Chemical signatures of palaeotsunamis: A forgotten proxy?

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A range of diagnostic criteria are required to help identify palaeotsunami deposits. Although chemical signatures have long been used as indicators of palaeosalinity in sedimentary sequences, their use in the studies of palaeotsunami deposits is rarely applied and is yet to be widely recognised. The chemical composition of interstitial water in coastal ponds and sedimentary sequences in coastal marshes and lagoons has been used successfully as an indicator of tsunami inundation. Chemical signatures can also provide clues about the landward limit of runup of a tsunami, beyond the area of sediment deposition. Like most palaeotsunami proxies however, the preservation of chemical signatures varies with environmental conditions.

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1. Introduction

In the last 25 years, there have been a large number of publications dealing with the study and identification of palaeotsunami deposits, mostly from the Pacific Northwest (e.g., Atwater, 1987; Clague and Bobrowsky, 1994), Europe (e.g., Dawson et al., 1988), Japan (e.g., Minoura and Nakaya, 1991), Hawaii (e.g., Moore et al., 1994), Australia (e.g., Dominey-Howes, 2007) and New Zealand (e.g., Goff et al., 2001).

Much has been written and debated about the diagnostic signatures or criteria for historical and palaeotsunamis (Goff et al., 2001; Dominey-Howes et al., 2006; Dominey-Howes, 2007; Kortekaas and Dawson, 2007). These have been refined over the last few years, but their use and perceived relevance reflect a strong bias in the literature related to the expertise of those involved in the research. Researchers have access to a wide range of proxies, including geomorphological, stratigraphical, sedimentological, archaeological, anthropological, palynological, macro- and micropalaeontological evidence to identify historical and palaeotsunami deposits (Goff et al., 2001; Dominey-Howes et al., 2006; Dominey-Howes, 2007). It should however be noted that all criteria have never been reported from one site, and neither are they all found in one single deposit. Therefore, the lack of one or more proxies (e.g., foraminifera or diatoms, etc.) should not be taken as unique evidence to refute the tsunamiogenic origin of a specific deposit (Goff et al., 2001; Dominey-Howes et al., 2006; Goff et al., in press).

Unlike many proxies, chemical signatures have rarely been used to help identify palaeotsunami deposits and are even omitted from published lists of diagnostic criteria, such as the recent one by Bryant (2008). Like other proxies (e.g., microfossil assemblages), chemistry alone will not provide a definite answer to the origin of a deposit (tsunami vs. storm surge). It will however provide another piece of the puzzle, and in conjunction with similar tools out of the ‘toolkit’, it can help identify palaeotsunami deposits. The occurrence of specific diatom assemblages, for example, can indicate a marine source and help pinpoint the landward extent of seawater inundation, also providing information about the depositional process (Hemphill-Haley, 1996). Similar analogies have been drawn using foraminifera (Dominey-Howes, 1996; Dominey-Howes et al., 1998; Mamo et al., 2009), and microfossils have been used together with other signatures to help identify palaeotsunami deposits and also to attempt to differentiate between known tsunami and storm deposits (e.g., Kortekaas and Dawson, 2007).

The aim of this paper is to demonstrate the usefulness of chemical signatures for the identification of palaeotsunami deposits. Although reports of chemical indicators of palaeosalinity in sedimentary sequences date back many years, there has been some reluctance to adopt them for studies of palaeotsunami deposits.

2. Chemical signatures of marine influence in sedimentary sequences

High sulphur (S) concentrations have long been associated with the brackish/marine depositional environments of peat (Casagrande et al., 1977), due to the higher availability of sulphate in seawater compared to freshwater. Boron (B) had also been identified as a palaeosalinity indicator in coal (Swaine, 1962) and shales (Bobor and Gluskoter, 1973). However, an increase in B may be linked to an increase in the proportion of clay (Levinson and Ludwick, 1966) or organic matter content (e.g., Ward, 1980; Goodarzi, 1988; López-Buendía et al., 1999) rather than increasing salinity, thus complicating the interpretation. Dominik and Stanley (1993)
demonstrated the usefulness of S and B as palaeosalinity indicators in sedimentary sequences from the Nile Delta, although they showed that normalisation of B with beryllium (Be) was required to account for grain size variations. Chen et al. (1997) found that strontium (Sr), barium (Ba), Sr/Ba ratios, B and B/Ga (Ga = gallium) ratios were valuable lithofacies indicators of marine transgressions, mostly due to the higher concentrations of Sr, Ba and B in seawater compared to freshwater.

López-Buendia et al. (1999) however rejected Sr as a palaeosalinity indicator because of its occurrence in calcite and aragonite, but concluded from their study of a coastal marsh and an inland bog that the principal chemical indicators of palaeosalinity were B−/bromine (Br) ≈ iodine (I) ≈ sodium (Na) > S−/germanium (Ge) > uranium (U).

Relative maxima in the concentrations of Cl, I and Br found in cores from a freshwater peatland situated c. 10 km from the coast were attributed to sea spray, possibly associated with a major storm event in the region about 2500 years ago (Chagué-Goff and Fyfe, 1996). A tsunami however cannot be ruled out. The low levels of soluble compounds in the peat also suggested that although Cl, I and Br were probably first deposited as salts from sea spray, they were subsequently incorporated within the organic fraction of the peat (Chagué-Goff and Fyfe, 1996). Biester et al. (2004) also show that Cl, I and Br are transformed to organohalogens during peat decomposition, and that they are therefore retained. The lack of a concurrent Na relative maximum in the peatland, which would be expected since it is a key component of sea spray, has been attributed to its high solubility in the peat (Damman, 1978; Shotyk, 1988).

Stable isotope data in shells and carbonates have also been used as indicators of marine vs. freshwater origin. Increases in δ13C and δ18O in growth bands of fossil gastropod shells, although also being influenced by temperature and evaporation, were found to reflect increases in salinity, related to decreases in freshwater inflow (e.g. Ingram et al., 1998). Shell δ13C can also be used as a proxy for salinity (e.g., McConnaughey and Gillikin, 2008), despite a number of factors affecting carbon fractionation.

3. Use of normalisation

The concentration of many trace elements has been found to be strongly dependent upon the grain size distribution (e.g., Ackermann, 1980). To aid comparison, other elements, such as caesium (Cs), rubidium (Rb), lithium (Li), aluminium (Al), titanium (Ti) and iron (Fe), which are not dependent on grain size, have often been used in order to correct for grain size distribution. Ackermann (1980) used Cs for correction, as Cs in quartz, the main constituent of sand, is about 10 times lower than in clay minerals, while Al and then Li were the elements of choice by Din (1992). However, Van der Weijden (2002) warns against normalisation using a common divisor, because it can cause spurious correlations between normalised elements. He shows however that Al can still be used to provide a realistic estimate of the elemental content in organic matter.

4. Chemical signatures to help identify palaeotsunami deposits

The study of chemical indicators associated with palaeotsunami inundations is in its infancy. Minoura and Nakaya (1991) and Minoura et al. (1994) were the first researchers to use geochemistry in the study of modern (1983 Japan Sea Tsunami) and historical tsunami deposits in northeast Japan. This followed their observations that the 1983 tsunami resulted in the flooding of coastal marshes and precipitation of calcite. Furthermore, seawater remained in ponds for several months after tsunami inundation, as shown by measured increases in salinity. Analysis of interstitial water revealed relative maxima in chloride (Cl−), calcium (Ca2+) and magnesium (Mg2+), and low Mg2+/Ca2+ ratios associated with the sandy layers in the mud sequences of the coastal marshes. Relative maxima of sulphate (SO4 2−) and sodium (Na+) were however only observed at some of the sites (Minoura et al., 1994). Increases of Ca2+ and Mg2+ were attributed to the reaction of seawater with the carbonic acids in the pond water and also to skeletal carbonate from marine shells carried in by the tsunami (Minoura et al., 1994). Abundant brackish and marine diatoms in the sandy layers confirmed the marine influence (Minoura et al., 1994), and correlation with historical events was obtained with 13C and/or Pb210 dating. Minoura and Nakaya (1991) concluded that geochemical signatures could also be used to identify seawater inundation by smaller tsunamis that did not deposit sediment. It is interesting to note however, that a comparison of their two studies shows that not all the expected ‘seawater’ elements, such as Na+ and SO4 2−, exhibited maxima in concentrations in the sand layers identified as tsunami deposits in all sites investigated.

Sediment chemistry, as opposed to the chemistry of interstitial water, has since been successfully used to identify palaeotsunami deposits in conjunction with other proxies (Chagué-Goff and Goff, 1999; Goff and Chagué-Goff, 1999; Chagué-Goff et al., 2002; Goff et al., 2004b; Schlichting and Peterson, 2006; Nichol et al., 2007).

Goff and Chagué-Goff (1999) reported relative maxima in Fe and/or S, and organic matter, associated with units of fine sediments (clay and/or silt) in New Zealand tidal marshes. These were tentatively attributed to tsunami inundations and chronologically linked with local fault ruptures. Mineralogical analysis showed that the concurrent maxima in Fe and S were due to the presence of pyrite, and that high Cl concentrations reported in one Fe–S–rich layer in at least one of the sedimentary sequences were attributed to the presence of halite (Chagué-Goff and Goff, 1999). Although S occurrence as pyrite represents a post-depositional process, S and Fe can be used as indicators of palaeosalinity and flooding frequency, also because Fe-rich sediments are more likely to fix S than Fe-poor sediments (Thomas and Varekamp, 1991). Much of the S can also occur in association with organic matter, reflecting the uptake of SO4 2− from seawater (Dominik and Stanley, 1993; Chagué-Goff et al., 2002). Halite has been reported at the surface of salt marshes and its formation attributed to evaporation at the marsh surface and oversaturation of Na and Cl (e.g., Chagué-Goff et al., 2001; López-Buendia et al., 2007). The absence of halite in underlying layers was attributed to dissolution as a result of flushing or capillary action (Chagué-Goff et al., 2001), although evidence of biogenic halite in association with halophytic plants has been reported and can explain its low mobility over periods over several thousand years (López-Buendia et al., 2007).

In a study of a coastal lagoon in northern Hawke’s Bay, New Zealand, a fining-inland gravel to sand unit was interpreted as a 6300 years BP palaeotsunami (Chagué-Goff et al., 2002). A marine diatom assemblage was found to coincide with small relative maxima in Na/Rb, although S data were equivocal. The latter was possibly caused by the coarse size fraction and low organic matter, which are not conducive to the uptake and preservation of S (Chagué-Goff et al., 2002).

A fining-upward sequence reported from both the seaward and landward side of Okarito Lagoon (South Island, New Zealand), was characterised by relative maxima in Fe, S, Ti, Sr, Ba, Ca and Na, and abundant marine and brackish diatoms (Fig. 1) (Goff et al., 2004b; Nichol et al., 2007). Its chemical composition indicated saltwater inundation, either directly as sediment mixed with seawater (Fe, S, Sr, Ba, and Na) or the presence of inwashed shell material (Ca), the latter only seen on the seaward side. Ti is not a salinity indicator per se, but an indicator of high-energy deposition causing iron sand enrichment during inundation (Goff et al., 2004b; Nichol et al., 2007). Marine (Polyhalobian) and fully-brackish (Mesohalobian) diatoms also showed a maximum in the fining-upward sequence on the seaward side of the lagoon (Fig. 1), and a marked increase in broken valves in this interval suggests reworking by high-energy flows. This comprehensive suite of sedimentological, geochemical and micro-palaeontological evidence allowed the researchers to infer a tsunamigenic origin for the fining-upward sequence, which was dated at 1826 AD, based on Pb210 dating of the landward sequence.

Schlichting and Peterson (2006) used Br (bromine) as marine marker coinciding with an increase in marine diatom abundance to confirm the marine origin of an anomalous sand sheet identified as a palaeotsunami deposit in Oregon using sedimentological proxies.
Stratigraphy, organic matter, geochemistry and diatom data from two cores (OK10 and OK6a) taken on the landward, and seaward sides, respectively, of Okarito lagoon, West Coast of South Island, New Zealand. The fining-upward sequence, as determined by particle-size analysis, exhibited a sharp basal contact and was characterised by relative maxima in S, Fe, Ba, Sr, Na and Ti at the base, as determined by X-ray fluorescence (modified after Goff et al., 2004b). Diatom data also suggest a marine origin and high-energy flows. Dating control was provided by Pb$^{2+}$ in core OK10. The sequence was characterised by a very low organic content (LOI). Sedimentological, geochemical and micropaleontological evidence confirmed the tsunamigenic origin of the fining-upward sequence (the area between the two horizontal dashed lines defines the approximate boundaries of the tsunami deposit). Details about the methodology can be found in Goff et al. (2004b).
Sediment chemistry has also been successfully used to refute an inferred palaeotsunami deposit. Dominey-Howes et al. (2006) found that the chemistry of a sequence reported as being deposited by a tsunami (Bryant, 2001) was that of a weathered soil, and did not contain any evidence of saltwater inundation. This conclusion was corroborated by the lack of sedimentological and stratigraphic indices usually found in deposits laid by tsunamis (Dominey-Howes et al., 2006).

Chemical signatures of saltwater inundation may also be recorded beyond the landward limit of sediment deposition, where no sedimentological evidence can be found (Minoura and Nakaya, 1991; Chagué-Goff and Fyfe, 1996). Salt residues were observed following the 17th July 2006 Java tsunami, on the landward side of a rice paddy field (Figs. 2 and 3), 35 m further inland than the maximum extent of sediment deposition of 720 m (Fritz et al., 2007; J. Goff, pers. comm., 2009), and also more recently in a wetland in southern Samoa during the post-tsunami survey following the South Pacific Tsunami of 29 September 2009 (pers. observation). Similarly, marine diatom assemblages have been reported beyond the landward limit of sand or silts deposited by a tsunami (Hemphill-Haley, 1996). Therefore, both chemical and microfossil proxies can be used to provide clues about the landward limit of tsunami inundation.

Like other proxies, there is no simple recipe for using geochemistry as a tool to identify palaeotsunami deposits. As reported above, not all elements thought to indicate saltwater inundation occur in all deposits, and the absence of one or other element can be due to a range of processes, such as the lack of uptake and/or preservation as a result of sediment particle size and composition, and post-diagenetic processes leading to dilution and/or dissolution of the saltwater indices. Sand layers in peaty deposits might also allow for circulation of oxygenated sub-surface flows after burial, possibly resulting in the formation of oxides. These can however be subjected to post-depositional oxidation–reduction processes, related to biochemical processes and/or various stages of oxygenation due to changes in water table and moisture regimes (e.g. Phang et al., 2004). Iron oxide stains have indeed been reported by Tuttle et al. (2004), although they occurred in the recent 1991 storm deposit, as opposed to the 1929 tsunami deposit. Thus, geochemical signatures acquired through marine inundation and/or formation of post-depositional minerals may be lost through subsequent leaching and dissolution. Mobilisation and redistribution of chemicals might also complicate the interpretation of chemical signatures left by marine inundation. Similar taphonomic problems are faced for microfossil proxies (e.g. Mamo et al., 2009).

5. Chemical signatures of palaeostorms or palaeotsunamis?

Distinguishing between historical and palaeotsunami and storm deposits is not a simple task. The task is further complicated by equivocal proxies and only a few studies have attempted to differentiate between these two types of high-energy events (Nanayama et al., 2000; Goff et al., 2004a; Tuttle et al., 2004; Kortekaas and Dawson, 2007; Morton et al., 2007). The question thus arises, whether chemical signatures could be used to distinguish palaeotsunami and palaeostorm deposits. A number of elements reflect a marine influence due to their higher concentration in seawater than in freshwater. As storms are most often associated with heavy rainfall and a marked terrestrial component, and assuming that we are dealing with the same site within a single stratigraphic sequence and a similar type of sediment, one could speculate that the marine signal might...
be expected to be diluted in a storm deposit, as opposed to that of a tsunami, and mixed with a terrestrial input. However, only further research, which could include stable isotope analysis, will be able to ascertain this, and again the evidence will have to be corroborated by other proxies.

6. Conclusions

The identification of palaeotsunami deposits relies on a wide range of diagnostic criteria, with each criterion adding a piece to the puzzle of the origin of the deposit. Chemical signatures have seldom been used as diagnostic criteria, while sedimentological and micropalaeontological proxies have tended to be the indicators of choice. Chemical signatures for palaeosalinity in peat, coal and sedimentary sequences can be applied to the study of palaeotsunamis, as long as the depositional environment is conducive to the uptake and preservation of saltwater indices. Elements, which have successfully been used as proxies for tsunami inundation are S, Cl, Na, Sr, Ba and Ca in sediments, as well as $\text{SO}_4^{2-}$, $\text{NO}_3^{-}$, $\text{Ca}^{2+}$ and $\text{Mg}^{2+}$ in interstitial water and Ti as indicator of high energy. Like other proxies, their use is not straightforward, but in conjunction with other indicators, they help identify palaeotsunami deposits. They can also fill a gap and provide the missing clue when other proxies are missing (e.g., foraminifera and/or diatoms) or provide equivocal results. Furthermore, like some micropalaeontological signatures they can be recorded beyond the landward limit of sediment deposition.

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