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1 Mineralogical, geochemical and magnetic signatures of surface sediments from the

2 Canadian Beaufort Shelf and Amundsen Gulf (Canadian Arctic)

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18 Highlights

- 19 1. Tracking sediment provenance in the Canadian Beaufort Shelf and Amundsen Gulf
- 20 2. Coupling grain size, mineralogical, geochemical and magnetic proxies in the western Canadian Arctic
- 21 3. Dolomite–K-feldspar and Ca-Mg characterize southwestern Banks Island
- 22 4. Phyllosilicates-magnetite-Fe-oxides and Al-K-Ti-Fe-Cr-V-Zn-P characterize the Canadian Beaufort
- 23 Shelf
- 24 5. The Canadian Beaufort Shelf and Amundsen Gulf are separated into four sedimentological provinces
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27 Abstract

28 Mineralogical, geochemical, magnetic, and siliciclastic grain-size signatures of 34 surface 29 sediment samples from the Mackenzie-Beaufort Sea Slope and Amundsen Gulf were studied in order to 30 better constrain the redox status, detrital particle provenance, and sediment dynamics in the western 31 Canadian Arctic. Redox-sensitive elements (Mn, Fe, V, Cr, Zn) indicate that modern sedimentary deposition within the Mackenzie-Beaufort Sea Slope and Amundsen Gulf took place under oxic bottom-32 33 water conditions, with more turbulent mixing conditions and thus a well-oxygenated water column 34 prevailing within the Amundsen Gulf. The analytical data obtained, combined with multivariate 35 statistical (notably, principal component and fuzzy c-means clustering analyses) and spatial analyses, 36 allowed the division of the study area into four provinces with distinct sedimentary compositions: (1) the Mackenzie Trough-Canadian Beaufort Shelf with high phyllosilicate-Fe oxide-magnetite and Al-K-Ti-37 Fe-Cr-V-Zn-P contents; (2) Southwestern Banks Island, characterized by high dolomite-K-feldspar and 38 Ca-Mg-LOI contents; (3) the Central Amundsen Gulf, a transitional zone typified by intermediate 39 40 phyllosilicate-magnetite-K-feldspar-dolomite and Al-K-Ti-Fe-Mn-V-Zn-Sr-Ca-Mg-LOI contents; and 41 (4) mud volcanoes on the Canadian Beaufort Shelf distinguished by poorly sorted coarse-silt with high 42 quartz-plagioclase-authigenic carbonate and Si-Zr contents, as well as high magnetic susceptibility. Our 43 results also confirm that the present-day sedimentary dynamics on the Canadian Beaufort Shelf is mainly 44 controlled by sediment supply from the Mackenzie River. Overall, these insights provide a basis for 45 future studies using mineralogical, geochemical, and magnetic signatures of Canadian Arctic sediments 46 in order to reconstruct past variations in sediment inputs and transport pathways related to late 47 Quaternary climate and oceanographic changes.

Keywords: mineralogy; elemental geochemistry; magnetic properties; surface sediment; sediment
provenance; redox condition; Canadian Beaufort Shelf; Amundsen Gulf.

2

50 **1. Introduction**

51 Sedimentation in the Arctic Ocean is characterized by high terrigenous input from the 52 surrounding continents with different petrographic signatures (Harrison et al., 2011). These sediments 53 are delivered into the Arctic Ocean mainly as suspended particulate matter and bed loads from several 54 large river systems (notably, the Mackenzie, Kolyma, Lena, Ob, Yenisei, Pechora, and Severnaya Dvina; 55 Holmes et al., 2002; Wagner et al., 2011) and from coastal erosion, and then dispersed by ocean currents 56 (summarized in Stein, 2008). Furthermore, in shallow margins, suspended terrigenous particles can also 57 be incorporated in sea ice during its formation and then be transported via ocean currents over long 58 distances throughout the Arctic Ocean, to finally settle far from their source of origin (e.g., Bischof et al., 59 1996; Darby et al., 2012, 2006).

Taking this into account, a number of studies have characterized the mineralogical and 60 geochemical composition of the detrital sediments over the continental shelf from the Eurasian Basin 61 (e.g., Vogt, 1997; Schoster et al., 2000; Viscosi-Shirley et al., 2003; Stein, 2008; Bazhenova, 2012), 62 Chukchi Sea-Bering Strait (e.g., Asahara et al., 2012; Linsen et al., 2014), and Chukchi-Alaskan margin 63 (e.g., Naidu et al., 1982; Naidu and Mowatt, 1983; Ortiz et al., 2009; Darby et al., 2012) to decipher: (1) 64 variations in detrital particle provenance, (2) climate and atmospheric circulation changes in the source 65 areas on adjacent landmasses, and (3) changes in sediment propagation and ocean-current pathways. 66 67 However, few studies provide a general view of the surface detrital provenances and sediment-dispersal patterns within the Mackenzie-Beaufort Sea Slope and Amundsen Gulf (e.g., Naidu et al., 1971; 68 Bornhold et al., 1975; Pelletier, 1975; Davidson et al., 1988; Hill et al., 1991; Darby et al., 2011; Vonk et 69 70 al., 2015) compared to other Arctic continental shelf regions. To our knowledge, no general 71 mineralogical and geochemical distributions of the Canadian Beaufort Shelf and Amundsen Gulf are 72 available today. Such studies may provide a baseline to better interpret, in terms of sediment dynamics 73 and climate change, the mineralogical and geochemical signatures preserved in the southern Beaufort Sea sedimentary records, which may then help to place current western Arctic climate change (e.g.,
Kwok et al., 2009) into its broader context.

76 In this study, a multi-proxy analysis was carried out on the bulk detrital fraction of surface 77 sediment samples from the Mackenzie-Beaufort Sea Slope and Amundsen Gulf in order to: (1) 78 characterize the spatial distribution patterns of siliciclastic grain size, magnetic properties, bulk minerals, 79 and elemental geochemistry in surface sediments; (2) identify different sedimentary provinces, source 80 areas, and transport pathways of terrigenous material; and (3) better constrain modern sediment 81 dynamics within the western Canadian Arctic. Overall, this study provides a unique opportunity to 82 compare mineralogical, geochemical, magnetic, and siliciclastic grain-size signatures within the 83 Mackenzie-Beaufort Sea Slope and Amundsen Gulf area.

84

85 **2. Environmental setting**

86 **2.1. Regional morphology**

The Canadian Beaufort Shelf is a shallow platform located along the northwestern Canadian 87 coast in the southeastern Beaufort Sea (western Arctic Ocean; Figure 1). It is bordered to the west by the 88 Mackenzie Trough and to the east by the Amundsen Gulf. This shelf is cut by several partially infilled 89 cross-shelf channels: the Ikit Trough, the Kugmallit Channel, and the Niglik Channel (Blasco et al., 90 91 2013). Moreover, several hundreds of conical mounds, locally referred to as pingo-like-features (Shearer 92 et al., 1971), occur across the Canadian Beaufort Shelf (Blasco et al., 2013). Based on seismic reflection 93 profiles, water column acoustic anomalies (Paull et al., 2007; Blasco et al., 2013; Saint-Ange et al., 94 2014; Paull et al., 2015) together with geochemical composition of pore waters, gas and sediments (Paull et al., 2015), these conical features are now regarded as mud volcanoes. On the other hand, the 95 96 Amundsen Gulf is a large channel (400-km-long, 200-km-wide, average water depth of 300 m; Stokes et al., 2006) that connects the southeastern Beaufort Sea to the Canadian Arctic Archipelago (Figure 1). It
is bordered by the Banks Island Shelf to the north and by the Mackenzie Shelf to the southwest.

99 The Canadian Beaufort Shelf and Amundsen Gulf are nearly completely covered by sea ice 100 (pack-ice and landfast ice) from September/October to May (Barber and Hanesiak, 2004; Galley et al., 101 2008), with great annual and interannual variability (e.g., Schell et al., 2008; Bringué and Rochon, 102 2012). In summer, freshet from the Mackenzie River, wind forcing, and rising air temperatures result in 103 ice-free conditions over the shelf by late July and over the slope in August (O'Brien et al., 2006). In 104 addition, in the middle of landfast ice from Amundsen Gulf, the ice-free zone forms part of the Cape 105 Bathurst Polynya which develops during winter at approximately the same location under the action of 106 winds, currents and upwellings of warmer water (Arrigo and van Dijken, 2004). In this ice-free area, 107 stronger westerly to northwesterly winds induce turbulent mixing of the water column (e.g., Magen, 108 2007; Forest et al., 2008; Tremblay et al., 2014).

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110 **2.2. Surrounding geology**

The Mackenzie River drainage basin covers a large part of western Canada ($\sim 1.8 \times 10^6$ km²; 111 112 Carson et al., 1998; Hill et al., 2001). Three main geological units characterize the Mackenzie River 113 basin (Millot et al., 2003); (1) the North American Cordillera (including the Rocky and the Mackenzie 114 Mountains) in the western part, distinguished by volcanic and immature volcano-clastic sediments in the 115 western Canadian orogenic belt (Stikine terrane) and carbonates and slates in the Mackenzie Mountains; 116 (2) the Interior Platform (lowlands), composed of marine and non-marine sedimentary rocks (Cambrian 117 to Cretaceous limestones, shales, and sandstones); and (3) the Canadian Shield in the eastern part, 118 typified by old silicate rocks (Archean granites and gneisses) from the Slave Province (Padgham and 119 Fyson, 1992). In turn, Banks Island is composed of Cretaceous shale and sandstone, Upper Devonian sandstone and shale, and Tertiary–Quaternary glacial deposits, which are rich in dolomite clasts as well
as quartz and feldspar grains (Bischof et al., 1996; Bischof and Darby, 1999).

122

123 **2.3. Sedimentation**

124 The Mackenzie River is the fourth-largest (after the Yenisei, Lena, and Ob rivers) Arctic river in 125 terms of freshwater discharge (~420 km³/yr; Wagner et al., 2011), but the first in terms of sediment 126 discharge (~127 Mt/yr; Carson et al., 1998). This large suspended sediment discharge of the Mackenzie 127 River forms a large sediment plume (generally 2–3 m thick; Hill et al., 1991) on the Canadian Beaufort 128 Shelf (Figure 1). The transport of suspended sediments within this plume is affected by the ice cover, 129 winds, and currents (Carmack and Macdonald, 2002). In winter, the Mackenzie River discharges are 130 trapped on the inner shelf by the stamukhi (a field of ice fragments), which acts as an inverted dam and 131 causes the formation of the "floating freshwater" lake Herlinveaux (Macdonald et al., 1995). In summer, 132 the plume's position is greatly affected by prevailing winds, with winds from the northwest pushing the 133 plume along Tuktovaktuk Peninsula (Giovando and Herlinveaux, 1981), and winds from the southeast 134 pushing the plume westward, beyond the Mackenzie Trough (MacNeil and Garrett, 1975). In addition, 135 although coastal erosion is an important local sediment supply near the shoreline, its estimated 136 contribution (~7 Mt/yr) is dwarfed by that of the Mackenzie River (Carmack and Macdonald, 2002). 137 Furthermore, the suspended particulate matter supply to the Amundsen Gulf is much smaller than to the 138 Canadian Beaufort shelf as no large rivers discharge into the Gulf (Macdonald et al., 1998). Indeed, 139 small rivers located to the east Mackenzie River, such as the Anderson, Horton and Hornaday Rivers (Figure 1), have a weak mean annual freshwater discharge ($<146 \text{ m}^3/\text{s}$; R-ArcticNet database: 140 141 http://www.r-arcticnet.sr.unh.edu/v4.0/index.html; Lammers et al., 2001), and therefore, contribute 142 weakly to the sedimentation in the eastern Mackenzie Shelf and Amundsen Gulf. Consequently, modern 143 sedimentation rates are high within the Mackenzie Trough (~40-320 cm/ka; Macdonald et al., 1998; Richerol et al., 2008a; Durantou et al., 2012) and on the nearby continental shelf and slope (~100–200
cm/ka; Barletta et al., 2008; Scott et al., 2009; Bringué and Rochon, 2012). To the east, these modern
sedimentation rates decrease towards Amundsen Gulf (<80 cm/ka; Macdonald et al., 1998; Schell et al.,
2008), where sediment hardly accumulates at all (Hill et al., 1991).

On the Canadian Beaufort shelf, most of the surficial seabed sediments are predominantly composed of Holocene marine olive-grey silts and clays (e.g., Pelletier, 1975; Hill et al., 1991; Barletta et al., 2008; Scott et al., 2009). Surface sediments from the Amundsen Gulf are composed of a relatively thin layer of olive-grey silty clay overlying a diamicton of brownish red color with abundant pebbles and cobbles (Bennett et al., 2008; Schell et al., 2008; Scott et al., 2009).

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154 **2.4. Oceanic circulation**

155 Oceanic circulation in the southeastern Beaufort Sea is dominated by the anticyclonic Beaufort 156 Gyre (BG), which pushes both surface currents and sea-ice westward at the shelf break (Figure 1). 157 Conversely, closer to shore around the 50-m isobath, the Beaufort Undercurrent transports both Pacific 158 and Atlantic waters eastwards along the continental margin and into Amundsen Gulf (e.g., Aagaard, 159 1984; Pickart, 2004; Bringué and Rochon, 2012; Durantou et al., 2012). In general, surface waters 160 influenced by the anticyclonic BG enter Amundsen Gulf near southwestern Banks Island and exit near 161 Cape Bathurst (Lanos, 2009). At a regional level, these surface circulation regimes are mainly controlled 162 by changes in the phase of large-scale atmospheric patterns such as the Arctic Oscillation (AO; Darby et 163 al., 2001; Macdonald et al., 2005) and the Pacific Decadal Oscillation (PDO; Overland et al., 1999; 164 Durantou et al., 2012), which are both significant natural patterns in global climate variability.

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166 **3. Material and methods**

167 **3.1. Samples**

168 A total of 34 surface sediment samples were collected at different depths in the Canadian 169 Beaufort Shelf and Amundsen Gulf on board the Canadian Coast Guard Ship (CCGS) Amundsen during 170 the CASES (Canadian Arctic Shelf Exchange Study) 2004 expedition (Rochon and onboard participants, 171 2004). The sampling was performed using a box core sampler (0.5 m x 0.5 m x 0.5 m) wherein the 172 uppermost 5 mm of sediment was recovered in order to collect only the sediment-water interface 173 (Richerol et al., 2008b). Based on the regional morphology, mud volcanoes distribution (Blasco et al., 174 2013), and the influence of the Mackenzie River plume (Richerol et al., 2008b; Scott et al., 2008), the 175 surface sediment samples were divided into four main geographical areas (Figure 1): (1) Mackenzie 176 Trough-Canadian Beaufort Shelf, (2) mud volcanoes, (3) central Amundsen Gulf, and (4) southwestern 177 Banks Island.

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179 **3.2. Analytical procedure**

180 3.2.1. Grain-size distribution

Sediment grain-size analyses were performed on the detrital fraction of the sediment using a 181 182 Beckman Coulter LS13320 laser diffraction grain-size analyzer, which has a detection range of 0.04– 183 2000 µm. Samples were deflocculated by successive washing with distilled water after the removal of 184 organic matter and biogenic carbonate of the sediments with 10 mL of hydrogen peroxide (30 % H_2O_2) 185 and 10 mL of hydrochloric acid (0.5M HCl), respectively. Biogenic silica was not removed as it appeared to be negligible (likely less than 1%, as suggested by its no detection in the bulk sediment XRD 186 187 diffractograms). The grain-size distribution and statistical parameters (e.g., mean, sorting) were 188 calculated using the moment methods in logarithmic (phi - ϕ) scale and the GRADISTAT software 189 (Blott and Pye, 2001). Furthermore, the end-member modelling algorithm (EMMA) developed by 190 Weltje (1997) and adapted by Dietze et al. (2012) was subsequently applied to the grain-size data in 191 order to extract meaningful end-member (EM) grain-size distributions and estimate their proportional 192 contribution to the sediments. The cumulative explained variance (r^2) was calculated to assess the 193 minimum number of EMs needed for a good estimate of our grain-size data (Weltje, 1997; Dietze et al., 194 2012). In general, grain-size distribution and end-member modelling analysis can be used to investigate 195 the sedimentary transfer regiweme because sediment grain-size distribution (primarily driven by 196 sedimentary processes) reflects transport conditions (e.g., Montero-Serrano et al., 2009, 2010a; Simon et 197 al., 2012; Dietze et al., 2012).

198 3.2.2. Bulk magnetic properties

Low-field magnetic susceptibility (k_{LF}) was measured on bulk sediment samples using a 199 200 Bartington MS2E. The k_{LF} values primarily reflect changes in the ferrimagnetic concentration (e.g., 201 magnetite or titanomagnetite), but they are also sensitive to magnetic grain-size variations (Dunlop and 202 Özdemir, 1997). In order to explorer the possible presence of ultrafine superparamagnetic (SP) particles, 203 Frequency-dependent magnetic susceptibility was measured in some bulk sediments samples, at low-204 (0.465 kHz; k_{lf}) and high- (4.65 kHz; k_{hf}) frequencies, on a Bartington Susceptibility Meter (model MS2B) with a dual-frequency sensor (Dearing, 1999). The per sample measuring time is 10s and data 205 output is in SI units (10⁻⁵ SI). Each measurement was repeated at least five times and the readings were 206 207 averaged. The measurement of hysteresis loops and derived properties, including saturation remanence (M_r), saturation magnetization (M_s), bulk coercive force (H_c), and remanent coercive force (H_{cr}) were 208 209 determined using an alternating gradient force magnetometer (AGM) MicroMag 2900 from Princeton 210 Measurements Corporation. The M_{rs}/M_s and H_{cr}/H_c ratios can be used as grain-size proxies (the so-called 211 Day plot) as well as to identify the magnetic domain state when the principal remanence-carrier mineral 212 is magnetite (Day et al., 1977; Dunlop, 2002; Stoner and St-Onge, 2007).

213 3.2.3. Bulk sediment mineralogy and elemental geochemistry

214 Before the bulk mineralogical and geochemical analysis, the sediment samples were rinsed five

times with distilled water after the removal of organic matter fraction with 10 mL of hydrogen peroxide

(30 % H₂O₂). Next, sediment samples were ground with a McCrone micronizing mill using 5 mL of ethanol and grinding times of 5–10 min to obtain a homogeneous powder. The slurry was oven-dried overnight at about 60°C and then slightly homogenized with an agate mortar to prevent any agglomeration of finer particles during drying. Aliquots of these sediment samples were used for bulk mineralogical and geochemical analysis.

221 Bulk mineral associations were studied by X-ray diffraction (XRD). The random powder samples 222 were side-loaded into the holders and analysed on a PANalytical X'Pert Powder diffractometer. This instrument is fitted with a copper tube (Cu K-alpha = 1.54178 Å), operating at 45 kV and 40 mA and a 223 post-diffraction graphite monochromator. Samples were scanned from 5° to 65° two-theta in steps of 224 225 0.020° two-theta and a counting time of 2 seconds per step. For the semi-quantification of the major 226 mineralogical components, bulk sediment XRD scans were processed in the software package X'Pert 227 High-Score Plus (PANalytical) using the Rietveld full-pattern fitting method (e.g., Young, 1993; 228 Grunsky et al., 2013). This method permits the semi-quantification of whole-sediment mineralogy with a 229 precision of 5–10% for phyllosilicates and 5% for non-phyllosilicates minerals. The quality of the 230 Rietveld fitting procedure was evaluated for the R-profile and goodness-of-fit (GOF). The R-profile 231 quantifies the difference between the observed and calculated patterns, whereas the GOF is the ratio 232 between the R-weighted profile (RWP: best least-squares fit between observed and calculated patterns) 233 and R-expected theoretical (Rexp; best possible value for the residual). An R-values profile between 20-30 % and a GOF of fewer than 3 are typically adequate in the Rietveld refinement of geological samples 234 235 (e.g., Young, 1993). The major mineralogical components quantified by this technique are: quartz, 236 potassium (K) feldspar (microcline + orthoclase), plagioclase feldspar (albite + anorthite), amphibole 237 (hornblende), pyroxene (augite), magnetite, Fe-oxides (hematite + goethite), dolomite, and 238 phyllosilicates (biotite, muscovite, illite, chlorite, and kaolinite).

239	A total of 14 elements (Al, Si, K, Mg, Ca, Ti, Mn, Fe, P, Sr, V, Cr, Zn, and Zr) were analysed by
240	energy dispersive X-ray fluorescence (EDXRF) spectrometry using a PANalytical Epsilon 3-XL. Before
241	EDXRF analysis, loss on ignition (LOI) was determined gravimetrically by heating the dried samples up
242	to 950°C for two hours. Subsequently, samples were treated by borate fusion in an automated fusion
243	furnace (CLAISSE® M4 Fluxer). Samples weighing ~0.6 g were mixed with ~6 g of lithium borate flux
244	(CLAISSE, pure, 49.75% Li ₂ B ₄ O ₇ , 49.75% LiBO ₂ , 0.5% LiBr). The mixtures were melted in Pt-Au
245	crucibles (95% Pt, 5% Au), and after fusion the melts were cast to flat disks (diameter: 32 mm; height: 3
246	mm) in Pt-Au moulds. Acquired XRF spectra were processed with the standardless Omnian software
247	package (PANalytical). The resulting data are expressed as weight percent (wt.% ; Al, Si, K, Mg, Ca, Ti,
248	Mn, Fe, P) and micrograms per gram (µg/g; V, Cr, Zn, Sr, Zr). Procedural blanks always accounted for
249	less than 1% of the lowest concentration measured in the sediment samples. Analytical accuracy and
250	precision were found to be better than 1-5% for major elements and 5-10% for the other elements, as
251	checked by an international standard (USGS SDC-1) and analysis of replicate samples. Because Al and
252	Si are associated to clay minerals, aluminosilicates and quartz, and Ca is associated to carbonates, the
253	ternary plot Al-Si-Ca (expressed as oxides) was used here in order to obtain a general geochemical
254	classification of the sediments (e.g., Brumsack, 1989; Meinhardt et al., 2014). In addition, to compare
255	the relative enrichment of redox-sensitive elements (Mn, Fe, V, Cr, Zn), we calculated enrichment
256	factors (EF) by comparing Al-normalized metal concentrations to those of average shale (Wedepohl,
257	1991): $X_{EF} = [(X/Al)_{sample} / (X/Al)_{average shale}]$. In practical terms, EF > 3 represents a detectable authigenic
258	enrichment of an element over average shale concentrations, whereas $EF > 10$ represents a moderate to
259	strong degree of authigenic enrichment (e.g., Tribovillard et al., 2006).

In addition, in order to identify the presence of authigenic minerals (such as, carbonate, iron oxides, and greigite) within the mud volcanoes province, some bulk sediments and white crusts observed in the samples were analyzed using a JEOL 6460LV scanning electron microscope equipped with an Energy Dispersive X-ray Spectrometer (SEM-EDS). The image and EDS analyses were obtained with a
backscatter detector, an accelerating voltage of 20 kV and a spot size of 60 (probe diameter).

265

266 **3.3. Statistical and spatial approach**

267 The mineralogical and geochemical data are of a compositional nature, that is, they are vectors of 268 non-negative values subjected to a constant-sum constraint (usually 100%). This implies that relevant 269 information is contained in the relative magnitudes, so statistical analysis must focus on the ratios 270 between components (Aitchison, 1986). Under this framework, a principal component analysis (PCA) 271 was performed on the mineralogical and elemental geochemical dataset with the goal of finding 272 elemental and mineralogical associations with similar relative variation patterns that may be interpreted 273 from a palaeoenvironmental standpoint (e.g., von Evnatten et al., 2003; Montero-Serrano et al., 2010b, 274 2015; von Eynatten et al., 2016). Likewise, a fuzzy c-means clustering analysis was performed using the 275 mineralogical dataset with the goal of ascertaining whether the differences observed between each 276 sedimentological province are statistically valid. The results from the fuzzy c-means clustering are 277 visualized by a silhouette plot (Kaufman and Rousseeuw, 2009), where each sediment sample is 278 represented by a bar (silhouette width) that ranges from 0 (no similarity) to 1 (identical). Thus, the 279 silhouette plot allows a visualization of the quality of the clustering and the distinctiveness of the 280 sediment samples (Borcard et al., 2011). Prior to all multivariate analyses, a log-centred (clr) transform 281 was applied to the data (Aitchison, 1990). The clr transform is derived by dividing each variable (e.g., 282 mineral percentage, element concentration) by the geometric mean of the composition of the individual 283 observations and then taking the logarithm. This operation removes statistical constraints on 284 compositional variables, such as the constant-unit sum, and allows the valid application of classical 285 (Euclidean) statistical methods to compositional data (Aitchison, 1986; 1990). All statistical calculations 286 were conducted with "R" software using the packages "StatDA" (Reimann et al., 2008), "compositions" (van den Boogaart and Tolosana-Delgado, 2008), "vegan" (Oksanen et al., 2015), and "cluster" (Maechler et al., 2015). Finally, the scores from the first two principal components of the log-centred data were used to produce interpolated compositional maps in ArcGIS® software. The interpolated maps were generated using the Spline with Barriers algorithm available in ArcGIS® Spatial Analyst Tools. This method of interpolation produced a smooth surface with values in the range of the scores of data points using a minimum curvature spline technique (Childs, 2004). However, the interpolated surfaces closest to the borders are biased by this method and should be interpreted with caution.

All analytical data presented are available in the supplementary material.

295

4. Results and Interpretations

297 **4.1. Grain-size distribution**

298 The mean sediment grain size (phi scale) ranges from 8.31 (clay) to 6.79 (fine silts), with 299 minimum phi values within the southwestern Banks Island province (Figure 2A). Interestingly, although 300 all sediment samples are poorly sorted (values of $1.25 < \sigma_{\phi} < 1.75$; Figure 2B) in the study area, relatively 301 higher sorting (values up to 1.75) is found in some samples from the mud volcanoes area in the Canadian 302 Beaufort Shelf (samples 403, 609, 709, 711, 712, 805) as well as in the southwestern Banks Island 303 (samples 2015 and 415) (Figure 2B). In correspondence with this result, the end-member modelling 304 analysis (EMMA) produced a three-EM model to explain more than 91% of the total variance (Figure 305 3A). The sediment samples are characterized by three grain classes with different grain-size distribution 306 (Figure 3B-C): (1) the EM1 end-member (mean $\sim 8.8 \ \phi$; bimodal distribution) is associated to the clay to 307 very fine silt fraction and characterizes many samples in the shallow Canadian Beaufort Shelf; (2) the 308 EM2 end-member (mean ~ 6.3 ϕ ; bimodal distribution) is associated to the fine silt fraction, which is 309 most of the sediments studied; and (3) the EM3 end-members (mean ~5.9 ϕ ; trimodal distribution) correspond to the medium to fine silt fraction of samples 403, 805, and 709 from the mud volcanoes in
the Canadian Beaufort Shelf and 415, 312, and 315 from the southwestern Banks Island.

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313 **4.2. Magnetic properties**

314 Hysteresis loops of representative samples are shown in Figure 4A. All hysteresis loops display 315 saturation fields (<250 mT) and shapes suggesting the assemblage of magnetic grains is mainly 316 dominated by magnetite. Similarly, the M_{rs}/M_s - H_{cr}/H_c crossplots or Day plot (Figure 4B) illustrate that 317 most of the magnetic grains within the sediment samples are composed of pseudo-single domain (PSD) 318 magnetite (Dunlop, 2002). However, some poorly sorted and coarser samples (106 and 109 from the 319 central Amundsen Gulf; 609 and 709 from the mud volcanoes in the Canadian Beaufort Shelf; Figure 320 3B) show a scattered distribution on the Day plot (Figure 4B), most probably reflecting the presence of coarser magnetite grains (e.g., Lisé-Pronovost et al., 2009; Brachfeld et al., 2009). Magnetic 321 susceptibility values range between 4.4 and 35.7 ($x10^{-5}$ SI units; Figure 5A), with maximum values 322 323 recorded in samples from the southwestern Banks Island coast (samples 215 and 415), central Amundsen 324 Gulf (sample 115) and mud volcanoes in the Canadian Beaufort Shelf (samples 400, 403, 609, 709 and 325 803). Differences in the frequency-dependent susceptibility for these last sediment samples are 326 negligible ($k_{lf}/k_{hf} \sim 1$; Table S1), suggesting low to very low contents of superparamagnetic grains. The 327 magnetic susceptibility, M_{rs}/M_s and H_{cr}/H_c ratios show similar distribution patterns (Figure 5), most 328 likely suggesting that magnetic susceptibility changes are driven by magnetic grain-size variations. 329 Interestingly, the southwestern Banks Island coast and Amundsen Gulf grains have slightly lower ratios 330 of H_{cr}/H_c and higher ratios of M_{rs}/M_s than the Mackenzie Trough-Canadian Beaufort Shelf samples 331 (Figure 5B-C and 12F), possibly indicating a slighter finer magnetic grain size. Similar results in 332 magnetic mineralogy have also been reported in the sedimentary records from the Mackenzie Trough-333 Canadian Beaufort Shelf area (e.g., Barletta et al., 2010; Barris, 2012).

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335 **4.3. Bulk mineralogy composition**

336 The Mackenzie Trough-Canadian Beaufort Shelf and Amundsen Gulf bulk mineralogy (Figure 337 S1 in the auxiliary material) is dominated by quartz (28-64%), phyllosilicates (15-48%), dolomite (3-338 35%), Na-plagioclase (4-11%), and K-feldspar (3-10%), and by lower proportions of Fe-oxides (0.2-10%)1.5%), calcite (<1%), magnetite (<0.7%), and pyroxene (<1.0%). Amorphous silica (e.g., diatoms) was 339 340 not detected in the XRD diffractograms in the studied samples due to its low content (likely <1%). 341 Quartz, K-feldspar, plagioclase, phylosilicates and dolomite represented more than 91% of the overall 342 mineral concentration in the sediment samples. In order to reduce dimensionality in the data and identify 343 mineral associations, a principal component analysis (PCA) was conducted (Figure 6A). This analysis 344 indicates that PC-1 (65.37% of the total variance) is positively associated with dolomite and negatively 345 associated with phyllosilicates, Fe-oxides, and magnetite, whereas PC-2 (19.54% of the total variance) is 346 positively associated with plagioclase and quartz and negatively associated with K-feldspar as well as 347 dolomite, magnetite, and Fe-oxides (Figure 6B). The spatial distributions of the PC-1 and PC-2 348 mineralogical scores (Figure 7) reveal that the southwestern Banks Island and central Amundsen Gulf 349 provinces are relatively enriched in dolomite and K-feldspar, whereas the Mackenzie Trough-Canadian 350 Beaufort Shelf is characterized by higher contents in phyllosilicates, Fe-oxides, and magnetite (Figure 351 7A). Intermediate PC-1 scores and high PC-2 scores (up to 1.4) are observed in the coarse multimodal 352 samples from the mud volcanoes province and reflect intermediate and higher concentrations of dolomite 353 and plagioclase-quartz, respectively (Figure 7B).

354

355 **4.3. Elemental geochemistry**

The major element composition (Figure S2 in the auxiliary material) in the Mackenzie Trough-Canadian Beaufort Shelf and Amundsen Gulf sediments is dominated by Si (25–37 wt.%), Al (4–10 wt.%), Ti (0.2–0.5 wt.%), Ca (1–8 wt.%), Fe (2–6 wt.%), K (1–3 wt.%), Mg (0.6–5 wt.%), P (0.2–0.8 wt.%), and Mn (0.02–2.2 wt.%), while the minor and trace elements are dominated by V (120–325 $\mu g/g$), Zr (134–227 $\mu g/g$), Sr (107–209 $\mu g/g$), Zn (84–205 $\mu g/g$), and Cr (81–163 $\mu g/g$). LOI values range from 3 to 17.5 wt.%.

362 The ternary plot Al₂O₃-SiO₂-CaO (Figure 8A) illustrates that the sediments from the Mackenzie 363 Trough-Canadian Beaufort Shelf are dominantly composed of detrital material similar to average shale, 364 whereas the central Amundsen Gulf and southwestern Banks Island sediments show higher detrital 365 carbonate contents (dolomite) and plot along the mixing line from average shale to the detrital carbonate 366 end-member. Some sediment samples from the mud volcanoes (samples 403, 609 and 709) and central 367 Amundsen Gulf (sample 106) are enriched in quartz (SiO₂ >45%; Figure 8A). Furthermore, Fe/Al and 368 Mn/Al display a high positive linear correlation (r=0.75; Figure 8B), indicating a geochemical 369 relationship most likely similar to Fe–Mn oxyhydroxide phases. The enrichment factors of redox-370 sensitive elements (V, Cr, Zn) reveal modest authigenic enrichment (1 to 3) in all sedimentological 371 provinces compared to average shale values (Figure 9). Fe shows no detectable authigenic enrichment in 372 any sediment samples, whereas Mn has moderate-to-strong authigenic enrichment (EF > 5) in the central 373 Amundsen Gulf and southwestern Banks Island and modest enrichment (EF < 3) in the Mackenzie 374 Trough-Canadian Beaufort Shelf.

Principal component analyses based on these major, minor, and trace elements (Figure 10A) reveal that the PC-1 scores (71.05% of the total variance) are positively associated with Ca-Mg-LOI and negatively associated with Al-K-Ti-Fe-Cr-V-Zn-P, whereas the PC-2 scores (11.27% of the total variance) are positively associated with Si-Zr-Cr-Ca-P and negatively associated with K-Fe-V-Mg-LOI (Figure 10B). Importantly, Mn was omitted in the PCA because of the large influence on the 380 geochemical variability. The Mn surface distribution show higher concentrations in central Amundsen 381 Gulf and southwestern Banks Island sediments compared to the Canadian Beaufort Shelf (Figure 11A). 382 Moreover, the spatial distributions of the PC-1 and PC-2 elemental geochemical scores (Figure 11B-C) 383 reveal similar trends as those observed in the bulk minerals (Figure 7), with large positive PC-1 scores 384 (Ca-Mg-LOI) in the southwestern Banks Island and central Amundsen Gulf provinces and large negative 385 PC-1 scores (Al-K-Ti-Fe-Cr-V-Zn-P) in the Mackenzie Trough-Canadian Beaufort Shelf province. Some 386 coarse multimodal samples from the mud volcanoes province are characterized by positive PC-2 scores, 387 which are associated with Si-Zr-Cr and Ca-P (Figure 11B).

388 SEM-EDS analysis performed on white crusts observed in mud volcanoes sediments (e.g., station 389 609; Figure S3 in the auxiliary material) reveal Ca-Mg peaks, suggesting that these white crusts most 390 probably represent authigenic carbonate minerals (notably, low Mg-calcite and/or dolomite). In addition, 391 authigenic iron sulfides (such as, pyrite and greigite) were not observed in the bulk sediment particles 392 during the SEM-EDS analysis.

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394

395 4.4. Relationship between grain size, bulk mineralogy, elemental geochemistry, and magnetic 396 properties

In order to explore the relationship among the grain size and PC scores from the bulk mineralogy and elemental geochemistry, a bivariate correlation was conducted (Figure 12). Note that sediment samples from the mud volcanoes province were not plotted because they are formed by different sedimentary processes. PC-1 scores derived from bulk mineralogy and elemental geochemistry exhibit a good-to-modest negative correlation with the mean grain size on the phi-scale, respectively (Figure 12A-B). This correlation suggests that variations in the mineralogical and geochemical signatures of surface sediments from the Mackenzie Trough-Canadian Beaufort Shelf and Amundsen Gulf are not only 404 dominated by the relative contribution of the different sediment sources, but are also a function of grain-405 size sorting. In fact, a west-east trend in sediment composition and grain size may be observed in Figures 406 2A, 3A, 4C, 5c and 12A-C. In the Mackenzie Trough-Canadian Beaufort Shelf sediments, the 407 phyllosilicate-Fe-oxide-magnetite (negative PC-1 scores from XRD) and Al-K-Ti-Fe-Cr-V-Zn-P 408 (negative PC-1 from XRF) contents are highest in the clay to very fine silt fraction (7.5 $\leq \phi \leq 8.5$), whereas 409 dolomite-K-feldspar (positive PC-1 scores from XRD) and Ca-Mg-LOI (positive PC-1 scores from 410 XRF) contents are highest in the very fine to fine silt fraction ($6.5 < \phi < 7.5$) of the southwestern Banks 411 Island sediments. Interestingly, there is a weak negative correlation with the mean grain size in phi units 412 for the magnetic susceptibility (k_{lf}) (Figure 12C), suggesting that the k_{lf} variability can only partly be 413 explained by the grain-size sorting. In general, the lowest magnetic susceptibility (k_{lf}) is recorded in the 414 clay to very fine silt fraction, where the magnetite and Fe-oxide contents are highest (Figure 6A), with an 415 inverse trend observed in the fine silt fraction.

To explore the relationship among the magnetic susceptibility (k_{if}) and the mineralogical and geochemical signatures in the surface sediments, a bivariate correlation was also performed. A good positive correlation is observed between all detrital proxies used here (Figure 12D-F), suggesting that: (1) the minerals and chemical composition of detrital particles are highly intercorrelated, and thus, phyllosilicates–Fe-oxides–magnetite are associated with Al-K-Ti-Fe-Cr-V-Zn-P contents, whereas dolomite–K-feldspar are associated with Ca-Mg-LOI contents; and (2) magnetic minerals (notably, magnetite) are derived mainly from detrital sources.

Since the distribution of geochemical elements in the Canadian Beaufort Sea-Amundsen Gulf region is closely linked to the mineralogical distribution (Figure 12E), either may be used to discriminate regional groups (or provinces) with a different sedimentary composition. Consequently, we performed a fuzzy c-means clustering analysis based on four unassociated key minerals (quartz–phyllosilicates–K- feldspar-dolomite; Figure 6A) to ascertain whether the differences between each predefined province are statistically valid. The ordination diagram and silhouette plots corroborates that sediment samples can be divided into four homogeneous clusters or sedimentary provinces (Figure 13), as indicated by their relatively high membership value (up to 0.60; Figure 13B): (1) Mackenzie Trough-Canadian Beaufort Shelf, (2) southwestern Banks Island, (3) central Amundsen Gulf, and (4) mud volcanoes.

432

433 **5. Discussion**

The spatial variations observed in the siliciclastic grain size, magnetic properties, mineralogical, and geochemical record from the Mackenzie Trough-Canadian Beaufort Shelf and Amundsen Gulf sediments are likely related to changes in redox conditions, surface detrital provenance, and sediment dispersal of the land-derived particles delivered by the Mackenzie River to the shelf and coastal erosion.

438

439 **5.1. Sedimentary redox conditions**

440 Given that Mn forms a highly insoluble oxyhydroxide where oxic conditions prevail (Burdige, 441 1993; Calvert and Pedersen, 2007), the moderate to strong degree of Mn enrichment (EF > 5) in the 442 central Amundsen Gulf and southwestern Banks Island sediments compared to the Canadian Beaufort 443 Shelf suggests that more oxic conditions prevail at the sediment-water interface in this area (Figure 9 and 444 Figure 11A). In agreement with previous geochemical studies (e.g., O'Brien et al., 2006; Mucci et al., 445 2010; Tremblay et al., 2014), we hypothesize that these differences in oxic conditions are likely related 446 with more turbulent mixing of the water column. Indeed, despite the pronounced stratification (Tremblay 447 et al., 2014), strong winds in conjunction with recurrent ice-free conditions for much of the year likely 448 led to strong vertical mixing within Amundsen Gulf. These conditions produce a well-oxygenated water column, but also promote the settling of Fe-Mn oxyhydroxide particles onto the seafloor (e.g., 449 450 Macdonald and Gobeil, 2012). Conversely, on the Canadian Beaufort Shelf, the large supply of 451 terrigenous organic matter from the Mackenzie River keeps the oxic-anoxic boundary close to the 452 sediment-water interface and allows the reductive remobilization and release of Fe and Mn from 453 oxyhydroxide coatings into the uppermost sediments and overlying water column (Magen, 2007; Magen 454 et al., 2011). Alternatively, the high Mn-enrichment factors found in the central Amundsen Gulf and 455 southwestern Banks Island (Figure 9) may also be related to the lower detrital Al-rich minerals (Figure 456 8B) received in these zones as no large rivers discharge into Amundsen Gulf and only a small amount of 457 the Mackenzie River sediment plume reaches the Gulf (e.g., Hill et al., 1991; Macdonald et al., 1998). 458 However, as shown in Figure 9, redox-sensitive elements (Fe, V, Cr, Zn) are not authigenically enriched 459 (EF <3), supporting the idea that predominantly oxic conditions prevail at the sediment-water interface 460 (e.g., Magen, 2007). Accordingly, these redox-sensitive elements reveal a good association with Al, 461 advocating a mostly detrital origin (Figure 10A).

462

463 **5.2. Sedimentary provinces and sediment provenance**

The spatial distribution of principal component scores, together with fuzzy c-means clustering analysis, indicates that there are four provinces with distinct sedimentary compositions within the Mackenzie Trough-Canadian Beaufort Shelf and Amundsen Gulf, as described below.

467 5.2.1. Mackenzie Trough-Canadian Beaufort Shelf

This sedimentary region is the most widespread and is characterized by mineral (phyllosilicates, Fe-oxides, magnetite) and element (Al-K-Ti-Fe-Cr-V-Zn-P) associations mainly found in the finegrained aluminosilicate and Fe-Mn oxide fractions. The variability in detrital input delivered to the Canadian Beaufort Shelf via the Mackenzie River discharge and the subsequent hydraulic sorting most probably controls the distribution and accumulation of these minerals and elements. Note that sediment contribution to the Canadian Beaufort Sea by the Arctic Alaskan rivers and coastal erosion is relatively small compared to the Mackenzie River discharge (Hill et al., 1991). Likewise, with the exception of the 475 mud volcanoes areas, the PC scores from both bulk mineralogy and elemental geochemistry (Figures 7 476 and 11B-C), as well as the Al-Si-Ca relationship (Figure 8A), exhibit a homogeneous sedimentary 477 composition within this province. This homogeneity suggests a common detrital particle provenance and 478 supports the notion that the Mackenzie River sediment plume has a wide easterly propagation, thus 479 influencing modern sedimentation in the entire Canadian Beaufort Shelf area (e.g., Hill et al., 480 1991;Carmack and Macdonald, 2002; Richerol et al., 2008b).

481 The overall mineralogical and geochemical signatures characterizing this province, point to a 482 detrital input from a more aluminosiliciclastic-carbonate sedimentary source such as the Cambrian to 483 Cretaceous shales, sandstones and limestone cropping out in the Interior Platform. This assumption is in 484 agreement with the Sr isotope composition of suspended sediments from the Mackenzie basin (Millot et al., 2003), suggesting that the Mackenzie River system is mostly dominated by the tributaries of the 485 486 Interior Plain, with the northern Rockies and Mackenzie Mountains as secondary sources. In addition, 487 compared to the rivers of the Mackenzie and Rocky Mountains, those of the interior plains also supply 488 large amounts of terrestrial organic matter to the Canadian Beaufort Shelf (Millot et al., 2003), almost all 489 of which (~97%) is deposited on the inner to middle shelf (Macdonald et al., 1998; O'Brien et al., 2006). 490 Accordingly, the slightly lower magnetic susceptibility (k_{lf}) values in this province, where the iron-491 bearing mineral (magnetite and Fe-oxides) contents are highest, may be accounted for by a dilution effect from a high organic-matter supply (e.g., Bloemendal et al., 1992). The large amounts of 492 493 terrigenous organic material deposited on the Canadian Beaufort Shelf compared to Amundsen Gulf 494 (e.g., Macdonald et al., 1998; Magen et al., 2010), lead to organic matter diluting the highly magnetic 495 fine-grained material and, therefore, producing low magnetic susceptibility (k_{lf}) values.

496 5.2.2. Southwestern Banks Island

497 This province is characterized mainly by the association of dolomite–K-feldspar and Ca-Mg-LOI 498 and, to a lesser extent, by phyllosilicates, Fe-oxides and Mn (Figures 7 and 11A-C). Likewise, fine silt

499 contents are higher in this province compared to the Mackenzie Trough-Canadian Beaufort Shelf area 500 (Figures 2A and 3C). Since biogenic carbonates are only of subordinate importance (calcite <1%), 501 detrital carbonate (dolomite) is the main host mineral for Ca-Mg-LOI (Figures 7 and 11). In agreement 502 with petrographic and bulk mineralogy studies performed around Banks Island (e.g., Bader and Henry, 503 1961; Bischof et al., 1996; Vogt, 1997), we hypothesize that most of the fine silt detrital sediments in 504 this province are mainly supplied from coastal cliff erosion of fine-grained Pleistocene carbonate-rich 505 glacial tills and clastic sedimentary rocks cropping out on southwestern Banks Island. In fact, much of 506 the coastline along southwestern Banks Island and Amundsen Gulf is vulnerable to coastal erosion 507 (O'Brien et al., 2006). According to Belliveau (2007), the summer melting of ground ice on the 508 southwestern coast of Banks Island generally leads to slumping along coastal cliffs, which not only 509 creates large erosional areas, but also increases the amount of fine-grained sediments available that can 510 reach the coastline, especially during the open-water season (June-September) when southeast winds and 511 onshore storm winds predominate.

512 Compared to the Mackenzie Trough-Canadian Beaufort Shelf province, this zone has a less-513 extended modern detrital contribution to the eastern part of Amundsen Gulf (Figures 7A and 11B). Note, 514 however, that from a more regional perspective and based on the geochemical fingerprint of entrained 515 Fe-oxide mineral grains in Arctic Ocean sea ice. Darby (2003) suggests that the Banks Island shelf is the 516 most important source of sediment accumulated from ice floes in the Beaufort Sea, northern Chukchi 517 Sea, and the Chukchi Borderland area. Indeed, easterly winds dominate the ice-cover dynamics in the 518 Canadian Beaufort Sea region (Hill et al., 1991) and promotes a recurrent polynya (known as the Cape 519 Bathurst Polynya; Arrigo and van Dijken, 2004) on the southwestern coast of Banks Island (Figure 1). In 520 this shallow, ice-free area, stronger winds in fall and winter induce turbulent mixing of the water 521 column, leading to both a more oxygenated water column and the recurrent incorporation of shelf 522 sediments into the sea ice (a process known as suspension freezing; Reimnitz et al., 1993). These sea ice 523 and ice floes are subsequently transported westwards of the area via surface currents and the clockwise

524 BG current system (Darby, 2003).

525 5.2.3. Central Amundsen Gulf

526 This province represents a transitional zone between the Mackenzie Trough-Canadian Beaufort 527 Shelf and southwestern Banks Island sedimentary provinces and is characterized by intermediate 528 phyllosilicate-magnetite-dolomite-K-feldspar and Al-K-Ti-Fe-Mn-V-Zn-Sr-Ca-Mg-LOI contents 529 (Figures 7 and 11). Similarly to southwestern Banks Island sediments, fine silt contents are also higher in 530 this province compared to the Mackenzie Trough-Canadian Beaufort Shelf area (Figures 2A and 3A). 531 Thus, this zone consists mainly of minerals and elements resulting from a mixture of (1) the 532 aluminosiliciclastic and iron-bearing material likely derived from the Mackenzie River discharge and (2) 533 detrital carbonate and K-feldspar derived from coastal erosion of southwestern Banks Island. Indeed, re-534 suspended shelf sediment and sediment from the Mackenzie River discharge may reach Amundsen Gulf 535 in the summer when northwesterlies dominate (O'Brien et al., 2006). In contrast, because southwestern 536 Banks Island is not influenced by significant river discharges, coastal cliff erosion plays a more 537 important role in sediment supply and transport to the central Amundsen Gulf (Belliveau, 2007). 538 Alternatively, we cannot rule out that other secondary source rivers, such as the Horton and Hornaday 539 rivers (Figure 1), might also reach the Amundsen Gulf during break-up in the spring. Furthermore, 540 because the drainage basin of these rivers is covered by Palaeozoic sedimentary rocks as well as finegrained Pleistocene potassium-rich glacial till resulting from the glacial erosion of Precambrian 541 542 Canadian Shield rocks (e.g., Padgham and Fyson, 1992), they may also supply quartz and K-feldspar 543 minerals to the Gulf.

544 5.2.4. Mud volcanoes

545 This province is characterized by poorly sorted medium to fine silt sediments (Figures 2B and 546 3C) with high quartz-plagioclase, authigenic carbonate (low Mg-calcite and/or dolomite; Figures 7 and 547 S3) and Si-Zr (Figure 11) contents, as well as high magnetic susceptibility values (Figure 5A). As no 548 large rivers discharge to the east Mackenzie River (R-ArcticNet database) and coastal erosion is only an 549 important local sediment supply near the shoreline in the Canadian Beaufort Shelf (Carmack and 550 Macdonald, 2002), we hypothesize that detrital sediments supply within these specific areas are driven 551 by the mud volcanoes activity. Based on seismic stratigraphy studies performed along the eastern 552 Canadian Beaufort Shelf (Blasco et al., 1990; Hill et al., 1991; Batchelor et al., 2013) and submarine 553 mud volcanoes distribution (Blasco et al., 2013), we presume that the detrital sediment in these areas 554 derived from the subsurface sediment remobilisation of the fine-grained Pleistocene quartz-rich glacial 555 tills deposited in the Shelf following mud migration towards the surface. Indeed, fluidized/gasified 556 sediment eruptions associated with the mud volcanoes activity promote the remobilization and transport 557 to the seafloor of the sediments accumulated in the subsurface (Paull et al., 2007; 2015). This 558 interpretation is in agreement with previous studies on the mud volcanoes from the Canadian Beaufort 559 Shelf (e.g., Paull et al., 2007; Blasco et al., 2013; Paull et al., 2015) that suggest that the crest sediments 560 are significantly older than the moat sediments and they almost certainly pre-date the marine 561 transgression (Hill et al., 1993). In addition, the mud volcanoes are also most likely responsible for the 562 higher magnetic susceptibility values in these areas. Lower ratios of Mrs/Ms and higher ratios of Hcr/Hc 563 suggest that mud volcanoes sediments are dominated by coarser magnetite grains (Figure 4), which are 564 responsible for the increase in magnetic susceptibility recorded within this province. This is consistent 565 with the remobilization of fine till material towards the surface by the mud volcanos activity.

566 On the other hand, the high authigenic carbonate contents observed in this province may be 567 related with the gas hydrate decomposition (Moore et al., 2004; Yang et al., 2014). In fact, ascending 568 venting methane derived from the gas hydrate decomposition come in contact with seawater sulfate near 569 the seafloor, where an anaerobic oxidation of methane takes place, favoring the authigenic carbonate 570 precipitation (which may include calcite, dolomite, and/or aragonite). This environmental process has been proposed to operate in different mud volcanoes settings from Fram Strait (Ambrose et al., 2015),
South China Sea (Yang et al., 2014), Niger deep-sea fan (Bayon et al., 2007), and Mediterranean
(Pancost et al., 2001), among others.

To summarize, the spatial trends of our mineralogical, geochemical, and magnetic data, together with the fuzzy c-means clustering analysis, suggest that: (1) the Mackenzie River discharge is the main contributor of terrigenous sediments in the entire Canadian Beaufort Shelf, (2) southwestern Banks Island represents a secondary source of sediments in the central Amundsen Gulf, (3) sediments from the central Amundsen Gulf represent a mix of sediments derived from both the Mackenzie River discharge and coastal erosion of southwestern Banks Island, and (4) the Canadian Beaufort Shelf area is preferentially influenced by mud volcanoes activity (see summary in Figure 14).

581

582 **5.3.** Comparison to other circum-Arctic regions — mineralogical clues

583 Continents surrounding the Arctic Ocean comprise bedrock/soils characterized by different 584 petrographic signatures (e.g., Vogt, 1997; Stein, 2008; Harrison et al., 2008; Bazhenova, 2012; Fagel et 585 al., 2014; Linsen et al., 2014). Consequently, as suggested by Vogt (1997), surface sediments from 586 circum-Arctic source areas may be characterized by very specific K-feldspar/plagioclase (Kfs/Plg) and 587 quartz/(K-feldspar + plagioclase) (Qz/Fsp) ratios. To verify this notion, we compared our bulk 588 mineralogical data with other circum-Arctic regions (Figure 15), notably with surface marine sediments 589 from the Eurasian shelf (Stein, 2008; Bazhenova, 2012), Bering Strait (Linsen et al., 2014), as well as 590 with continental sediments from the northern Yukon Territory, Tuktoyaktuk Peninsula, Cape Bathurst, 591 and Banks Island (Vogt, 1997). Our findings indicate that sediments from the Mackenzie Trough-592 Canadian Beaufort Shelf, Amundsen Gulf, and Banks Island have higher Qz/Fsp (2 to 4) and Kfs/Plg 593 (>0.7) ratios compared to Eurasian sediments (Figure 15). In the study area, quartz and K-feldspar are 594 mainly supplied by the Mackenzie River discharge (which drain large sedimentary platforms) and by 595 coastal erosion of the Pleistocene potassium- and carbonate-rich glacial till cropping out on southwestern 596 Banks Island. In contrast, rivers from the Eurasian margin drain large basaltic provinces that can supply 597 high amounts of plagioclase (Stein, 2008). Plagioclase weathers much faster than K-feldspar as silicate 598 weathering preferentially attacks Na- and Ca-rich minerals (White et al., 2001). In glacial environments, 599 this implies that mechanical comminution of plagioclase is more effective than K-feldspar during the 600 initial stages of feldspar weathering (e.g., von Eynatten et al., 2016). As a result, plagioclase is enriched 601 over K-feldspar in the Eurasian shelf sediments compared to the Canadian Beaufort Shelf and Amundsen 602 Gulf, and thus yields sediments with lower Qz/Fsp (0.2-1) and Kfs/Plg (0.2-0.7) ratios (Figure 15). On 603 the other hand, sediment samples from the mud volcanoes province display higher Qz/Fsp (>3) ratios 604 similar to the glacial tills cropping out along the northern Yukon Territory (VH83-014), Tuktoyaktuk 605 Peninsula (VH85-045), and Cape Bathurst (VH85-006), suggesting a common detrital origin for these 606 sediments, which likely originate from the glacial activity of the Laurentide Ice Sheet. These also 607 support our interpretation that the mud volcanoes province are composed of glacial tills that have been remobilized from the subsurface of the Canadian Beaufort Shelf (see section 5.2.4.). Finally, little 608 609 difference is observed between surface sediments from the Bering Strait and Eurasian sediments (Figure 610 15) because Bering Strait sediments also contain significant amounts of plagioclase that mainly 611 originated from the Anadyr River drainage basin, where volcanic, granitic, and granodiorite rocks crop 612 out (e.g., Linsen et al., 2014).

613

614 6. Conclusions

The spatial variability of continental input, surface currents, and redox conditions within the
 Mackenzie-Beaufort Sea Slope and Amundsen Gulf was investigated through analyses of the grain size,

magnetic properties, and the mineralogical and geochemical composition of 34 surface sediment
samples. The results of this research yield the following generalizations and conclusions:

- 619
 1. The relative enrichment in redox-sensitive elements (Mn, Fe, V, Cr, Zn) suggests that modern
 620 sedimentary deposition within the Mackenzie-Beaufort Sea Slope and Amundsen Gulf took place
 621 under oxic bottom-water conditions;
- Strong winds together with recurrent ice-free conditions for much of the year appear to be a
 plausible scenario for explaining the well-oxygenated water column and, therefore, the high Mn enrichment factors observed in the Amundsen Gulf compared to the Canadian Beaufort Shelf;
- 625 3. The mineralogical, geochemical, and magnetic signatures of surface sediments allowed the 626 identification of four provinces with distinct sedimentary compositions: (1) the Mackenzie 627 Trough-Canadian Beaufort Shelf, characterized by minerals (phyllosilicates, Fe-oxides, 628 magnetite) and elements (Al-K-Ti-Fe-Cr-V-Zn-P) derived mainly from the Mackenzie River 629 discharges; (2) southwestern Banks Island, characterized by the association of dolomite-K-630 feldspar and Ca-Mg-LOI mainly supplied from coastal cliff erosion of Pleistocene potassium- and 631 carbonate-rich glacial tills as well as clastic sedimentary rocks cropping out on the island; (3) the 632 central Amundsen Gulf, which represents a transitional zone typified by intermediate 633 phyllosilicates-magnetite-K-feldspar-dolomite and Al-K-Ti-Fe-Mn-V-Zn-Sr-Ca-Mg-LOI 634 contents resulting from a detrital mix between the Mackenzie River discharges and coastal 635 erosion of southwestern Banks Island; and (4) the mud volcanoes distinguished by the association 636 quartz-plagioclase-authigenic carbonate and Si-Zr contents, as well as high magnetic 637 susceptibility values resulting from the remobilization of glacial tills deposited in the subsurface 638 of the Canadian Beaufort Shelf.

639 4. Our mineralogical data corroborate that K-feldspar/plagioclase and quartz/(K-640 feldspar+plagioclase) ratios (Vogt, 1997), together with detrital carbonate (dolomite), can be 641 successfully used to track changes in terrigenous sediment input from the Canadian Beaufort Sea, 642 Eurasian shelf, and Bering Strait.

Taken as a whole, our data provide a baseline for future studies using the mineralogical, geochemical, and magnetic signatures of sediment cores from the Mackenzie-Beaufort Sea Slope and Amundsen Gulf in order to reconstruct and document past variations in continental inputs and sediment dispersal related to climate changes.

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- 648

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995 **Figure captions**

996

997 Figure 1. Map of the Beaufort Sea and Amundsen Gulf illustrating the location of surface sediment 998 samples used in this study. Partially infilled cross-shelf channels (IT: Ikit Trough, KC: Kugmallit 999 Channel, NC: Niglik Channel) and distribution of mud volcanoes across the Beaufort Shelf are also 1000 illustrated (Blasco et al., 2013). MT represents the Mackenzie Trough; the thick white arrows represent 1001 the coastal surface circulation which is dominated by the Beaufort Undercurrent (here represented under 1002 westerly influence); the black line indicates the maximum extent of the Mackenzie River Plume; the 1003 light blue shading represents the extent of the spring ice-free zone.

1004

Figure 2. Mean grain-size (A) and sorting (B) distributions (phi units) for the Mackenzie-Beaufort Sea Slope and Amundsen Gulf sediments. Note that in (B) all sediment samples are poorly sorted, however, relatively higher sorting (values up to 1.75) is found in some samples from the mud volcanoes area, in the Canadian Beaufort Shelf as well as in the southwestern Banks Island.

1009

Figure 3. End-member modelling analyses (EMMA) performed on the grain-size distribution of the detrital fraction from the Canadian Beaufort Shelf and Amundsen Gulf. (A) The grain-size distribution of the first three end-members accounts for more than 91% of the total variance. (B) Three representative unmixed grain-size distributions derived from EMMA. (C) Log(EM1/EM2+EM3) and (D) Log(EM2/EM3) end-member ratios, which represent the relative proportion between clay/silts and medium-silt/coarse-silt, respectively.

1016

Figure 4. (A) Hysteresis loop curves for three representative samples from the Mackenzie Trough-Canadian Beaufort Shelf (MT-CBS), central Amundsen Gulf (cAG), and southwestern Banks Island (SWBI); and (B) Mrs/Ms and Hcr/Hc crossplot (Day plot) illustrating the magnetic grain size SD (single domain), PSD (pseudo-single domain), and MD (multi-domain) zonation (adapted from Day, 1977).

1021

Figure 5. Magnetic properties of surface sediments from the Canadian Beaufort Shelf and Amundsen
Gulf. (A) Spatial distribution of magnetic susceptibility (k_{1f}); (B) Spatial distribution of Mrs/Ms ratio;
(C) Spatial distribution of Hcr/Hc ratio. Note that all magnetic parameters show similar spatial
distributions.

1026

Figure 6. (A) Biplot of the PC-1 versus PC-2 obtained from the log-centred transformation of the bulk mineralogical data from the Canadian Beaufort Shelf and Amundsen Gulf sediments. (B) Loadings derived from the principal component analysis illustrating the weight (>0.1) of each mineral in the definition of each PC score.

1031

Figure 7. Map of PC-1 and PC-2 scores derived from the bulk mineralogical data from the Canadian
Beaufort Shelf and Amundsen Gulf sediments.

1034

Figure 8. (A) Al₂O₃-SiO₂-CaO ternary plot shows the overall composition of surface sediments from the
Canadian Beaufort Shelf and Amundsen Gulf in comparison with average shale (adapted from
Brumsack, 1989). (B) Log(Fe/Al) versus Log(Mn/Al).

1038

Figure 9. Box plots of the enrichment factors (EF) of redox-sensitive elements (Mn, Fe, V, Cr, Zn) from the Mackenzie Trough-Canadian Beaufort Shelf (MT-CBS), central Amundsen Gulf (cAG), and southwestern Banks Island (SWBI) sediments.

1042

Figure 10. (A) Biplot of the PC-1 versus PC-2 obtained from the log-centred transformation of the major-, minor-, and trace-element data from the Canadian Beaufort Shelf and Amundsen Gulf sediments.

1045 (B) Loadings derived from the principal component analysis illustrating the weight (>0.1) of each 1046 element in the definition of each PC score.

1047

Figure 11. (A) Spatial distribution of Log(Mn/Al). (B-C) Map of PC-1 and PC-2 scores derived from the
 major-, minor-, and trace-element data from the Canadian Beaufort Shelf and Amundsen Gulf sediments.

1050

Figure 12. Relationship between siliciclastic mean grain size (phi units), bulk mineralogy (PC-1 score),
elemental geochemistry (PC-1 score), magnetic susceptibility [Log(k_{lf})], and Log(Hcr/Hc).

1053

Figure 13. Silhouette (A) and principal coordinate ordination (B) plots resulting from the fuzzy c-means clustering analysis based on the bulk mineralogical data from the Canadian Beaufort Shelf and Amundsen Gulf sediments. The parameters listed on the right-hand side in (A) represent of number of sample in each cluster and their membership's values. 100% of the cluster memberships are correctly classified. In (B) each cluster is associated with a small "star" whose segment radiuses are proportional to its membership coefficient.

1060

Figure 14. Generalized map summarizing the main sedimentary provinces defined in this study. The
 thick white arrows represent surface currents.

1063

Figure 15. K-feldspar/plagioclase versus quartz/(K-feldspar+plagioclase) diagram illustrating the mineralogical difference between some circum-Arctic regions. Surface marine sediments from the Canadian Beaufort Sea (this study), Amundsen Gulf (this study), Eurasian shelf (Stein, 2008; Bazhenova, 2012), Bering Strait (Linsen et al., 2014), as well as continental sediments from the northern Yukon Territory, Tuktovaktuk Peninsula, Cape Bathurst, and Banks Island (Vogt, 1997) are plotted. 1069

1070 Supplementary tables

1071

Table S1. Frequency-dependent susceptibilities data for the some bulk sediments samples from the
Mackenzie Trough-Canadian Beaufort Shelf (MT-CBS), central Amundsen Gulf (cAG), southwestern
Banks Island (SWBI;), and mud volcanoes (MV) sediments.

1075

1076 Supplementary figures

1077

1078 Figure S1. Box plots of the main bulk mineral components present in the different sedimentary regions 1079 (Mackenzie Trough-Canadian Beaufort Shelf, mud volcanoes, central Amundsen Gulf, and southwestern 1080 Banks Island). The box plots show the median (horizontal line) and the box includes 50% of the 1081 distribution. Small crosses represent outliers.

1082

Figure S2. Box plots of the major-, minor-, and trace-element concentrations for the different sedimentary regions (Mackenzie Trough-Canadian Beaufort Shelf, mud volcanoes, central Amundsen Gulf, and southwestern Banks Island). The box plots show the median (horizontal line) and the box includes 50% of the distribution. Small crosses represent outliers.

1087

Figure S3. Photography (A) and scanning electron microscopic (B) images of white crusts observed in a representative mud volcano sediment sample (2004-804-609). EDS (Energy-dispersive X-ray spectroscopy) spectrum of white crusts (C). The red circle indicates the location of the EDS analysis. EDS spectrum is consistent with the presence of Ca-Mg authigenic carbonates (notably, low Mg-calcite). Na and Cl peaks reflect the high salt content of the pore waters. Other peaks (e.g., Si, Al, Fe, and P) are 1093 interpreted as background detrital sediment signals. Sulfides (such as, pyrite and greigite) were not 1094 observed in the SEM-EDS analysis. X-ray diffractogram (D) of the bulk sediment sample showing the 1095 presence of dolomite, calcite and low Mg-calcite. The salt content is represented by the halite peak. Note 1096 that the bulk sediment samples were not rinsed with distilled water for the SEM-EDS analysis, hence the 1097 presence of salt. Figure 1.



Figure 2.



Figure 3.



Figure 4.

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Figure 5.

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69°N

50 100 km

nzie

Fine magnetic ^{1.4} grains

1000 m

0 m

Horr

Figure 6.





Figure 7.



Figure 8.



Figure 9.



Figure 10.





Figure 11.



Figure 12.



Figure 13.
Figure 13

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Figure 14.

Figure 14

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Figure 15.

Figure 15

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Graphical abstract.

Graphical abstract



Sedimentary provinces:

- Preferential influence of discharge of the Mackenzie River (1)
- high phyllosilicates, Fe oxides, magnetite and AI-K-Ti-Fe-Cr-V-Zn-P contents
- Preferential influence of coastal erosion 2 high dolomite, K-feldspar and Ca-Mg-LOI contents
- Transition zone (mixture characteristics from zones 1 and 2) 3 intermediate phyllosilicates, magnetite, K-feldspar, dolomite, and Al-K-Ti-Fe-Mn-V-Zn-Sr-Ca-Mg-LOI contents
- Zone influenced by mud volcanoes (4)
 - high quartz-plagioclase-authigenic carbonates and Si-Zr contents, high magnetic susceptibility