

GEOCHEMICAL EVIDENCE FOR THE RECENT CHANGES IN A SALT MARSH, CHEZZETCOOK INLET, NOVA SCOTIA, CANADA

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Sediment cores were collected from the salt marsh located in the West Head of Chezzetcook Inlet, and analyzed to determine the recent changes in geochemistry as a result of both natural and anthropogenic influence. ¹³⁷Cs dating was used to determine the accretion rates and shows that the sedimentation rates have been slightly higher in the high marsh than in the middle marsh for the last 30 years, probably as a result of increased terrestrial sediment influx. Elemental distribution is mainly related to mineral matter abundance and variety (e.g. mica, halite). However, elevated concentrations of Fe, Ni, Co, Mo and As near the surface of the marsh sequence at the landward edge of the marsh is attributed to anthropogenic input related to the recent road construction, while an elemental enrichment at about 1 m depth is attributed to diagenetic processes, which have resulted in elemental precipitation as sulfides at the boundary between the oxidized and reduced sediment layer.

On a collectionné des carottes de sédiment du marais salé situé à West Head de Chezzetcook Inlet, et on les a analysés en but de déterminer les changements géochimiques dus aux influences naturelles et anthropogéniques. La datation au ¹³⁷cesium a été usagée pour déterminer les taux d'accrétion, et a démontré que les taux de sédimentation ont été un peu plus élevés dans le haut marais que dans celui du milieu pendant les dernières 30 années, probablement à cause d'un plus grand envahissement de sédiment terrestre. La distribution d'éléments se rapproche surtout à l'abondance et à la variété de matières minérales (par ex., le mica, la halite). Tout de même, les concentrations du Fe, Ni, Co, Mo et As près de la surface de la terre en haut du marais se rapporte à l'entrée anthropogénique pendant la construction récente d'un chemin. L'enrichement d'éléments à un mètre de profondeur est dû aux processus diagénétiques qui ont effectué la précipitation élémentaire de sulfures à la borne entre les couches de sédiment oxydé et réduit.

Introduction

Salt marshes act as natural sinks for fine-grained sediments and associated trace metals, and are also well protected from physical disturbance such as bioturbation; thus they have the potential to preserve an undisturbed record of environmental changes (McCaffrey and Thomson, 1980). Sediment cores from salt marshes have therefore often been used to assess the extent of contamination and to reconstruct the history of pollution in coastal areas (e.g. McCaffrey and Thomson, 1980; Thomas and Varekamp, 1991; Varekamp, 1991; Valette-Silver, 1993; Zwolsman *et al.*, 1993; Cundy and Croudace, 1995; Calloway *et al.*, 1996; Cochran *et al.*, 1998). The stratigraphic record of ¹³⁷Cs fallout, which provides information about the recent sediment accumulation rates, has also been used successfully in a number of studies to determine lateral variations in salt marsh accretion and their response to natural and anthropogenic influences (e.g. Delaune *et al.*, 1978; Craft *et al.*, 1993; Cundy and Croudace, 1995; Williams and Hamilton, 1995; Calloway *et al.*, 1996; Orson *et al.*, 1998).

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Chezsetcook Inlet is one of the most extensively studied estuaries of the Canadian Atlantic coast, as it provides a record of the coastal evolution and sea-level changes for the past 4000 years (Scott, 1977a; 1977b; Scott, 1980; Scott and Medioli, 1980; Scott *et al.*, 1987; Orford *et al.*, 1991; Carter *et al.*, 1992; Jennings *et al.*, 1993; Shaw *et al.*, 1993; Scott *et al.*, 1995). Investigations of the environmental changes in the last 200 years include studies of the recent evolution of the gravel barrier at Story Head (Orford *et al.*, 1991; 1995), the development of salt marshes at close proximity to the barrier, with a 2000 year long record (Jennings *et al.*, 1993), and the morphological changes in the estuary (Scott, 1980).

However, little is known about the most recent environmental changes in the salt marsh at the head of the inlet. A new road (Highway 107) was constructed across the northern part of the salt marsh in the late 1980's and there was some concern about the impact of road construction on the salt marsh. This paper discusses the vertical and lateral variations in geochemistry in the salt marsh, as a result of natural and anthropogenic influence.

Material and methods

Site description

Chezsetcook Inlet is located about 25 km ENE of Halifax, on the eastern shore of Nova Scotia. The estuary started forming as a result of submergence of the Nova Scotia coast and relative sea-level rise (RSLR) about 4000 years ago (Scott *et al.*, 1995). A drill core in the West Head of the estuary shows the presence of freshwater peat, overlain by a gradational salt marsh sequence, which grades sharply into 10 m thick intertidal mudflats (Scott, 1977a, 1980). The area investigated (44°42'N; 63°15'W) is situated in the West Head (Fig 1), and is mostly an area of middle marsh with steep-sided channels, as shown by the vegetation cover (Scott and Medioli, 1980). The stratigraphic record suggests that salt marsh formation in the West Head of Chezsetcook Inlet was initiated approximately 200 years ago, probably as a result of increased sedimentation related to settlement of European immigrants in the area (Scott, 1980). Road construction in the late 1940's and early 1950's promoted an increase of marsh area in the inlet (Scott, 1980) and probably increased sedimentation rates in the salt marsh in the West Head.

Bedrock geology of the immediate area is dominated by quartzites, gneiss, greywackes and slates of the Goldenville Formation, Cambro-Ordovician age, which is part of the Meguma Group (McBride, 1978). Surface geology consists mainly of Quartzite Till, a stony till derived from local sources (Stea and Fowler, 1979).

Marsh vegetation consists primarily of *Spartina patens*, which is typical of middle marsh, with *Spartina alterniflora* in the lowest marsh elevations. In the high marsh, the vegetation is dominated by Cyperaceae and *Juncus gerardii* (Scott and Medioli, 1980).

Sample collection

Two long cores were taken with a Russian-type auger in the middle marsh (core CH1a; 2 m long) and high marsh (core CH2a; 0.90 m long) in 1991 (Figure 1). In 1993 four surface cores (maximum 0.20 m depth) were taken in the middle (CH1b and CH4) and high (CH2b and CH3) marsh. Cores CH3 and CH4 were taken close to the new highway built in 1985-87. Core CH2b was collected near the western tip of the West Head of Chezsetcook Inlet (same location as core CH2a), and core CH1b was taken on the south side of the median channel in the middle marsh, at the same location as core CH1a (Fig 1).

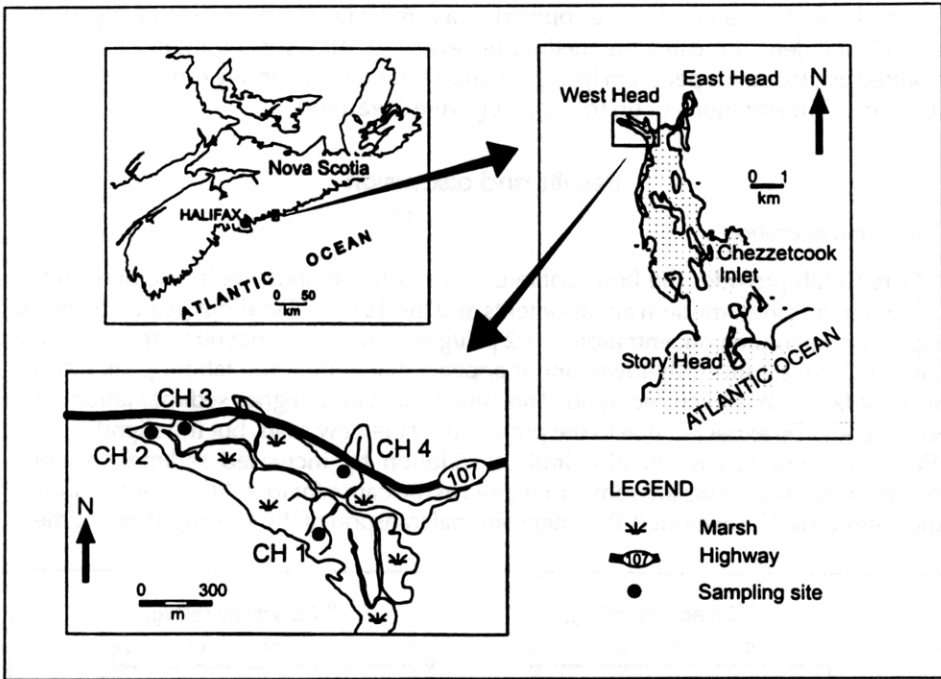


Fig 1 Location map of the salt marsh in the West Head of Chezzetcook Inlet, Nova Scotia, Canada, showing core locations. CH1 indicates the sampling site for cores CH1a and CH1b, while CH2 indicates the sampling site for cores CH2a and CH2b.

Chemical and mineralogical analyses

All samples were dried at 60°C overnight to constant weight, prepared for chemical analysis by crushing using an agate pestle and mortar and then divided into four splits. One split was analyzed by Instrumental Neutron Activation Analysis (INAA). Major and trace elements were determined on a dry weight basis, while the weight loss on ignition (LOI) was determined on a second split by ashing at 1000°C. Mineralogy of a third split of the long cores (CH1a and CH2a) was determined by X-ray diffraction (XRD), and the fourth split from the long cores was also analyzed using a Sulfur-Leco Determinator for total sulfur content.

¹³⁷Cesium dating

¹³⁷Cesium (half life = 30 years) is an anthropogenic radionuclide byproduct of nuclear weapon testing or nuclear accidents. It was first recorded in 1953, which corresponds to the onset of deposition in sediments as a result of atmospheric nuclear weapon testing, and peaked in 1963/64, which can be related to a maximum in atmospheric nuclear testing activity in 1962 followed by the Nuclear Test Ban Treaty of 1963 (Carter and Moghissi, 1977). ¹³⁷Cesium has a strong affinity for clay and fine organic particles and is therefore relatively immobile in sediments (e.g. Tamura, 1964; McHenry *et al.*, 1973).

Samples for ^{137}Cs analysis were counted on a gamma ray spectrometer having a solid state Ge (Li) detector, using the method of Lewis (1974). Concentrations of ^{137}Cs are reported as picocuries per gram (pCi/g) of dried sediment. Counting errors were about 10% and concentrations of up to 3.2 pCi/g were measured.

Results and discussion

Salt marsh accretion

Core CH2b provides the best profile of ^{137}Cs concentration, as it resembles other fallout records from modern environments and the 1964 ^{137}Cs fallout peak is resolved (Fig 2). A maximum concentration of 3.2 pCi/g is recorded. Concentrations of ^{137}Cs in core CH3 are significantly lower and it appears that neither the fallout peak nor the onset of fallout has been recorded. This might indicate a higher sedimentation rate, which could be expected due to the proximity of the new road, but the slightly lower ^{137}Cs concentrations might also indicate dilution by increased sediment supply. Bioturbation and mixing are sometimes cited as the cause for low ^{137}Cs concentrations, and they can not be excluded. Post-depositional transport of ^{137}Cs through the sediment

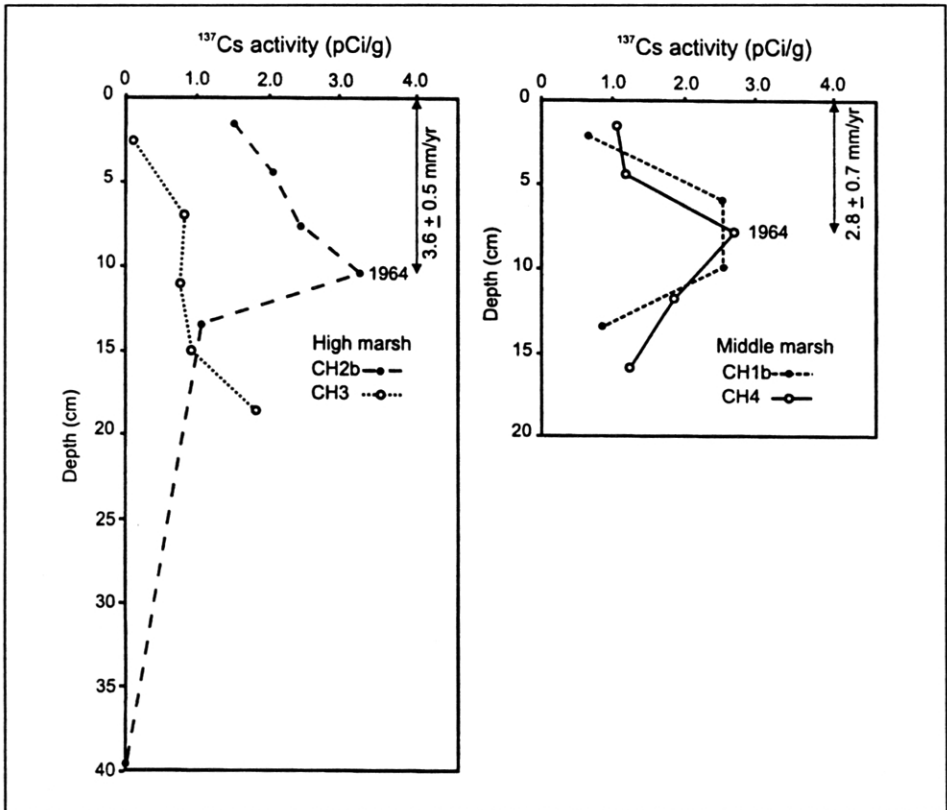


Fig 2 Distribution of ^{137}Cs with depth in cores CH1b, CH2b, CH3 and CH4, Chezzetcook Inlet. Estimated accretion rates are shown for the period 1964-1993. See Fig 1 for core sampling locations. Cores CH1b and CH2b were taken at sampling sites CH1 and CH2, respectively.

column via porewater diffusion or advection has also been reported (Ritchie and McHenry, 1990) and could account for the ^{137}Cs profile in core CH3.

The fallout profiles of cores CH1b and CH4 are more poorly resolved than that of core CH2b, but the peak fallout can still be resolved, in particular for core CH4, with a maximum ^{137}Cs concentration of 2.6 pCi/g (Fig 2). The data also suggest that the accretion rates show little variation between the south side and the north side of the salt marsh, the latter being at close proximity to the new highway. As it is not certain whether the onset of fallout in 1954 is detected in any of the cores, only the peaks in activity are used for determining the mean sediment accumulation rate for the period 1964-1993.

The depth of the fallout peak in cores CH1b, CH4 and CH2b suggests an estimated mean accretion rate of 2.8 mm/y in the middle marsh and 3.6 mm/y in the high marsh for the last 30 years. Data obtained from core CH3 are not considered here, due to possible mixing and post-depositional transport of ^{137}Cs . These data suggest that the accretion rates might be slightly higher in the high marsh than in the middle marsh. Previous studies have suggested that sedimentation rates are usually greater in the lower marsh than in the high marsh, due to more frequent flooding and greater sediment input (e.g. Thomas and Varekamp, 1991). However, in the present study, there might be an additional terrestrial sediment influx due to surface runoff and road building in the high marsh area, which could exceed the sediment input from tidal flooding (Chagué, 1994).

Sediment accumulation rates in the salt marsh also slightly exceed the mean rate of relative sea-level rise (RSLR) for the last 1000 years, which is 2 mm/y (Scott *et al.*, 1995). Evidence for the last 200 years also suggests that accretion rates have exceeded RSLR, as the salt marsh in West Head of Chezzetcook Inlet has evolved from low to high marsh (Scott, 1977a; 1980). Increased terrestrial sediment input and surface runoff caused by human activity is likely to have contributed to the slightly higher sediment accumulation rate. Although the number of cores investigated is limited, this study suggests that the recent road construction does not appear to have resulted in a considerable increase in sedimentation rate on the north side of the salt marsh.

Geochemistry and mineralogy

Weight loss on ignition (LOI) averages 42% and 41% in cores CH1a and CH2a, respectively (Fig 3). Sampling at finer intervals in the shallow cores shows that LOI decreases from the surface (maximum of 77%) to 15-20 cm depth (minimum of 37%) and averages 44% (Chagué, 1994). The higher LOI at the surface is attributed to the abundance of fresh plant material, while the decrease with depth is due to degradation of organic material and trapping of fine inorganic material by the marsh vegetation.

The distribution of Al, Mn, K and Mg in cores CH1a and CH2a is similar and shows a negative relationship with LOI distribution (Fig 3). This suggests that these elements are associated with the inorganic fraction of the marsh sediments, mainly in clay minerals, such as mica (illite) and chlorite, as well as in feldspars (as determined by XRD; Chagué, 1994).

Sodium and Cl concentrations show a marked increase toward the surface in core CH1a, which is attributed to the presence of halite. XRD analysis of the core reveals up to 16% halite at the surface of core CH1a, while the mineral is absent or only present in small amounts in the subsurface sediments and in core CH2a. Halite probably reflects important evapotranspiration and resulting oversaturation of Na and Cl at the marsh surface during drought periods between tidal flooding. The absence of halite in almost all other samples analyzed by XRD is attributed to dissolution *in situ* as a result of flushing or capillary action. Freshwater runoff is also more significant at the western tip of West Head (location of core CH2a), and thus enhances dissolution of halite.

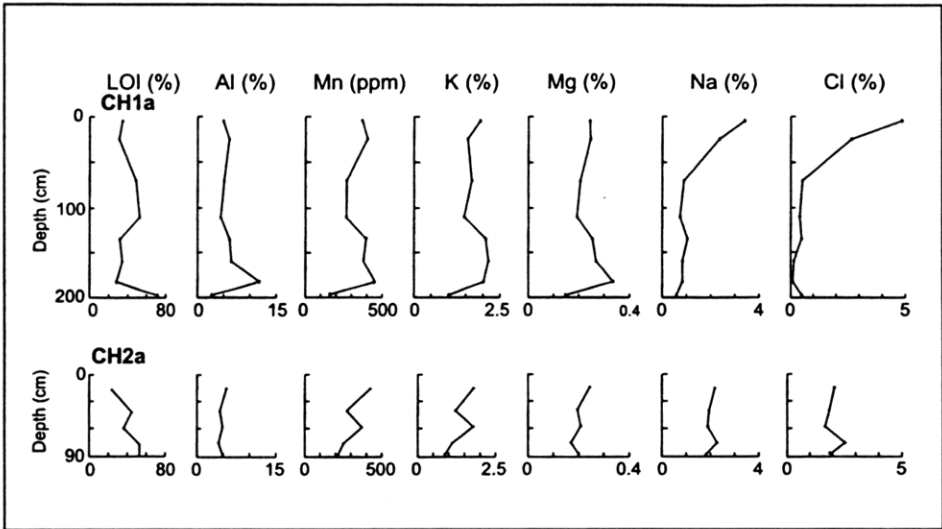


Fig 3 Weight loss on ignition (LOI), aluminium (Al), manganese (Mn), potassium (K), magnesium (Mg), sodium (Na) and chlorine (Cl) distribution in cores CH1a (middle marsh) and CH2a (high marsh). See Fig 1 for core sampling locations. Cores CH1a and CH2a were taken at sampling sites CH1 and CH2, respectively.

Iron concentrations average 3.4% in the core taken in the middle marsh (CH1a) and 1.9% in the core taken in the high marsh (CH2a). Fe concentrations in salt marsh sediments have been interpreted as being representative of the abundance of fine-grained matter in the marsh and are thus being used as a proxy for flooding frequency (Varekamp, 1991; Thomas and Varekamp, 1991; Chagué-Goff and Goff, 1999; Goff and Chagué-Goff, 1999), since Fe occurs mainly as fine-grained oxides in coastal and estuarine environments (Coonley *et al.*, 1971). Low and middle marsh areas are flooded more frequently than high marsh areas, and thus trap more fine-grained material. Therefore, the higher Fe content measured in the middle marsh than in the high marsh can be interpreted as a result of more frequent tidal flooding. In core CH1a, there is an enrichment of Fe, As, Co, Mo and S at a depth of 1.00-1.20 m (Fig 4), which does not coincide with a decrease in LOI (Fig 3) and thus can not be attributed solely to an increase of the mineral matter content in that particular interval. The Fe-rich band occurs at the upper boundary of the low marsh with the middle marsh (this delineation is based on macrophyte remains and foraminifera assemblages; Scott, 1977a; 1980). Enrichments of Fe and other metals have been found at the boundary between oxidizing and reducing conditions in tidal marsh sequences, where mobilization and reprecipitation as sulfides occur (e.g. Zwolsman *et al.*, 1993). McCaffrey and Thomson (1980) and Cochran *et al.* (1998) have also reported the presence of iron sulfide phases associated with metals. The coincidence of the Fe and S peaks is attributed to precipitation of Fe sulfides due to sulfate reduction (e.g. Zwolsman *et al.*, 1993). Pyrite was the dominant S mineral fraction determined by XRD, and microscopic observations revealed pyrite occurring mostly as framboids in association within the cortex of rootlets and rarely as small anhedral crystals (Chagué, 1994). Such an association of framboidal pyrite with plant rootlets has been frequently reported in salt marsh environments (e.g. Cohen *et al.*, 1983; Altschuler *et al.*, 1983; Oenema, 1990).

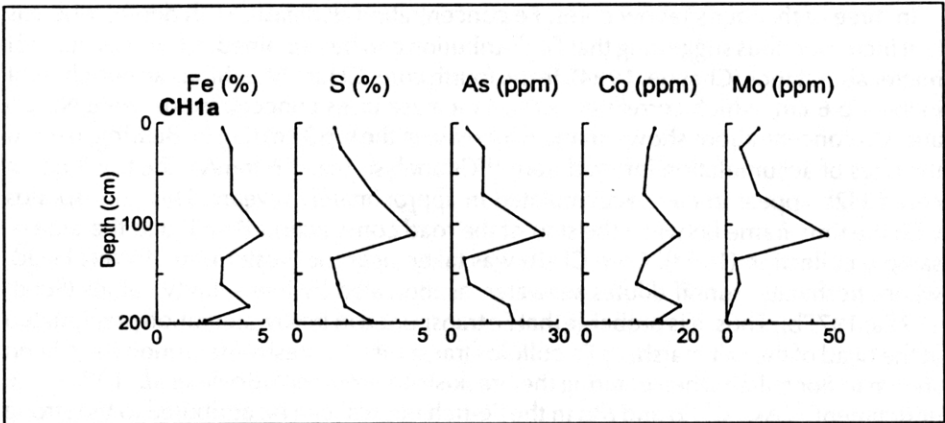


Fig 4 Iron (Fe), sulfur (S), arsenic (As), cobalt (Co) and molybdenum (Mo) distribution in core CH1a (middle marsh). Note the enriched interval at 1.00-1.20 m depth. Core CH1a was taken at sampling site CH1 (see Fig 1 for sampling location)

Previous studies also show that S and Fe are commonly correlated, as Fe-rich sediments are more likely to fix S than Fe-poor sediments (e.g. Thomas and Varekamp, 1991; Chagué-Goff and Goff, 1999). Thomas and Varekamp (1991) also report similar correlations in Fe-rich bands (except for As) and attribute this increased supply of trace elements in the pre-pollution period (before input of anthropogenic contamination) to an increase in flooding frequency. In the present study, As, Mo and Co are probably derived from the Goldenville Formation slates, which provide a local source for these elements (Graves and Zentilli, 1988). Their elevated concentrations in the interval at 1.00-1.20 m in core CH1a can be attributed to their association and precipitation as sulfides in the upper boundary of the low marsh sequence.

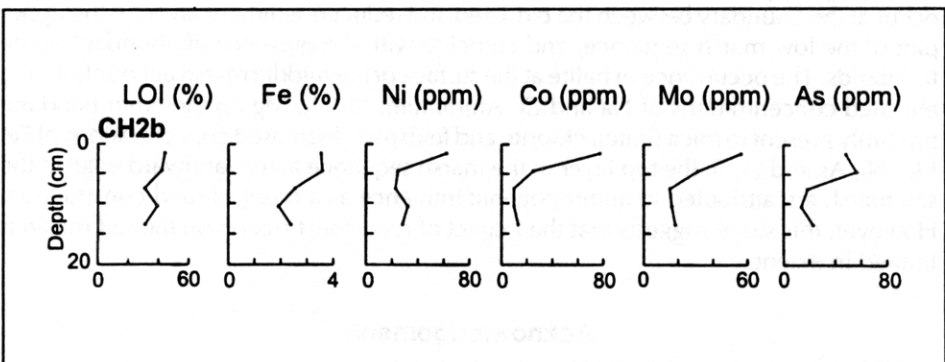


Fig 5 Weight loss on ignition (LOI), iron (Fe), nickel (Ni), cobalt (Co), molybdenum (Mo) and arsenic (As) distribution in core CH2b (high marsh). Core CH2b was taken at sampling site CH2 (see Fig 1 for sampling location).

In three of the four shallow cores, Fe concentration decreases with depth, whereas LOI increases, thus suggesting that Fe distribution can be explained in terms of mineral matter abundance (Chagué, 1994). In the fourth core, CH2b, Fe exhibits an enrichment in the top 6 cm, which correlates with an increase in As concentration, while Ni, Co and Mo concentrations show a marked increase in the top 3 cm (Fig 5). Bearing in mind the rates of accumulation inferred from ^{137}Cs analysis (ca. 3.6 mm/y), the top 3 cm of core CH2b appear to have accumulated in approximately 8 years. This corresponds with the time frame between the start of the road construction (1985) and the time of sample collection (1993). Core CH2b was taken near the western tip of West Head, where freshwater runoff dilutes seawater, as indicated by low salinity values (Scott, 1977a; 1977b). Thus, it is probable that Fe transported in freshwater runoff precipitated at the head of the salt marsh, as Fe colloids transported in freshwater runoff have been shown to flocculate when entering the brackish environment (Boyle *et al.*, 1977). The enrichment of As, Ni, Co and Mo in the Fe-rich interval can be attributed to the strong affinity of these elements for Fe-oxides and hydroxides (e.g. Lee, 1975). These elements were probably released in dust and runoff in association with the recent road construction (e.g. heavy equipment, Goldenville Formation slate used as road fill), transported into the salt marsh by freshwater runoff, and precipitated with Fe-oxides when entering the brackish environment. Elevated concentrations were not found in cores CH3 and CH4, which are located approximately 20 m from the highway. Therefore it is suggested that contamination associated with the most recent road construction is fairly localized in the salt marsh.

Conclusions

Variations in accretion rates, geochemistry and mineralogy in Chezzetcook Inlet salt marsh were determined using ^{137}Cs dating, INAA, Sulfur-Leco and XRD. The depth of the peaks in ^{137}Cs activity was used to estimate the accretion rates and suggests that they are slightly higher in the high marsh (CH2b, CH4 - 3.6 mm/y) than in the middle marsh (CH1b - 2.8 mm/y), possibly as a result of increased terrestrial input related to road construction in the area. Accretion rates are also slightly higher than the rate of relative sea level change.

Elemental distribution in the sediment cores is controlled by both natural and anthropogenic processes. Mobilization and reprecipitation of trace elements as sulfides occur at the boundary between the oxidized and reduced sediment layer, in the upper part of the low marsh sequence, and coincide with the presence of abundant pyrite framboids. The occurrence of halite at the surface of the middle marsh accounts for the elevated concentrations of Na and Cl. Aluminum, Mn, K, Mg on the other hand are probably present in mica (illite), chlorite and feldspars. Increased concentrations of Fe, Mo, Ni, As and Co in the top layer of the marsh sequence at the landward edge of the salt marsh are attributed to anthropogenic influence as a result of road construction. However, this study suggests that the impact of road construction on the salt marsh is limited in extent.

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