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Depositional variability of estuarine intertidal sediments and implications for metal distribution: an example from Moreton Bay (Australia)

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ABSTRACT

This study examines the patterns of depositional variability, sediment geochemistry and metal distribution in intertidal areas of Moreton Bay, southeast Queensland, Australia. Recent concern over increasing human impact on the bay has generated the need to obtain evidence on how the disturbance of the depositional setting might affect the natural estuarine environment. Sediment stratigraphy, major, and trace element analyses of sediment cores show that the sedimentation pattern is unique to each intertidal site. Disturbed ^{210}Pb and ^{137}Cs activity profiles of some of the cores indicate that sediment reworking occurs across the intertidal flats up to a depth of at least 80 cm. With some notable exceptions, an accurate geochronology of the surface sediments could not be established due to low ^{210}Pb activities and sediment mixing. Thus, an increase in Pb, Zn and Cu towards the surface sediments observed at various sites is attributed to both anthropogenic contribution following the rapid urban development in the last century and to post-depositional diagenetic processes, bioturbation and sediment re-suspension induced by tides, storms or floods. Sediment cores are representative only of the local sedimentation and may not always allow extensive correlation to larger areas. Vertical profiles of heavy metals reflect the different

depositional environment controlled by the complex hydrodynamics of the bay. Local hydrologic, physical, and tidal conditions might induce metals redistribution at different scales. This information is of critical importance in view of sediment remobilization caused by future development such as dredging, intertidal areas reclamation or excavation of new navigational channels.

Keywords: Metals; sediment cores; sediment mixing; ^{210}Pb; estuaries; Moreton Bay.

1. INTRODUCTION

Moreton Bay, located in southeast Queensland is one of the largest estuarine systems in Australia and is internationally recognized for its biodiversity and ecological significance (Dennison and Abal, 1999). Last century's accelerated urban, agricultural and industrial development led to substantial modifications of the region with only 28% of the catchment area remained undisturbed after intensive land clearing, deforestation, agriculture and urbanization (Capelin et al., 1998; Neil 1998; Dennison and Abal, 1999). The western sides of the bay are facing continuous pressure from new developments and intensive alterations of the intertidal areas, such as the establishment of the Brisbane port and airport and the construction of recreational harbours. Sediment deposition in marine coastal environments is a natural process controlled by geography, geology, geomorphology, and climate variability. However, following the rapid urbanization, sediments transported from the catchment to the bay have increased together with the associated metal loads and nutrients (Neil, 1998; Dennison and Abal, 1999; Duke et al., 2003; Cox and Preda, 2005; Healthy Waterways 2012; Morelli et al., 2012), and represent a major environmental problem. For example, nitrate and phosphate concentrations in the Brisbane River waters increased by 22 and 11 times, respectively, in the last 50 years (Dennison and Abal, 1999; Duke et al., 2003; Cox and

Preda, 2005). Frequent blooms of *Lyngbya*, a toxic cyanobacteria, in the western side of the bay possibly reflect high nutrient contents in the waters, with western Moreton Bay considered to be eutrophic (Dennison and Abal, 1999) and to have an altered hydrogeological regime (Logan et al., 2010). Most importantly, sediments deposited in bays and estuaries represent a potential sink for anthropogenic-derived metals in any aquatic environment because of their large adsorption capability (Siegel, 2002). Hence, estuarine sediments deposited in the past may be more contaminated than recent sediments (e.g. Pirrie et al., 1997; Cearreta et al., 2000; Cundy et al., 2003). Post-depositional disturbance by reworking processes (bioturbation, floods, tidal currents or dredging) can result in sediment re-suspension and release of potential contaminants into the food chain (Tessier and Campbell, 1988), thus contributing to the long-term contamination of many estuaries (e.g. Forstner and Wittman, 1983; Eyre and McConchie, 1993; Swales et al., 2002; Cundy et al., 2003; Forstner and Salomon, 2008; Smith et al., 2008; Larrose et al., 2010).

Due to its sub-tropical location, Moreton Bay is subjected to seasonal intense floods during the wet season, resulting in significant events of sediment deposition and/or resuspension (Neil, 1988). Consequently, it is important to have a clear understanding of the depositional setting, distribution and source of sediments together with trace elements variability to support any management strategy for the protection of this estuarine ecosystem. A number of studies have characterized trace metal distribution in Moreton Bay sediments (Cox and Preda, 2005; Brady et al 2014a; Brady et al. 2014b; Morelli and Gasparon, 2014) and provided some information at the whole-bay scale. A study on sediment cores revealed considerable variability in sedimentation rates across four intertidal areas (Morelli et al., 2012). It remains to be established, however, whether the depositional and geochemical properties of the intertidal sediment profiles are consistent across the bay. This characterization is critical to identify the natural and anthropogenic components of the sediments, and to monitor possible sediment contamination in view of any disturbance of the sediment column due to excavation and maintenance of navigational channels, intertidal areas reclamation or increased recreational activities.

This study on estuarine sedimentation and geochemistry documents the complexity of Moreton Bay intertidal areas. Results of nine intertidal sediment cores are discussed in this article and are integrated with an additional set of data from four cores described in a previous study (Morelli et al., 2012). Stratigraphic observations, major and trace element compositions, and ^{210}Pb and ^{137}Cs activities were integrated to 1) characterize the sedimentary settings of the intertidal sites, 2) identify the geochemical variability among the sediments of different sites and 3) identify the processes that control vertical metal distribution along the bay sediments.

2. MATERIALS AND METHODS

2.1 Regional setting

Moreton Bay in Southeast Queensland is a semi-enclosed estuarine embayment approximately 110 km long and about 35 km wide with an average depth of 6.8 meters (Fig. 1). The bay is delimited to the east by three sand dune barrier islands (Moreton, North and South Stradbroke) and includes five major catchments: the Brisbane River, Logan/Albert Rivers, Pine Rivers, Pumicestone Passage, and Caboolture River. The climate is humid subtropical with a dry winter season (June to September) and a humid, hot summer season (November to April) and is subjected to the El Niño Southern Oscillation (ENSO) (Eslami-Andargoli et al., 2009). Hydrographic data for the Brisbane and Logan Rivers record long-term periods of low runoff alternating with periods of high runoff. During the year south-easterly winds dominate, with north easterly sea breeze in summer afternoons and south-westerly to north-westerly winds in winter (Stephens, 1992). The prevailing south-easterly winds push the water to the north inducing a northward circulation. The patterns of sedimentation are strongly controlled by ebb and flow tidal currents entering the bay from the North Entrance between Moreton and Bribie islands, the South Passage, and the Jumpinpin Passage (Newell, 1971; Milford and Church, 1976; Church, 1979). Along the shallower western side of Deception Bay and Bramble

Bay water circulation is mainly influenced by river deposition (Maxwell, 1970) and driven by northward weak tidal currents following the shoreline (Patterson, 1992). Waterloo Bay is characterized by a hyposaline, turbid environment with restricted water circulation and shallow bathymetry. In South Moreton Bay, since the opening of Jumpinpin Bar in 1896, oceanic tides and waves enter from the Jumpinpin Passage and travel northwards, mixing with the Logan delta flow (Steele and Mendoza, 1993) and leading to the erosion of the existing low energy tidalites and the formation of the present tidal channels among the islands.

The dominant lithologies in the Moreton Bay region are Jurassic lithic feldspathic sandstone, siltstone and shale (Marburg Formation), Devonian/Carboniferous shale, mudstone argillite, chert and greywacke (Neranleigh Fernvale Beds), Miocene alkali-olivine basalts (Main Range Volcanics), Triassic andesites (Neara Volcanics), polymictic pebble to boulder conglomerate mixed with feldspathic sandstone (Esk Formation) and Quaternary alluvium sand, silt and clay (Lohe, 1980; Ewart et al., 1985; Ham, 1993; Walker, 1998; Caitcehon et al., 2001; Heim, 2002; Douglas et al., 2003; Willmott, 2004; Douglas et al., 2007).

2.2 Sampling methods

Sampling was designed to target depositional areas of Moreton Bay characterized by fine sedimentation, because of the preferential partitioning of trace metals to fine particles (Schorer, 1997; Siegel, 2002). The analysis of Google Earth images and a recent Moreton Bay Seafloor Map (SEQ Catchment, 2008) allowed preliminary qualitative identification of areas of current sediment deposition. The observation of aerial photos taken since 1946 (obtained from the Queensland Department of Natural Resources and Mines – DNRM; unpublished) made it possible to identify changes in the coastline and the evolution of the major channels and sand bars. Sediment cores were collected from intertidal mud-sand flats close to mangrove-vegetated western shorelines in Deception Bay (G9), Bramble Bay (G24, G26, G16), Waterloo Bay (G14, G15, G21, G23), and

South Moreton Bay (G29, G32, G27, G30) (Fig. 1). Cores G37, G34, G6, and G31 were collected in a previous campaign and results of ^{210}Pb, ^{137}Cs, and trace metal trends have been discussed in Morelli et al. (2012). Hand-pushed aluminium tubes 150-300 cm long were used to collect the sediment cores. The tubes were then filled with seawater and a plug inserted in the top opening of the core prevented sediment loss as the tubes were slowly extracted by hand. A diamond saw was used to make two longitudinal cuts in the aluminium casing, and the sediment cores were split into two halves with a nylon string (Morelli et al., 2012). After qualitative description of their stratigraphy, lithology, and possible bioturbation, the cores were sliced with an acid-cleaned plastic cutter at 3-5 cm intervals from the surface to 50 cm. The lower part of the cores was sampled with a 10 cm resolution. Samples were stored in plastic containers in a dark cold room (T= 2-4 °C) until analysis. One fraction of the sample was used for geochemical analyses after oven drying at 60 ^{o}C, and the other to establish grain size and organic matter content (Loring and Rantala, 1992). In addition to the cores described in Morelli et al. (2012), five cores (G16, G14, G21, G29, G32) were chosen for radiometric isotope measurements. Four samples in the top 75 cm were analyzed for ^{210}Pb and ^{137}Cs from cores G16, G29, and G32 and six samples in the top 150 cm from cores G21 and G14.

2.3 Analytical methods

Grain size analysis was carried out using a laser particle size analyzer (Malvern 2000, Mastersizer) after wet sieving the samples to the <1 mm fraction. Organic matter was determined using weight-loss-on-ignition (LOI), a method of approximating the organic and carbonate content in sediments (Dean, 1974; Heiri et al., 2001; Boyle, 2004). After oven drying the sample at 105 °C to constant weight, the organic matter was determined from the loss of mass after heating the sediments at 550 °C for four hours (Dean, 1974; Heiri et al., 2001).

Metal concentrations were determined by complete dissolution using a hot-plate open beaker digestion procedure with screw-top Teflon beakers. A combination of ultrapure acids (HNO_3+HF, HNO_3, HCl+HNO_3) was used sequentially until complete sediment dissolution was achieved. For each batch of samples, laboratory blanks, procedural blanks, replicates, and certified reference materials (AGV-2 -USGS Andesite and MESS-3 -USGS marine sediment from the Beaufort Sea) were analysed for analytical quality control. Sample preparation was performed in the Radiogenic Isotope Laboratory (Class 100 to 1000) at The University of Queensland. All plastic lab wares were acid cleaned and rinsed with Milli-Q water before use. Concentrations of Zn, Pb, Cu, Ni, Co, As, Th, Ba, Rb, Sc, and REE were determined by Inductively Coupled Plasma Mass Spectrometry (ICP-MS - Thermo X7 ICP-MS) and Al, Na, Ca, Fe, Mn, Ti and K were analysed by Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES – Perkin Elmer Optima 3300). Concentrations of trace elements analysed were all above the detection limits and concentrations in the procedural blanks were in the pg/g range and therefore negligible. The precision of the method based on multiple determinations of reference standard materials (AGV-2 thirteen replicates; MESS- 3, seven replicates) was very good. For AGV-2, relative standard deviation (RSD) values were < 5% for all the elements except Cu (RSD 7%) and Pb (RSD 9%); for MESS-3, RSD typically ranged from 1 to 3% with the exception of Cu (6%). Analytical accuracy compared to the certified standard values of AGV-2 and MESS-3 was within RSD < 5% for all the elements of interest, except for Gd (9%) in MESS-3.

The activities of radionuclides ^{210}Pb and ^{137}Cs were measured using a Compton suppression gamma spectrometer located at ANSTO (Australian Nuclear Science Organization). Approximately 30-40 grams of sample were used for the analysis. ^{210}Pb activity was determined using the 46.5 keV peak and ^{226}Ra activity was calculated using ^{214}Pb and ^{214}Bi at 351.9 keV and 609.3 keV, respectively. Unsupported ^{210}Pb activity was calculated by subtracting the ^{226}Ra activity from the ^{210}Pb activity. ^{137}Cs activity was determined from the 662 keV peak after subtraction of the ^{214}Bi peak interference.

3. RESULTS

3.1 Lithology and grain size

Moreton Bay intertidal sediments are predominantly sand to sandy silt (Fig. 2). In the north and south side of Deception Bay (G37 and G34) sediments are characterized by silt and clay (mud ~ 50%), while fine sand with several shell deposits dominates in the western side of Deception Bay (G9), and is underlined by yellow-brownish solid clay below ~ 82 cm (Fig. 3). Silt forms the top 50 cm of core G6 in the Pine River. Bramble Bay sediments are formed by grey-brown very fine-to-fine sand with layers of ~1-2 cm white-grey bivalves. The top meter of core G14 in Waterloo Bay is composed of very fine grey sand changing to fine dark grey mud (70%) below 90 cm, similar to the deposit at the bottom of cores G37 and G34. At the mouth of Tingalpa Creek (G21 and G23) fine sand is the dominant sediment fraction. In South Moreton Bay (G29 and G32) grey brown very fine to medium sand is mixed with ~20-30 % mud, changing in the lower part of the cores to white-grey fine to coarse sand with black grey coarse grains (stratigraphic logs are shown in Fig.3). LOI at 550 °C ranges from 1% to 18 % and is positively correlated with the fine sediments fraction in each core (r^2=0.60 to 0.88).

3.2 ^{210}Pb and ^{137}Cs radionuclides

Total ^{210}Pb activities ($^{210}Pb_{tot}$) were detected in the top 100-150 cm (Fig. 4) ranging from a maximum of 56.7 Bq/kg (G34) to a minimum of 7.6 Bq/kg (G16) with their maxima at or near the top of the cores. Unsupported ^{210}Pb ($^{210}Pb_{uns}$) activities in the surface sediments varied from 9.3 Bq/kg in core G29 to a minimum of 4.1 Bq/kg in core G21. In core G16 $^{210}Pb_{uns}$ was found only in the top layers, and low $^{210}Pb_{tot}$ (<10 Bq/kg) and constant values with depth indicate possible

sediment mixing or sediment loss. Very low $^{210}Pb_{uns}$ in core G14 (~7.3 Bq/kg) showed a slight decrease with depth. Total and unsupported ^{210}Pb in cores G29 and G32 decreased with depth, but the low $^{210}Pb_{uns}$ surface activities (9.3 ± 3.2 and 6.9 ± 2.1 Bq/kg, respectively) do not allow calculation of a meaningful age for the sediments. In the top ~10 cm the constant activities of $^{210}Pb_{tot}$ are indicative of sediment mixing. In all the cores, the low ^{137}Cs activities in the top 30-40 cm (Fig. 4) do not show the typical atmospheric testing fallout (Longmore et al., 1983; Simms et al., 2008). In core G16 ^{137}Cs was only detected in surface sediments (0.4 ± 0.2.Bq/Kg) and at 81 cm (0.6 ± 0.3 Bq/kg). Activities of ^{137}Cs were very low in G14 surface sediments (0.8 ± 0.3 Bq/kg) and in core G21 (0.5 ± 0.2 Bq/kg), but were below detection in core G29. In core G32, ^{137}Cs ranged from 1 ± 0.2 Bq/kg to 1.8 ± 0.4 Bq/kg. The peak in ^{137}Cs (1.8 ± 0.4 Bq/kg) at 15 cm could correspond to ~1963, suggesting sediments are at least younger than 1963, although this cannot be conclusively stated due to the lack of ^{210}Pb data.

3.3 Sediment source and geochemistry

Intertidal Moreton Bay sediments are more enriched in Al_2O_3, CaO and Na_2O than K_2O (A-CN-K Nesbitt and Young, 1984) and are geochemically similar to the Marburg Formation, a mixture of meta-sedimentary Jurassic feldspathic sandstone, siltstone and shale (Fig. 5). Sediment geochemistry is also influenced by the Basaltic Volcanics, the Main Range Volcanics (Miocene alkali-olivine basalts) and the Neranleigh Fernvale Beds (Devonian/Carboniferous shale, mudstone argillite, chert and greywacke) in agreement with previous studies on Moreton Bay sediment sources (Caitcheon et al., 2001; Douglas et al., 2003).

Average concentrations of PAAS-normalized REE (PAAS-Post Archean Australian Shale, Taylor and McLennan, 1985) in each core (Fig. 6) show that sediments are slightly enriched in Eu ($Eu_{(PAAS)}$ = 0.23-0.74) and in HREE (heavy REE) relative to LREE (light REE). In Deception Bay

(G9) sediments are enriched in Gd and the weathered clay at the base of G9 has a distinctive LREE enrichment relative to HREE ($La/Lu_{(PAAS)}$=1.18), a positive Gd anomaly ($Gd/Lu_{(PAAS)}$=1.88) and a Eu negative anomaly. The northern part of Waterloo Bay (G14) is characterized by similar REE patterns as Bramble Bay (G16, G26, G24) and enrichment in MREE ($Gd/Lu_{(PAAS)}$=1.46) compared to Tingalpa Creek (G21, G23). Sediments in South Moreton Bay (G29 and G32) are characterized by MREE enrichment over HREE.

Figure 7 shows that the La, Sc, Th and Co concentrations of Moreton Bay sediments cluster around the PAAS and MUQ composition. Ratios of La/Sc and Th/Sc are low for the bottom sediments of Deception Bay (G34 and G37) and Waterloo Bay (G14) while Bramble Bay (G24, G26, G16) and Waterloo Bay (G14 top 100 cm) sediments are characterized by the highest ratios of Co/Th (>3). Barium and Rb range between 51.8 and 357.6 µg/g, and between 5.7 and 74.6 µg/g, respectively (Tab. 1) and are enriched in sediments of cores G29, G32, G21, G6 (Fig. 7).

Depth profiles of trace metals are shown in Fig. 8. Concentrations are normalized against Al to account for variations in the mineral fraction and grain size (Loring and Rantala, 1992). Metal concentrations vary between 10-82 µg/g for Zn, 4-17 µg/g for Pb, 3-27 µg/g for Cu, and 3-52 µg/g for Ni. Increasing concentrations of Zn and Pb towards the surface are found in cores G16, G24 and G14. Similar trends are observed in the Al-normalized profiles at site G21 (Zn/Al and Cu/Al) and G23 (Pb/Al) showing that the increasing profiles are not an artefact of variable grain size. In contrast, high Zn concentrations (~62 µg/g), Pb (~12 µg/g) and Cu (~16 µg/g) found in the top 5 cm in South Moreton Bay (G29) are associated to an increase in the fine fraction. Maximum concentrations of Zn, Pb, Cu and Ni at 40 cm in core G26 and at 75 cm in core G24 are associated with a high LOI (~10%).

4. DISCUSSION

4.1 Spatial and temporal variability of the depositional environment

Results of sediment geochemistry and their relationship with source rocks are consistent with earlier observations suggesting a mixed sedimentary source for Moreton Bay (Morelli et al., 2012). The little variations among the sites of the PAAS-normalized REE patterns (Fig. 6) are associated either to a lack of REE variability of the source material, or more likely to REE homogenization following weathering, sediment transport and deposition. Lanthanum, Sc, Th, and Co cluster around the Australian Post Archean Shale composition (PAAS, Taylor and McLennan, 1985) and around the average values for sediments in Queensland -MUQ (MUd from Queensland; Kamber et al., 2005). The ratios La/Sc, Th/Sc, and Co/Th are often used to distinguish mafic from felsic sources (Taylor and McLennan, 1985; Singh, 2009; Singh, 2010). Sedimentation in the northern part of Moreton Bay (Deception Bay and Bramble Bay) is more influenced by mafic sources, while the southern part of the bay (Waterloo Bay and South Moreton Bay) is dominated by felsic materials and characterized by enrichment in Ba and Rb. Nevertheless, as shown by the core stratigraphy (Fig.3) the depositional patterns are unique to each site and correlation across Moreton Bay cannot be easily made. Once sediments are deposited, several reworking processes at a local scale may result in geochemical signatures that differ from the original input in terms of compositional variability and degree of homogenisation.

In Bramble Bay the top 1.50 meters of the intertidal flats are dominated by relatively homogeneous very fine- to fine sand, in contrast with the sediment distribution map of Heggie et al. (1999), where the south west coast of Bramble Bay is mostly characterized by mud deposition. Bramble Bay is characterized by the longest water residence time in Moreton Bay (over 60 days) and fine sediments on the seabed are easily resuspended (Dennison and Abal, 1999). Hence, the low content in mud (~10%) found along the three cores (G26, G24, G16) suggests that tidal currents transport silt and clay materials outside of the intertidal area. Compared to the other embayments, sediments are more enriched in Co and La, suggesting a dominantly mafic source.

Sedimentation in the northern and southern part of Waterloo Bay shows compositional and textural differences. The top 100 cm of core G14 are geochemically close to the mafic composition of Bramble Bay (high Co/Th and Sc/Th ratios). In the southern part of Waterloo Bay (G21 and G23), the lower Co/Th and Sc/Th ratios suggest that sedimentation from the Tingalpa Creek is more influenced by felsic sources. The area is characterized by high depositional variability as shown by the different grain size and stratigraphy of the two nearby cores as well as by the marked difference in the Rb/Al vs Ba/Al ratios. Sedimentation is most probably driven by small-scale hydrodynamic processes. Strong currents, different tidal inflow/outflow or floods may erode, re-suspend and re-deposit sediments resulting in preferential deposition of fine particles, for example at site G23.

In Southern Moreton Bay (G32, G29), the dominance of fine to medium sand and the similar geochemistry indicate that the recent sedimentation is affected by strong hydrodynamic conditions, not allowing most of the fine sediments transported by the Logan River to deposit. The different La/Sc and Co/Th ratios and the predominant clay to sandy silt at site G31 could thus represent the finer sediment fraction from the Logan River carried northward by currents. The quartzose marine sand characterized by low ratios of La/Sc and Th/Sc, typical of felsic rocks, found in the lower sediments of cores G29 and G32, is associated to the opening of the Jumpinpin Bar in 1896, when high-energy marine sand transported from the ocean into the bay deposited over the fine sediments rich in organic matter typical of the old low energy tidal environment.

The only common sedimentary feature found across Moreton Bay is the weathered clay at the base of cores G9, G15, G27 and G30. This layer is correlated to the Pleistocene sub-aerial weathering surface exposed during the period of lowered sea level (Jones et al., 1978; Hekel et al., 1979; Clark, 1998) and constrains the deposition of the sediments considered in this study to the Holocene. Consequently, a qualitative correlation of the mud-clay deposits found below 100 cm in cores G37, G34 (Deception Bay), and G14 (Waterloo Bay) and their similar Sc/Th and Co/Th ratios, suggests that they may correlate to the sedimentation of the quiescent basins formed during the Holocene transgression (described by Flood, 1978, Jones et al. 1978, and Hoffman, 1980) filling the

depressions of the pre- Holocene topography. We infer that in the area of G37, G34 and G14, the weathered clay is found at the base of mud deposits (dashed grey line in Fig. 3). The lack of marine mud above the Pleistocene surface on the west side of Deception Bay (core G9) highlights the variability of the depositional environment characterized here by higher energy, erosion and preferential deposition of sand.

4.2. Implications for sediment mixing

Profiles of ^{210}Pb suggest that post-depositional diagenetic processes have influenced sediment accumulation in most of the Moreton Bay intertidal areas, although the sites considered in this study were selected as the most likely to have preserved their original stratigraphy. Sediment mixing occurred at least in the top 10-15 cm and it can affect deep sediments. For example in core G16 $^{210}Pb_{uns}$ activities are constant to a maximum depth of about 80 cm and ^{137}Cs is recorded at 85 cm. Where ^{210}Pb activities have an exponential decay below the surficial mixing layer (core G14, G29, G32), it is conceivable that those sediments were deposited at least in the last 100-150 years (Appleby and Oldfield, 1992). However, the low $^{210}Pb_{uns}$ activities in the surface sediments (less than 10 Bq/kg) do not allow derivation of a sufficiently accurate age model. A peak in ^{137}Cs activity (1.8 ± 0.4 Bq/kg) at 15 cm in core G32 constrains the deposition of the sediments to the last 50-60 years, yet for this core it is difficult to infer where $^{210}Pb_{uns}$ and ^{137}Cs activities reach close to zero values because no samples were analysed below 30 cm. A profile with these characteristics may result from the combination of low ^{210}Pb activities, bioturbation, physical mixing or sediment loss caused by waves and tidal action, responsible for transporting radionuclides downward in the sediment column, as commonly occurs in such environments (Heijnis et al., 1987; Crusius et al., 2004; Ruiz-Fernandez et al., 2009). As discussed by Palinkas and Nittrouer (2007), variations in ^{210}Pb activities can also be related to episodic delivery of sediments, such as flood sediments supplied in higher concentrations that commonly preclude particles from scavenging the usual

amount of ^{210}Pb. The overall low amount of mud content is also consistent with low ^{210}Pb activities and the variability of mud/sand fraction along the cores is also known to cause differences in activities (e.g., He and Walling, 1996; Ruiz-Fernandez et al. 2009). Numerous studies have documented the partitioning of ^{137}Cs and other radionuclides among different particle size fractions of soils and sediments (e.g. Walling and Woodward, 1992; He and Walling, 1996; Dubrulle et al., 2007; Korobova et al., 2014) and how the mineralogical composition of suspended and bottom sediments may affect the fate of ^{137}Cs (Lujaniene et al., 2005). For example, Walling and Woodward (1992) found that the activities of ^{137}Cs decrease with increasing particle size. The variability of ^{137}Cs found in Moreton Bay sediments coupled with the absence of marked increases are possibly explained by the combination of a non-homogenous grain size together with the low ^{137}Cs activities typical of the southern hemisphere, since original ^{137}Cs fallout here is five times lower than in the northern hemisphere (UNESCAR, 2000; Pfitzner et al., 2004).

In the absence of a reliable temporal record the observed metal trends are associated to either a recent metal contribution from anthropogenic inputs, or to the homogenization of the sediment profile and the redistribution of metals induced by mixing processes. For example, in Bramble Bay similar concentrations of Zn, Pb, Ni and Cu are found in core G16 and G24, but metals show no consistent vertical trends along the intertidal area with only Zn/Al increasing towards the surface in both cores. Surface sediments at these sites were classified as only moderately contaminated with Ni and As (Morelli and Gasparon, 2014). Bramble Bay is one of the most environmentally degraded parts of Moreton Bay (Healthy Waterways, 2012), with the Brisbane River, Kedron Brook and Pine River delivering nutrients and suspended sediments into the tidal region from their highly urbanized catchments. However, as shown by the ^{210}Pb activities and by the grain size variability, metals distribution in this intertidal area is controlled more by the hydrologic regime and by mixing processes than by the direct recent accumulation of metals in the sediments. In Waterloo Bay, core G23 sediments have significantly higher concentrations of Pb, Zn and Cu and increasing trends towards the surface of their metal/Al profiles compared to sediments of core G21. The shallow

bathymetry of Waterloo Bay and restricted water exchange with the open ocean probably induce different small scale hydrodynamic processes at the two sites, contributing to the distinctive metal trends, as also suggested by the ^{210}Pb activities, the geochemistry and the grain size distribution. At site G14 in the northern part of Waterloo Bay, the top 40 cm of sediments were deposited in the last 100 years based on ^{210}Pb results. The observed increase in Zn/Al, Pb/Al and Cu/Al in this interval is correlated to the development of the refinery located in Lytton and established in the 1960's, to the runoff from the Brisbane airport, and to the nearby wastewater treatment plants. The increase of the Al-normalized As profile from 80 cm to 20 cm, followed by a decrease in the top 20 cm is possibly associated to the same industrial development around the area. Surface sediments at this site are contaminated with As (Morelli and Gasparon, 2014). Thus, following sediment mixing further ecosystem deterioration could be induced by the redistribution towards the surface of the As found below 20 cm. In South Moreton Bay (cores G29 and G32) metal distribution is more strongly influenced by tidal exchange from the ocean rather than by the Logan River, as indicated by the lack of well-defined metal profiles. Although the data set shows some compositional variability, trace metal abundances do not differ substantially from the MUQ composition (Zn=73.47 mg/kg; Pb=20.44 mg/kg; Cu=32.36 mg/kg; Ni=31.57 mg/kg). In contrast, at site G31 increasing concentrations of Zn, Cr, Cu Cd and Pb in the top 60 cm were correlated to the anthropogenic development occurred in the last 100 years (Morelli et al., 2012) and surface sediments are contaminated with Ni and As (Morelli and Gasparon, 2014). These results suggest that the finer and more contaminated sediment fraction from the Logan River is transported northward by currents and deposited in a less mixed location, as also shown by the different grain size distribution in the three cores.

A comparison of cores collected in different areas of Moreton Bay suggests that post-depositional diagenetic processes and mixing are most likely responsible for vertical re-distribution of metals in the sediment column. Hence, the majority of the sites can provide information only on existing metal loads, but not on their temporal trends. There are, however, exceptions to this observation:

previous work showed that four of the sites (G37, G34, G6 and G31) have preserved an undisturbed sedimentation record for at least the last 100 years (Morelli et al., 2012).

Assessment of the anthropogenic contribution of metals in sediments is often achieved by comparison to geochemical backgrounds estimated using elemental concentrations in the continental crust, or in local, non-contaminated sites, or in the deepest sediments of cores (Reimann and de Caritat, 2005). However, as found in Moreton Bay, metal distribution in sediment cores does not always reflect a record of temporal contamination, as rapid sediment accretion or vigorous mixing often cause metal redistribution in the sediment column (e.g. Lee and Cundy, 2001; Cundy et al., 2003; Rosales-Hoz et al., 2003). Profiles of heavy metals and elemental ratios are unique to each site, reflecting the different depositional environments. Intertidal sedimentation is mostly driven by the hydrodynamic conditions of the bay, climate, topography of the catchment area as well as regional geology. However, at each site the depositional variability is often overprinted by localized factors: local tidal and wind conditions, different vegetation cover, variable rainfalls, and/or different urbanization and industrialization of each catchment. Results from the present study highlight the geochemical and depositional variability of Moreton Bay intertidal areas (summarized in Fig.9) showing that simplistic generalizations of temporal and spatial trends of metals in similar estuaries may lead to an incorrect assessment of current and past environmental impacts.

5. CONCLUSIONS

This sedimentary and geochemical investigation of Moreton Bay intertidal sediments provides critical information for the understanding of estuarine sedimentation processes and metal distribution. Sediments are made of sand to sandy silt and their geochemical signature reflects a mixture of different source rocks outcropping within the Moreton Bay region. The processes that may influence metals' behaviour in intertidal areas were identified. Physical and/or biological mixing disturbs the sediment sequence of the intertidal mudflat at least in the top 10 cm of most of

the sites, and up to a maximum depth of 80 cm. When interpreting increasing Pb, Zn and Cu concentrations towards the surface in the sediment column, it is difficult to discern the contribution of anthropogenic contamination from that caused by post-depositional diagenetic processes. Following mixing induced either by natural causes (e.g., flooding events) or by anthropogenic activities (e.g., dredging) long-term pollution problems could be induced by the release of metals associated with previously contaminated sediments. Further, with this contribution we have highlighted that sediment cores from intertidal areas of large estuarine embayments are often representative of a local depositional environment and may not always allow extensive spatial correlation over larger areas.

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HIGHLIGHTS

Sediment cores from intertidal estuarine areas represent local depositional sedimentation and/or metal accumulation
Sediment deposition across the intertidal flats is affected by reworking and mixing at different depths (up to 80 cm)
Vertical variability of Pb, Zn and Cd is associated to both anthropogenic and natural sources
Sediment remobilization caused by future developments (e.g. dredging) might induce metals redistribution

Fig 1

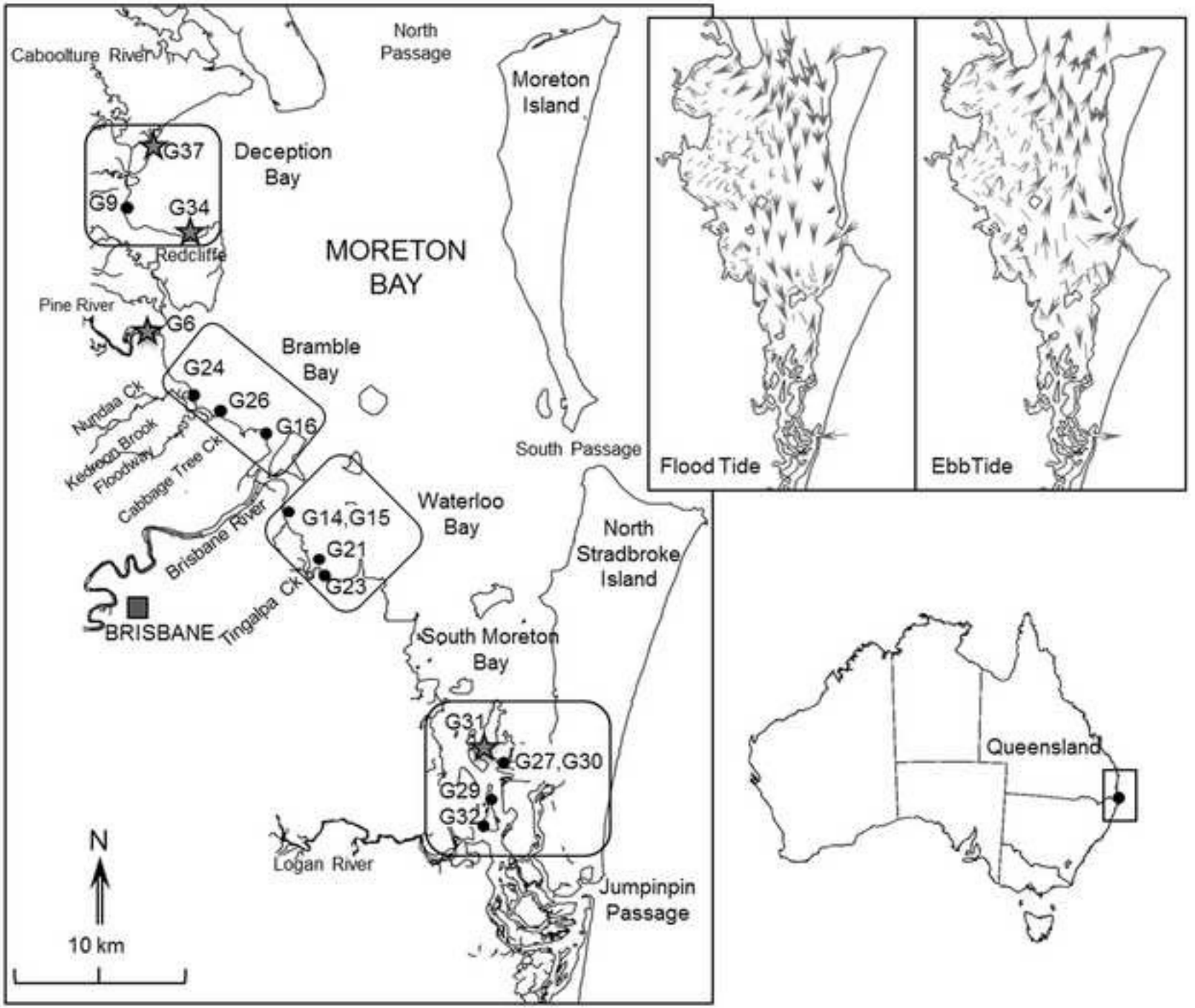


Fig 2

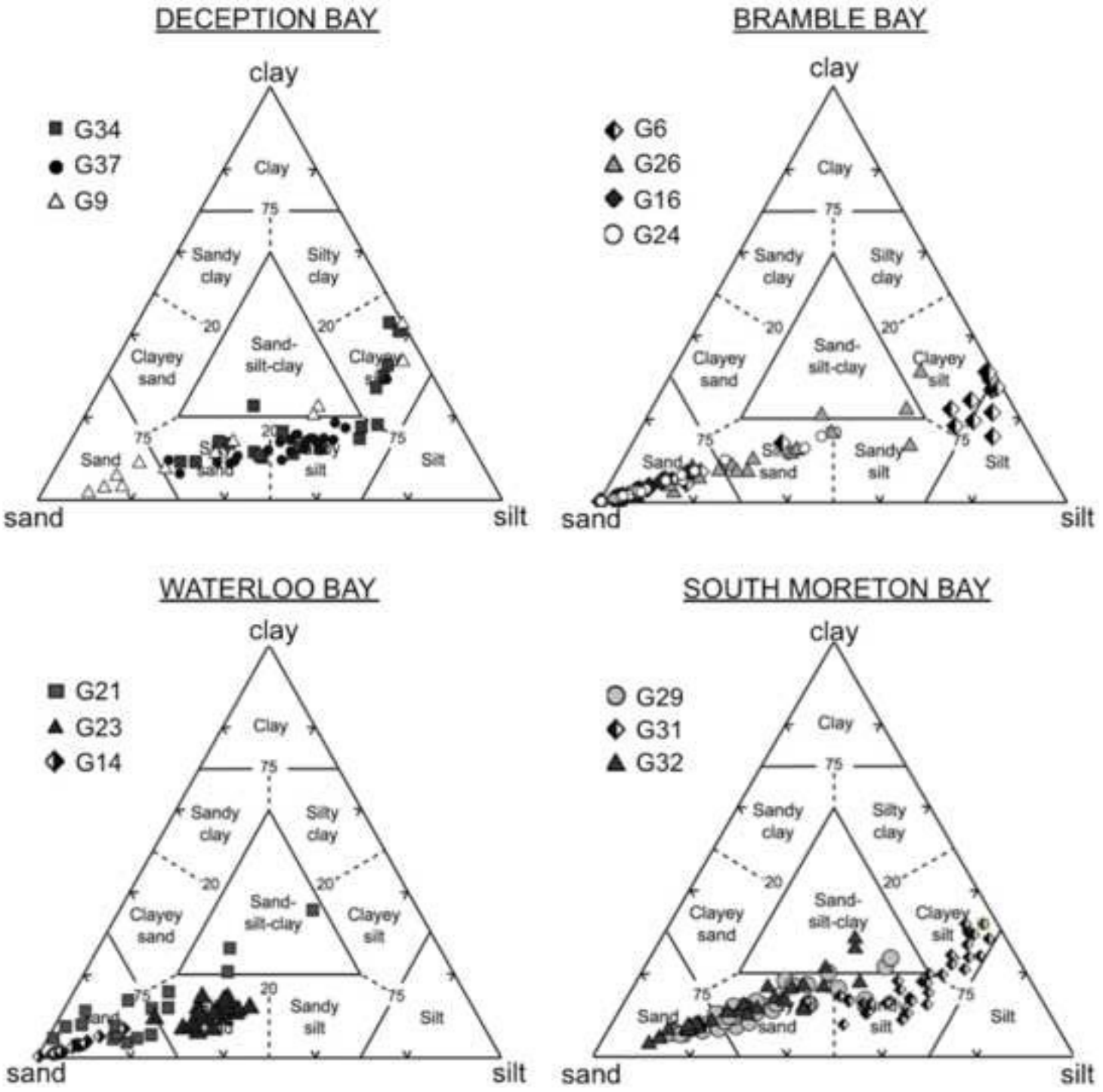
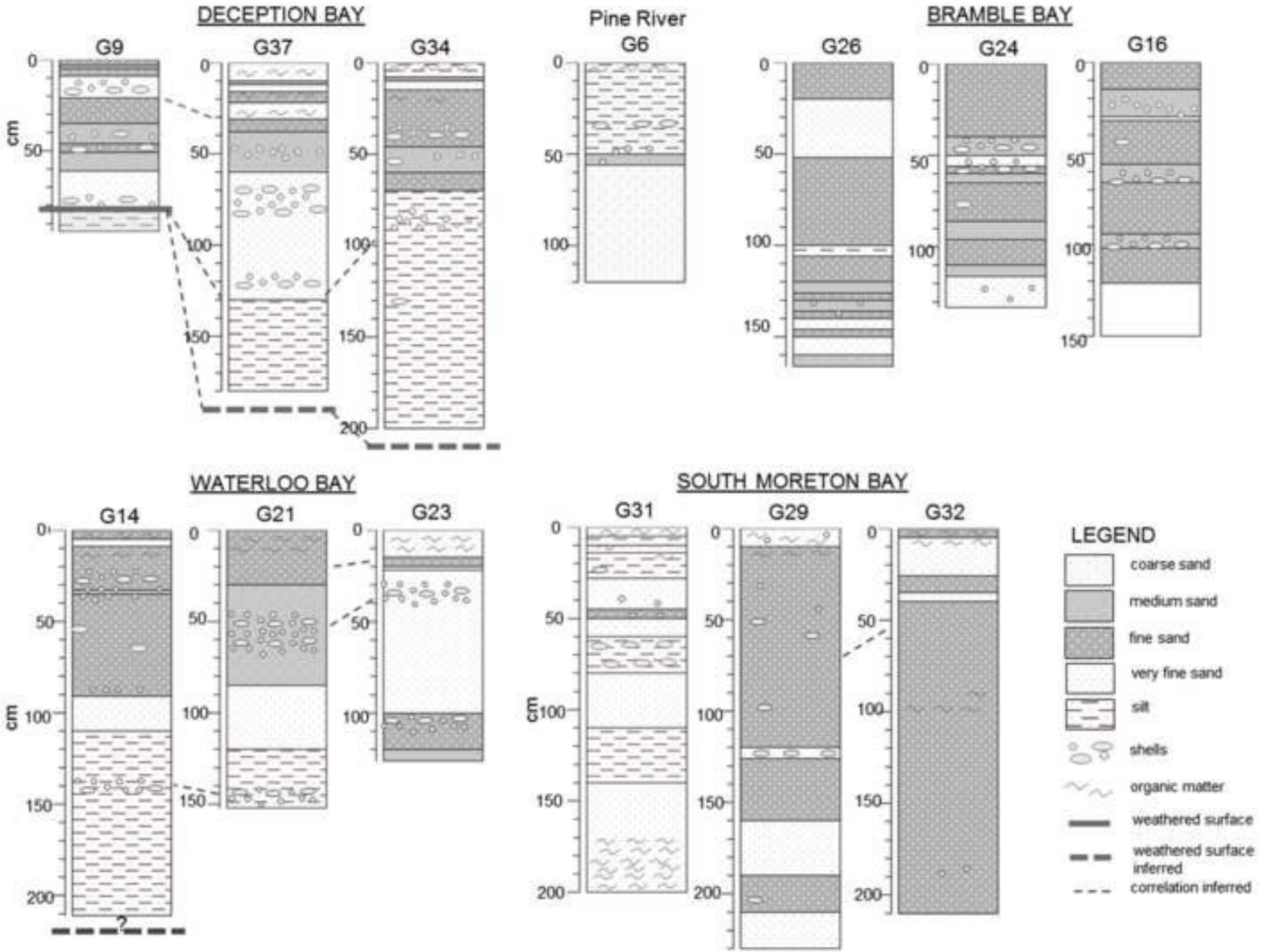


Fig 3



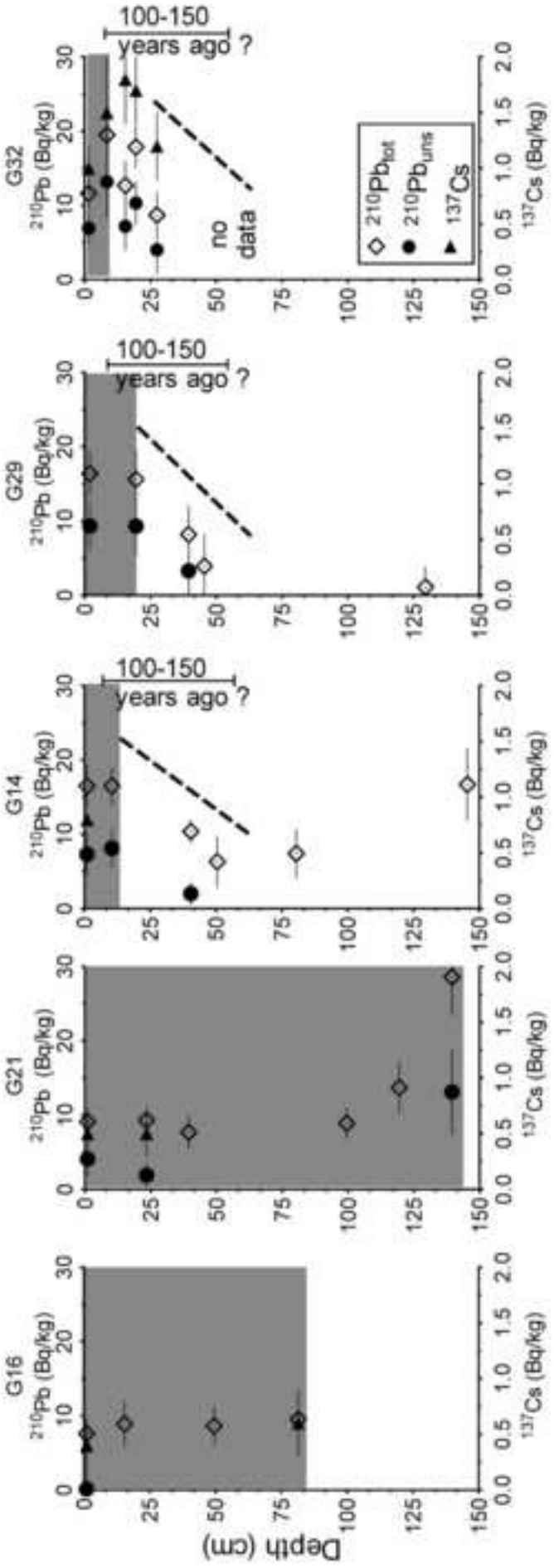
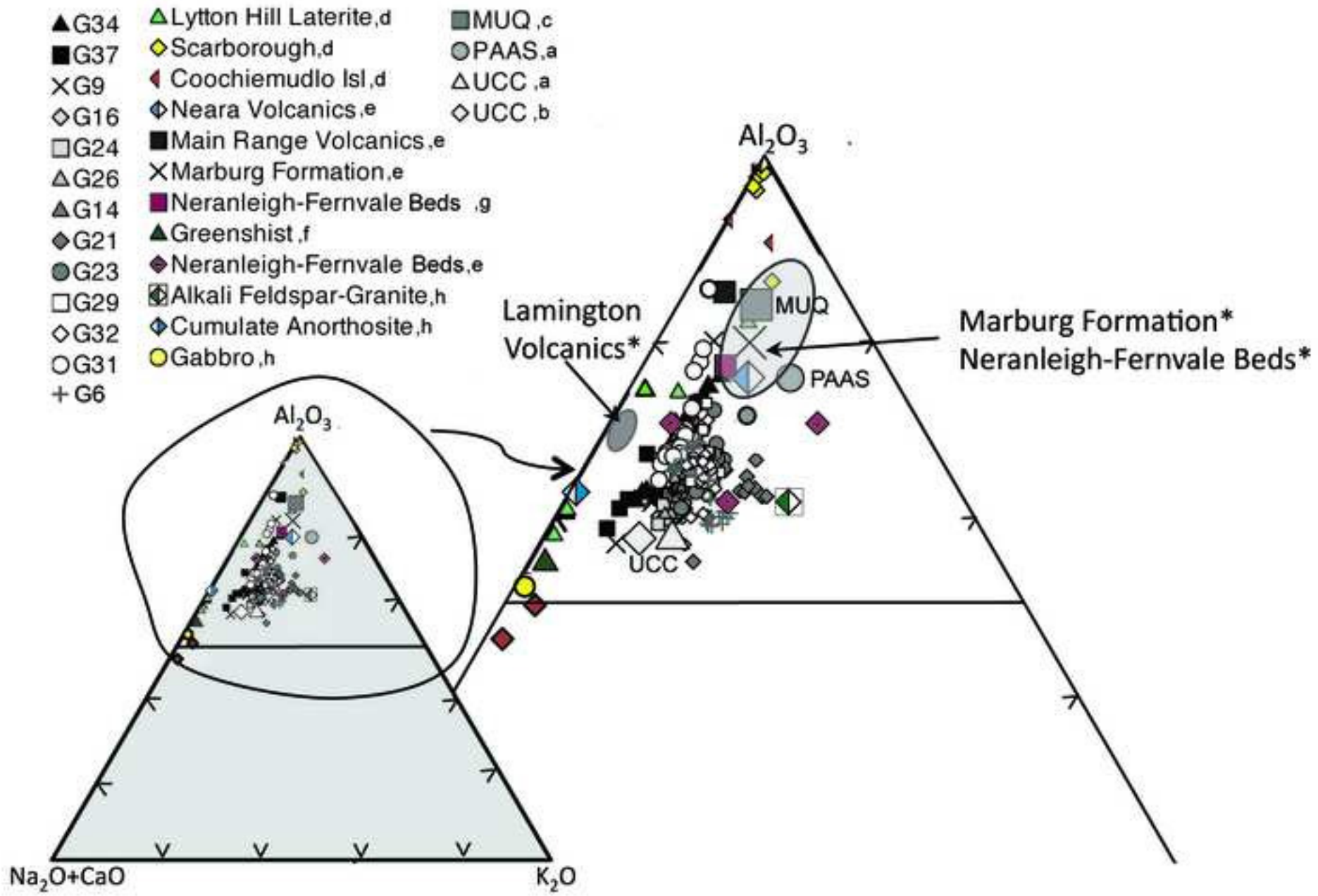

G32
G29
G14
G21
G16
100-150 years ago ?
no data
$^{210}Pb_{tot}$
$^{210}Pb_{uns}$
^{137}Cs
^{210}Pb (Bq/kg)
^{137}Cs (Bq/kg)
Depth (cm)

Fig 4

Fig 5



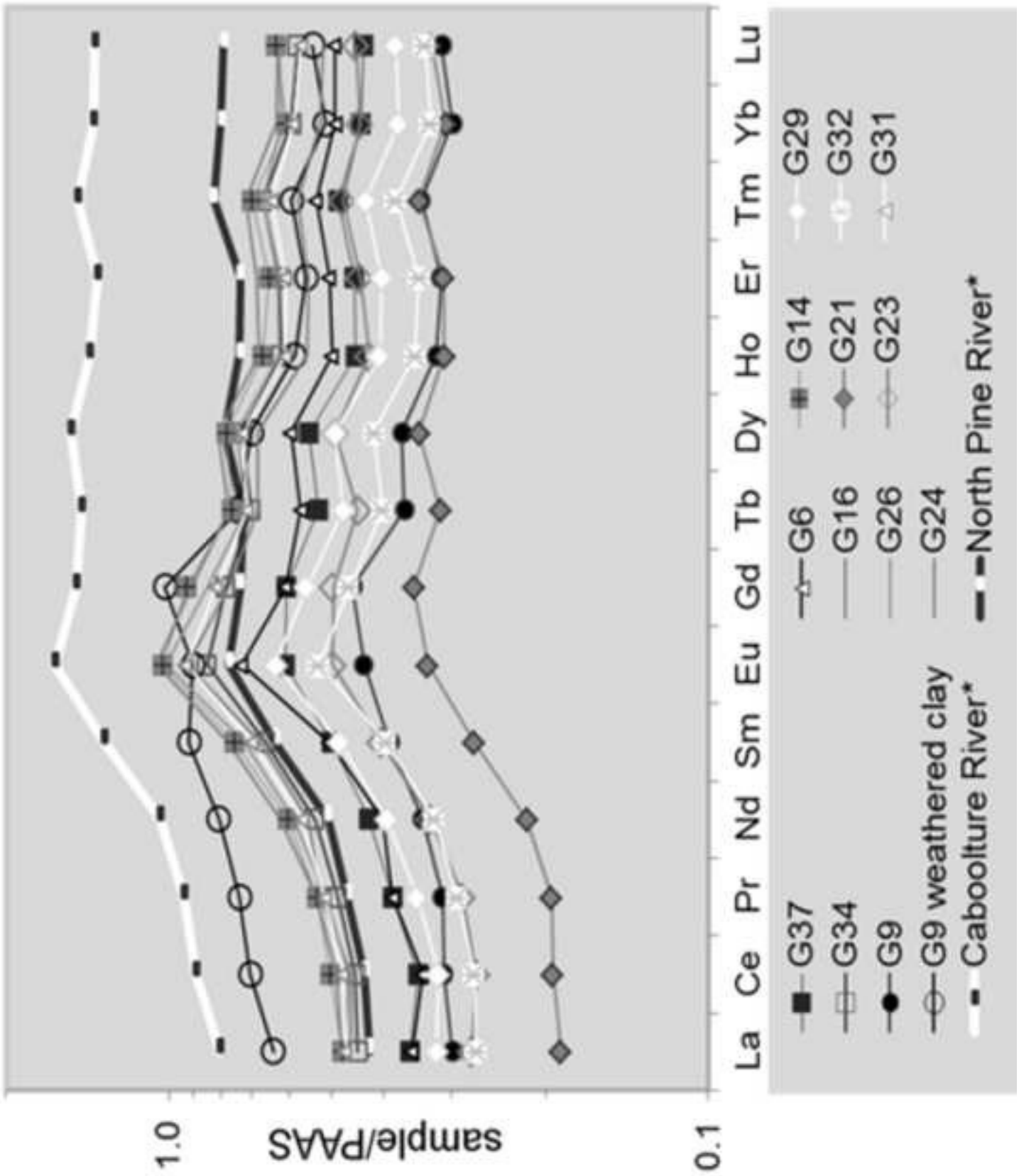

sample/PAAS
1.0
0.1
La Ce Pr Nd Sm Eu Gd Tb Dy Ho Er Tm Yb Lu
G37
G34
G9
G9 weathered clay
Caboolture River*
G6
G16
G26
G24
G14
G21
G23
North Pine River*
G29
G32
G31

Fig 6

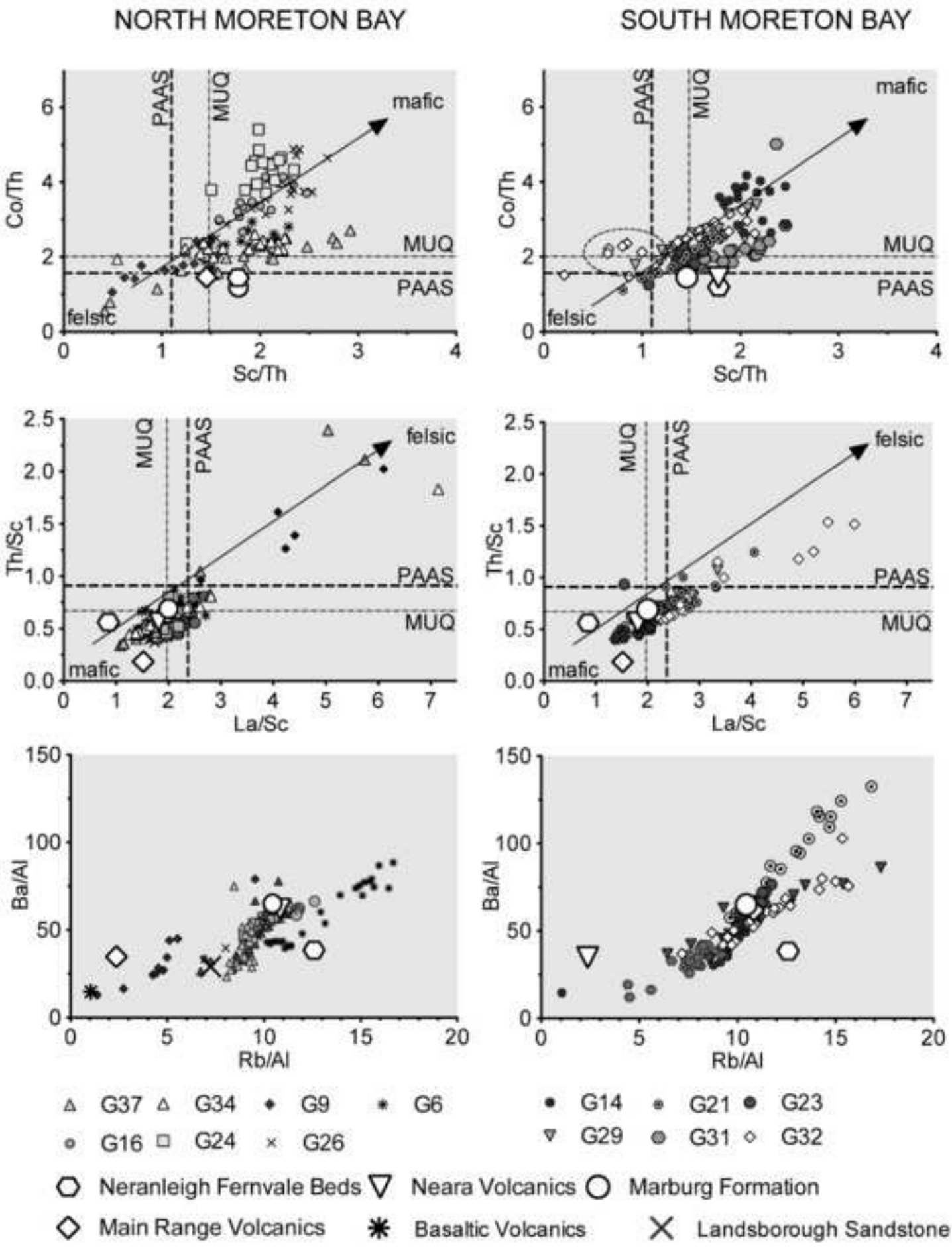


Fig 8

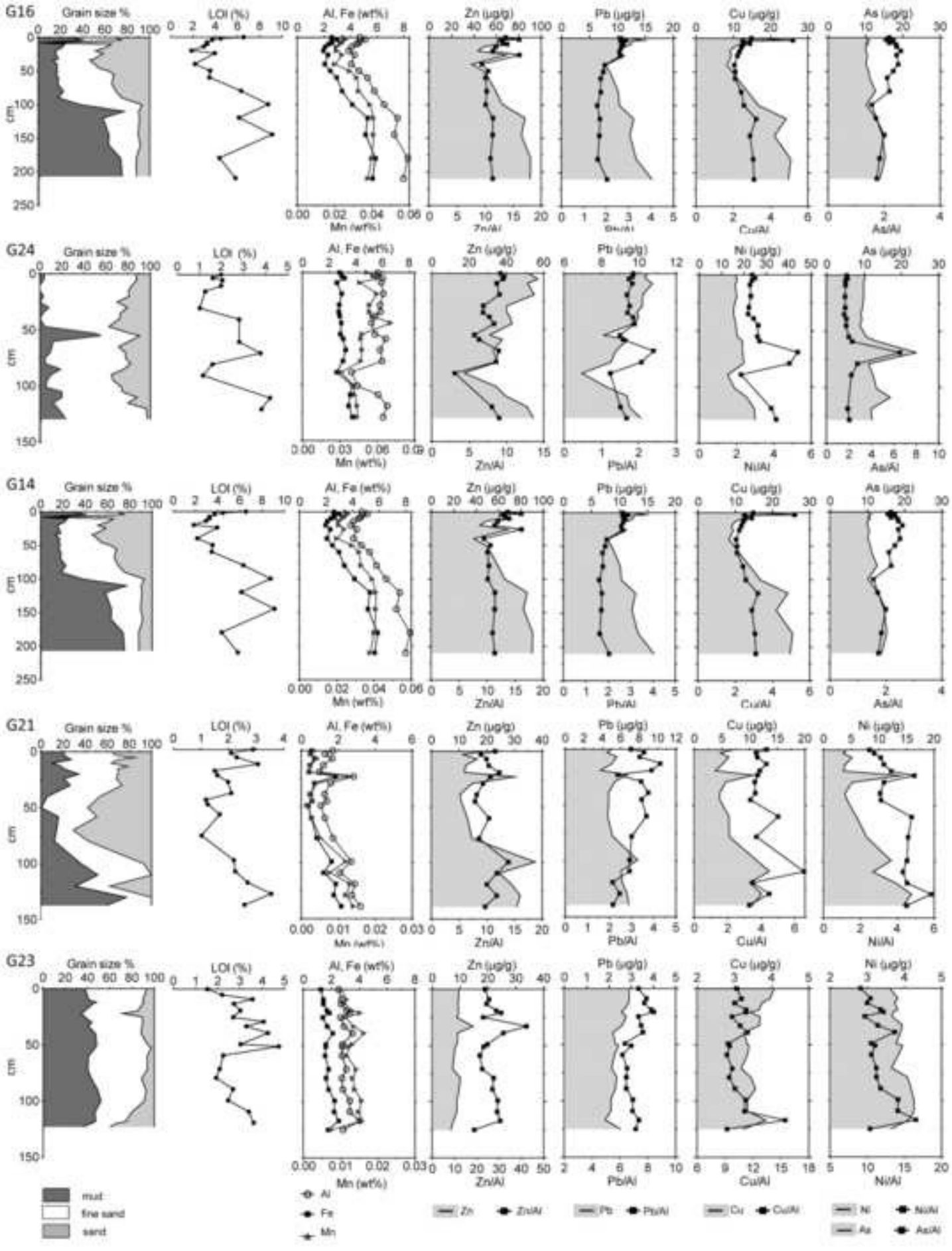
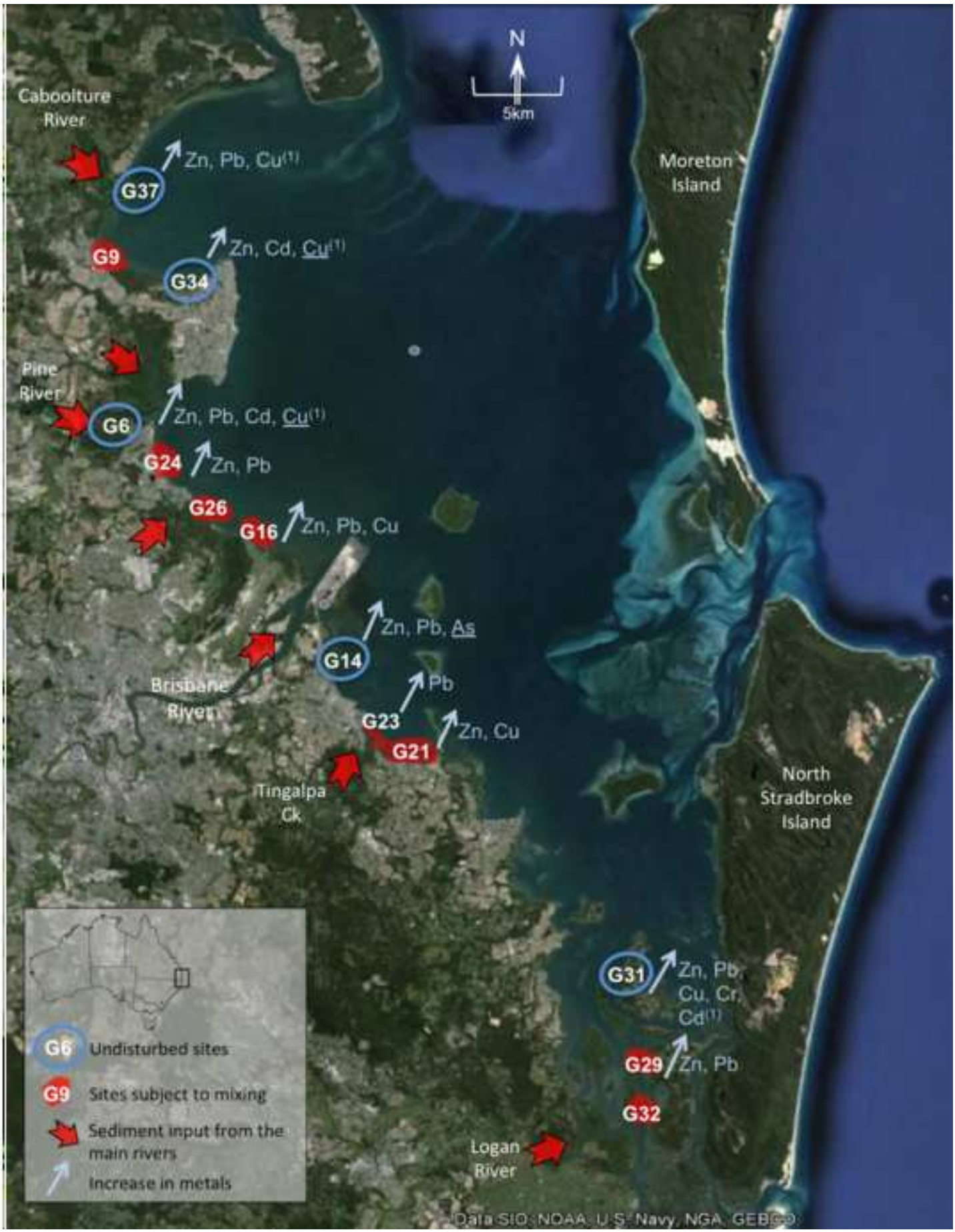


Fig 9



Core	G37	G34	G9	G6	G24
Al	3.5 ± 1.9	4.7 ± 2.6	2.3 ± 0.8	4.7 ± 1.4	5.6 ± 0.7
	6.8 - 0.7	9.2 - 0.9	4.2 - 1.2	6.5 - 3.0	6.4 - 3.7
Fe	2.2 ± 1.1	3.2 ± 1.7	1.2 ± 0.6	2.3 ± 1.5	3.0 ± 0.4
	4.1 - 0.5	6.1 - 0.6	2.3 - 0.4	4.1 - 0.7	3.7 - 2.6
Mn	0.0 ± 0.0	0.03 ± 0.02	0.0 ± 0.01	0.0 ± 0.0	0.1 ± 0.01
	0.0 - 0.0	0.1 - 0.01	0.03 - 0.005	0.0 - 0.0	0.1 - 0.03
K	0.6 ± 0.2	0.8 ± 0.4	0.5 ± 0.1	1.3 ± 0.1	1.5 ± 0.2
	1.0 - 0.2	1.3 - 0.2	0.6 - 0.3	1.5 - 1.2	1.6 - 1.1
Na	0.9 ± 1.0	1.0 ± 0.7	1.0 ± 0.2	0.9 ± 0.2	1.5 ± 0.2
	3.7 - 0.1	2.6 - 0.2	1.3 - 0.7	1.3 - 0.6	1.8 - 1.1
Ca	1.1 ± 0.6	1.3 ± 0.6	0.4 ± 0.4	1.6 ± 0.5	2.0 ± 0.3
	2.1 - 0.3	2.4 - 0.4	1.5 - 0.1	2.4 - 1.0	2.4 - 1.3
Ti	0.4 ± 0.2	0.5 ± 0.2	0.2 ± 0.1	0.4 ± 0.2	0.5 ± 0.1
	0.6 - 0.1	0.8 - 0.2	0.4 - 0.1	0.7 - 0.1	0.7 - 0.2
Sc	8.8 ± 5.2	10.2 ± 6.3	4.2 ± 1.7	8.7 ± 5.5	9.1 ± 1.5
	17.3 - 1.5	20.4 - 1.0	7.3 - 1.7	15.9 - 2.7	10.9 - 5.5
Co	8.6 ± 4.3	12.3 ± 5.7	7.2 ± 3.4	10.5 ± 5.7	19.2 ± 2.1
	15.6 - 2.5	20.3 - 3.5	13.9 - 3.7	17.7 - 3.7	22.0 - 12.7
Ni	14.6 ± 8.3	21.1 ± 11.9	7.6 ± 2.8	15.1 ± 10.0	17.7 ± 3.2
	29.8 - 2.7	40.6 - 3.7	12.5 - 4.1	28.5 - 3.9	24.9 - 12.6
Cu	10.3 ± 5.7	15.5 ± 8.2	5.1 ± 1.4	12.8 ± 9.2	9.4 ± 3.6
	20.3 - 2.0	30.1 - 4.3	7.8 - 2.9	27.1 - 2.9	22.4 - 7.1
Zn	30.2 ± 15.4	47.2 ± 26.7	22.4 ± 5.9	48.9 ± 33.2	42.7 ± 10.6
	56.3 - 5.5	103.0 - 10.4	32.1 - 15.7	94.4 - 14.9	57.0 - 17.3
As	11.2 ± 3.4	14.0 ± 6.1	6.9 ± 1.8	9.0 ± 4.7	11.8 ± 3.7
	17.2 - 4.8	29.0 - 5.9	9.3 - 3.7	16.4 - 2.9	24.0 - 8.9
Rb	29.4 ± 13.3	41.1 ± 21.9	21.2 ± 4.1	58.1 ± 8.9	57.3 ± 6.3
	56.2 - 7.7	74.6 - 7.9	27.1 - 12.8	74.1 - 46.5	66.7 - 41.0
Sr	100.1 ± 73.9	117.6 ± 62.7	77.2 ± 22.8	147.9 ± 26.4	206.6 ± 21.2
	256.7 - 18.8	238.7 - 33.4	133.5 - 45.8	190.1 - 114.7	235.9 - 136.1
Cd	0.07 ± 0.03	0.10 ± 0.07	0.04 ± 0.01	0.07 ± 0.04	0.06 ± 0.02
	0.12 - 0.02	0.35 - 0.02	0.06 - 0.03	0.15 - 0.03	0.12 - 0.04
Ba	129.2 ± 44.5	160.9 ± 55.4	147.1 ± 32.0	254.7 ± 18.7	316.5 ± 33.4
	204.2 - 55.7	240.4 - 70.1	224.4 - 106.2	295.2 - 213.9	357.6 - 230.7
La	13.6 ± 4.4	16.9 ± 6.5	12.6 ± 4.8	13.5 ± 7.3	18.2 ± 2.1
	23.6 - 7.4	27.1 - 7.1	24.3 - 7.1	22.9 - 5.2	20.7 - 12.0
Ce	27.6 ± 9.3	36.1 ± 13.8	28.0 ± 11.4	27.1 ± 15.7	39.0 ± 4.1
	48.5 - 14.0	57.1 - 14.7	56.6 - 15.7	47.2 - 9.5	44.3 - 27.3
Pr	3.4 ± 1.2	4.3 ± 1.7	3.1 ± 1.4	3.4 ± 2.0	4.7 ± 0.6
	6.1 - 1.7	7.0 - 1.6	6.6 - 1.7	6.0 - 1.2	5.4 - 3.1
Nd	13.6 ± 5.0	17.2 ± 7.0	12.3 ± 5.3	12.8 ± 7.7	19.9 ± 2.5
	24.5 - 6.3	27.7 - 6.3	25.8 - 6.7	22.7 - 4.4	22.7 - 13.3
Sm	2.8 ± 1.1	0.9 ± 0.4	2.5 ± 1.0	0.8 ± 0.4	1.2 ± 0.1
	5.0 - 1.2	1.6 - 0.3	5.1 - 1.3	1.3 - 0.4	1.3 - 0.8
Eu	0.7 ± 0.3	3.7 ± 1.6	0.5 ± 0.2	2.8 ± 1.7	4.3 ± 0.5
	1.3 - 0.2	6.1 - 1.3	1.0 - 0.3	5.0 - 1.0	5.0 - 2.9