediments likely supply a significant Mn flux to the Canada 530 Basin upper ocean (Measures, 1999; Rogalla et al., 2021). Because of the fast reversible oxidation / 531 reduction cycling of Mn, with timescale of a couple of weeks (oxidation rate 0.06 per day and reduction 532 rate 0.04 per day; Bruland et al., 1994; Rogalla et al., 2021), Mn is cycled back and forth between the 533 dissolved and oxidized pools multiple times once it is sourced from surface waters. While Mn is in its 534 oxidized phase, particles will sink at rates governed by their density and size, thus generating a downward 535 flux. This Mn flux is then entrained in wBSW, where Mn oxidizing bacteria populations are potentially 536 abundant and Mn oxidation rates may be greater. The lighter and smaller bacterially-mediated Mn oxides 537 (Fitzsimmons et al., 2017) that are formed in the wBSW barely sink (Stokesian settling velocities for 0.5-1 μ m particles < 17 m y⁻¹), and thus, would be concentrated in this water mass. These two mechanisms 538 described above are also expected to take place in AW in the BB, where sharp non-lithogenic pMn peaks 539 540 were observed in this study.

541 The pV maxima in wBSW and AW have relatively high non-lithogenic contributions (~70-92 and 542 ~55-75 %, respectively; Figures 3 and 6), attributed to the scavenging of dV onto the abundant authigenic 543 Mn oxides present in these subsurface waters. A similar pV peak in CB around the 26.6 kg m⁻³ isopycnal 544 surface (wBSW) was reported by Whitmore et al. (2019), who argued that the strong dV removal and the 545 subsequent pV enrichment is largely controlled by the adsorption of dV onto Mn oxides, strong scavengers 546 of many trace elements, in deep basins.

547 In the LS, lithogenic inputs govern the subsurface distribution of pAl and pFe (100%), as well as those of pV (~54-78 %) and pMn (~38-60 %), compared to CB and BB where authigenic Mn oxides exert a 548 549 dominant role shaping pV and pMn subsurface distributions (Figure 5). The diminished influence of pMn 550 oxides in the LS could be explained by micronutrient limitation (such as dissolved Mn or Fe) of Mn 551 oxidizing bacteria in this region (Colombo et al., 2020), and/or the relative isolation of LS from continental 552 margins with regards to CB and BB. Further research, such as bioassay/incubation experiments, genomic 553 measurements and taxonomic identification, is required to unravel the authigenic Mn cycling in CB and 554 BB, and the notable reduction of authigenic Mn formation in LS.

555 5.1.2 Boundary transport of resuspended sediments and near-bottom enrichment of pTE

The Arctic Circumpolar Boundary Current (ACBC; σ_{θ} > 27.9 and z> 350 m) is anticipated to modulate 556 557 the distributions of pAl, pV and pFe in CB, causing higher concentrations of these elements at CB2 station, 558 followed by CB4 and CB3 (Figure 3). The main ACBC components, the Fram Strait Branch (FSB) and 559 Barents Sea Branch (BSB), travel around the rim of the Arctic Ocean (Figure 1), mobilizing sediment 560 particles from the continental margin, and advecting these particle-rich waters to the Canada Basin interior 561 (Aksenov et al., 2011; Aguilar-Islas et al., 2013; Giesbrecht et al., 2013; Kondo et al., 2016; Xiang and 562 Lam, 2020). As a result of their locations, both CB2 and CB4 are more directly impacted by FSB and BSB 563 cyclonic circulation than CB3 station. This difference is notably reflected in substantially contrasting 564 concentrations of pAl, pV and pFe in open ocean CB stations (CB4 >> CB3), and much higher concentrations observed at the shelf-break CB2 station, where transmissivity values were lowest (Figures 565 566 1, 3 and S2). The relative isolation of CB3 station from the ACBC and enhanced advective transport of 567 dissolved Fe and Mn to CB4 has been recently documented (Grenier et al., 2019; Colombo et al., 2020).

Here we show that mid-depth lateral transport from continental margins also exerts a strong control over pAl, pV and pFe distributions, and possibly other lithogenic-derived elements. Preferential sinking of larger particles, with faster settling velocities than smaller ones, likely explains the high concentrations of pAl, pV and pFe observed close to the shelf-break and slope stations and their reduction along the flow path, as reported in studies where size-fractionated data were available (Lam et al., 2015b; Lam et al., 2018; Lee et al., 2018; Xiang and Lam, 2020).

574 The vertical distributions of pAl, pV and pFe are also strongly influenced by advective transport of 575 resuspended sediments from continental margins in the WGIW in BB (Figure 1). The highest concentrations 576 of pAl, pV and pFe were measured in Davis Strait (BB1), where the tidal stresses are the highest of BB (~0.016 to 0.069 m²s⁻²; Figures 3 and 7). These high concentrations are likely associated with the 577 578 resuspension of sediments in this region where intense cyclonic boundary currents have been recorded 579 (Tang et al., 2004). In contrast, pAl, pV and pFe were uniformly low at station BB2 (Figure 3), which is 580 located in the center of Baffin Bay (Figure 1), largely isolated from the cyclonic circulation of WGIW 581 (Colombo et al., 2019). Station BB2 has also low tidal stresses, suggesting reduced prevalence of sediment 582 resuspension (tidal stress $< 0.0009 \text{ m}^2\text{s}^{-2}$; Figure 6). Vertical distributions of pMn in CB and BB, in contrast 583 to pAl, pV and pFe, were similar among sampled stations, despite the disparate influence of advective 584 transport of resuspended particles from continental margins in both basins (Figure 3). This distinctive 585 behavior is linked to the relatively low lithogenic contributions of pMn (~ 5-30%; Figures 3 and 6) in FSB, 586 BSB and WGIW waters, which contrasts with those of pV and pFe (~30-70 and > 90%, respectively).

587 Distributions of particulate Al, V, and Fe in Labrador Sea deep waters (LSW, NSDW and DSOW) are 588 predominantly controlled by lithogenic sources (Litho_pV: ~ 55-77 and Litho_pFe: > 90%). Their 589 concentrations are relatively low and similar to those measured in the isolated central stations in CB and 590 BB (CB3 and BB2; Figure 3), which reveals reduced lateral transport of these elements from continental 591 slope regions. Compared to CB and BB deep waters, pMn has its lowest concentrations (Figure 3), and the strongest lithogenic contribution (~ 30-100%; Figure 5) in LSW, NSDW and DSOW, presumably related
to diminished Mn oxidizing activity in LS as discussed in section 5.1.1.





602 CB4), BB (BB1-BB3) and LS (LS2 and K1), with concentrations considerably higher than those measured 603 in overlying waters (Figure 3). At the Baffin Bay stations located along the slope (BB3) and Davis Strait 604 (BB1), the bottom samples are highly enriched in particulate elements, and have pTE:pAl ratios largely 605 exceeding those of UCC and relatively low lithogenic fractions (Litho_pV: 20 and 47%, Litho_pFe: 42 and 606 73%, Litho_pMn: 8 and 13%, for BB3 and BB1 respectively; Figures 3 and 6). The relatively low lithogenic 607 fraction of pFe is particularly notable as its distribution is dominated by lithogenic sources (close to 100%) 608 in the vast majority of the analyzed samples (Figure 5, lower panel). This evidence shows that redox 609 mobilization of reduced Mn and Fe species and their subsequent precipitation is an important source of pFe 610 and pMn for the deepest samples at BB1 and BB3. This signal is superimposed to that of the resuspension 611 of lithogenic particles as demonstrated by a spike in pAl. At station BB2 in central Baffin Bay, the presence 612 of authigenic Mn (redox-mobilized) differs from the lithogenic prevalence of pV and pFe (Litho pV: 64%, 613 Litho_pFe: 100%, Litho_pMn: 13%). This difference is likely related to lower productivity and organic 614 matter inputs in central Baffin Bay compared to shelf regions (Wyatt et al., 2013; Figures S2 and S3) and/or 615 increased organic matter remineralization in the water column at the deeper BB2 station (BB1 &BB2: 1000 616 m vs. BB2 > 2000 m). These environmental conditions would in turn presumably decrease reducing 617 conditions in the sediments at BB2 station, yielding a redox potential appropriate for Mn reduction, but 618 higher than that required for Fe reduction. In LS, where particulate concentrations rapidly increased towards 619 near-bottom waters (Figure 3), the deepest samples have pTE:pAl ratios similar to those of UCC and greater 620 lithogenic contributions (Litho_pV: 60%, Litho_pFe: 100 and 100%, Litho_pMn: 52 and 100% for LS2 621 and K1 respectively; Figure 5). The flow of deep water boundary currents (NSDW and DSOW) over bottom 622 topography in LS (Figure 1) is expected to cause intense sediment resuspension events (Middag et al., 623 2015), as suggested by the sharp decrease of dissolved trace elements that are particle reactive, such as dissolved Pb and ²³⁰Th at LS2 and K1 stations below 2500 m (Colombo et al., 2019; Grenier et al., 2018), 624 625 and consequently results in elevated lithogenic-derived particulate elements (Figure 3). This energetic 626 boundary current in LS, and associated sediment resuspension events, probably diminishes reductive

sedimentary processes, thereby explaining the lower contribution of authigenic pMn oxides to bottom LS
waters, as suggested by Ohnemus and Lam (2015).

5.2 Biogenic-derived elements: pP, POC and pN dynamics across the

630 productivity gradients of the Canadian Arctic Ocean

631 In the Canadian Arctic Ocean (CAO), particulate phosphorous (pP) as well as particulate organic 632 carbon and nitrogen (POC and pN) were elevated in the upper 100 m, where Chl-a peaked and 633 transmissivity dropped, indicating the dominant role of biological production in modulating vertical 634 distributions of these elements (Figures 3-5). In contrast, below ~100 m, the predominance of organic matter 635 regeneration is reflected in a rapid decrease of pP, POC and pN with depth, with concentrations remaining 636 uniformly low in deep waters (Figures 3 and 5). The spatial distributions of these elements revealed notable 637 differences between CB and the rest of the CAO, with extremely low pP, POC and pN in CB, similar to 638 those reported by Trimble and Baskaran (2005) and Brown et al. (2014), in contrast to those observed in 639 CAA, BB and LS (Figure 2). These differences are linked to the low Chl-a (Figures S2, S3 and S4), primary 640 productivity and export production rates in CB compared to the much higher values in the rest of CAO (Hill 641 et al., 2013; Varela et al., 2013; Wyatt et al., 2013; Crawford et al., 2015; Lehmann et al., 2019; Xiang and 642 Lam, 2020). The low levels of primary productivity in CB result from both light and nitrate limitation 643 (Tremblay & Gagnon, 2009; Tremblay et al., 2008), which could be further limited by dissolved Fe 644 availability in the summertime (during phytoplankton blooms) as described by Taylor et al. (2013).

Within the CAA, pP, POC and pN also display a spatial contrast between western and eastern CAA regions, with lower concentrations measured from M'Clure Strait to Viscount Melville Sound (stations CB1 and CAA8) versus those from Barrow Strait and Lancaster Sound (Figure 4). It should be noted that the stations CB1 and CAA8 in western CAA were sampled in early and late September, whereas eastern stations were sampled in early August, potentially affecting the distribution of pP, POC and pN due to the seasonal solar irradiance variation (Michel et al., 2015). However, the spatial differences in pP, POC and 651 pN distributions in the CAA observed in this study are consistent with the variability in phytoplankton 652 communities and regimes identified by Ardyna et al. (2011). The Beaufort Sea and the western CAA are 653 characterized by an oligotrophic flagellate-based system which transitions to an eutrophic diatom-based 654 system in eastern CAA (Lancaster Sound). Therefore, an interplay of multiple environmental processes 655 affecting light and nutrient availability may explain the above noted spatial differences in this shallow 656 environment. Sea ice pervasiveness (limiting solar irradiation), increased stratification (hindering nutrient 657 replenishment from bottom waters), and reduced availability of micronutrients (e.g. Fe) in the western 658 CAA, contrast with the longer ice-free season and enhanced mixing, sediment resuspension and 659 micronutrient availability of the eastern CAA (Ardyna et al., 2011; Colombo et al., 2021; Hughes et al., 660 2017; Michel et al., 2006; Michel et al., 2015). The distributions and concentrations of pP, POC and pN in 661 the CAO, and in the CAA in particular, agree with previous studies in the Arctic region (Trimble and 662 Baskaran, 2005; Wyatt et al., 2013; Brown et al., 2014), and correlate with primary production patterns 663 (Chl-a) retrieved from both satellite data and in-situ measurements during the Canadian Arctic 664 GEOTRACES cruise (CB and western CAA << eastern CAA, BB and LS; Figures 3, 5, and S2-S4).

665 The particulate C:N:P ratios from all particulate samples collected during the Canadian Arctic GEOTRACES cruise show strong POC:pN and POC:pP relationships in the deep basins ($R^2 = 0.94$ and 666 0.80, respectively) and the shallow CAA ($R^2 = 0.93$ and 0.63, respectively, Figure 7). Although C:N:P ratios 667 668 are known to be influenced by phytoplankton community composition, and hence, regional differences 669 exist, our data compare well with the canonical Redfield ratio (106:16:1) and with recent global elemental phytoplankton composition estimates (166.6:23.4:1; Martiny et al., 2013; Figure 7). Within the euphotic 670 671 zone, POC:pN and POC:pP ratios agree more closely with Redfield and Martiny ratios and the lithogenic 672 contributions to pP are negligible, whereas the deeper samples (> 100 m) are more dispersed, deviating 673 from phytoplankton ratios, and have slightly higher lithogenic contributions to pP (Figures 5 and 8). In the 674 upper 100 m of BB and LS, the most productive basins in the CAO, particulate C:N ratios (6.3 ± 1.3) closely 675 reflect phytoplankton stoichiometries (6.6-7.1; Martiny et al., 2013), while in the CAA these ratios are

676 somewhat higher (8.4 ± 2.6) . This is most probably related to enhanced terrigenous-derived organic matter 677 inputs with higher C:N ratios (Dittmar and Kattner, 2003) in this land-dominated environment where shelf-water interactions and freshwater inputs are increased (Colombo et al., 2021). In the upper 100 m of 678 679 CB, however, particulate C:N ratios are very scattered and deviate from Redfield and Martiny ratios, with 680 lower ratios at CB4 (3.0-7.4) and much higher ratios at CB2 and CB3 stations (6.7-19.9). A similarly high 681 POC:pN ratio (17.9) is observed in the upper 50 m at the entrance of M'Clure Strait (station CB1). Past 682 studies have described the large spatial variability in particulate C:N ratios in the CAO. While, extremely 683 low ratios (2.1-3.5) in central CB are associated with low concentrations of phytoplankton carbon and larger 684 contribution of small flagellates, higher ratios (> 10) are observed in shelf waters, which are potentially 685 related to the influence of freshwater inputs (riverine, glacial, and/or sea ice meltwater) of allochthonous 686 carbon detritus (Brown et al., 2014; Crawford et al., 2015; Fragoso et al., 2017). Lastly, compared with 687 phytoplankton stoichiometry, the consistently higher POC:pP ratios of the deepest samples in CB, BB and 688 LS, as well as in M'Clure Strait (Figure 7), may reflect the preferential remineralization of P relative to C 689 in sinking particulate matter (Faul et al., 2005).



Figure 7 Relationship between POC versus pN and pP in deep basins (upper panels) including the Canada Basin (CB), Baffin Bay (BB) and the Labrador Sea (LS and K1), and in the shallow Canadian Arctic Archipelago (lower panels; CAA). Note the logarithmic scale of the plots. The depth of the samples is indicated with the color scale; data are from all 16 stations and all depths. Linear regression lines and R² values are displayed in the figure (light gray), as well as Redfield and Martiny POC:pN (6.6 and 7.2) and POC:pP (106 and 167) stoichiometric ratios indicated by the dashed and dash dotted black lines (Martiny et al., 2013).

695 **6 Conclusions**

696 The present study investigates the processes which control the biogeochemical cycling of particulate 697 elements (Al, V, Fe, Mn, P, POC and pN) in the Canadian Arctic Ocean (CAO) deep basins, as well as pP, 698 POC and pN in the shallow Canadian Arctic Archipelago (CAA). A combination of both lithogenic sources 699 and enhanced scavenging onto Mn oxides appears to shape pV distribution in surface and subsurface waters 700 (Figure 8). Particulate Al, Fe and V have identical deep water profiles (z > 300 m), revealing the importance 701 of lateral transport of resuspended sediments from shelf/slope regions. The Canada Basin (CB), Baffin Bay 702 (BB) and the Labrador Sea (LS) are all characterized by strong deep water boundary currents (ACBC, 703 WGIW, NEADW and DSOW), which flow along shelf and slope regions, become enriched in resuspended 704 particles, and transport this signature offshore into the basins (Figure 8). Indeed, the stations sampled along 705 the flow path of the boundary currents have higher concentrations of pAl, pFe and pV than those located in 706 the central basins. The distributions of particulate Mn, in contrast, are governed by water column Mn^{+2} 707 oxidation, showing maximum concentrations between $\sim 100-300$ m across the CB and BB, presumably 708 related to strengthened activity of Mn-oxidizing bacteria -fueled by excess of dissolved Mn inputs advected 709 from shallow shelves- and photoinhibition relief in subsurface waters (Figure 8). The occurrence of Mn 710 oxides (non-lithogenic component) is greatly reduced in the LS, where the lithogenic sources dominate bulk 711 particulate distributions of Al, Fe, V, and also Mn, despite the low concentrations measured in this basin, 712 distant from continental margins. Benthic nepheloid layers are also abundant in the CAO, where above 713 mentioned particulate elements are enriched in the deepest samples due to sediment resuspension and/or 714 redox mobilization (Figure 8). The distributions and cycling of pP, POC and pN are tightly linked to the 715 strong productivity gradient present in the CAO, with the lowest values registered in the oligotrophic CB, 716 progressively increasing their concentrations towards the shallow CAA, BB and the LS, where large 717 phytoplankton blooms have been registered. Accordingly, the C:N:P stoichiometry in the upper 100 m 718 closely reflects that of phytoplankton, although higher C:N ratios have been observed in the land-dominated

- 719 CAA where terrigenous-derived inputs are likely increased. In the deep water column (> 100 m), where
- 720 remineralization is prevalent, C:N:P ratios deviate from canonical ratios (Figure 8).



721

Figure 8 Conceptual scheme of the dominant phases (biogenic, lithogenic and non-lithogenic) and key processes controlling the distributions of particulate Al, V, Fe, Mn, P as well as particulate organic carbon and nitrogen (POC and pN) across the Canadian Arctic Ocean (CAO). CB: Canada Basin, CAA: Canadian Artic Archipelago, BB: Baffin Bay, LS: Labrador Sea, wBSW: winter Bering Sea Water, ACBC: Arctic Circumpolar Boundary Current, AW: Arctic Water, WGIW: West Greenland Intermediate Water, LSW: Labrador Sea Water, DSOW: Denmark Strait Overflow Water. The size of the label fonts and symbols represent the relative abundance of particulate trace element and the importance of the described processes.

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736 Research Data

The particulate Arctic data reported in this study is available in the Research Data document (ElectronicAnnex).

739 Supporting Information

Tables S1 & S2, Figures S1-S6 and Supplementary Methods are included in the supporting information.

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