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Baseline

Mangrove sediments reveal records of development during the previous century (Coffs Creek estuary, Australia)

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ABSTRACT

A mangrove sediment core was studied to evaluate possible pollution of an urban estuary in Coffs Harbour, Australia. The heavy metal and nutrient profiles revealed a ~2.5-fold enrichment in more recent sediments. Lead-210 dating showed increasing phosphorous (P) and copper (Cu) accumulation following agricultural activity and population growth in the catchment after 1950. In contrast, nitrogen (N) did not show enrichment suggesting no external sources. Mercury (Hg) depositional fluxes and recent enrichment may be associated to an increase in fossil fuel emissions in the region. Down-core lead (Pb) profiles reflect an increase in leaded gasoline in the 1950s, then a decrease as a result of phasing out leaded gasoline in 1986. The heavy metal and nutrient depositional fluxes are well preserved in mangrove sediments and were related to historical events in the catchment.

Mangroves are important to estuarine systems by protecting shorelines from erosion, trapping sediments and acting as heavy metal and nutrient sinks (Duarte et al., 2013; Machado et al., 2016; Sanders et al., 2014a). Retention of contaminants is related to mangrove roots reducing tidal and fluvial water flow, promoting sediment deposition (Kirwan and Megonigal, 2013). Metal and nutrients in mangrove sediments may be linked to land use in adjacent catchments (Brady et al., 2014; Defew et al., 2005; Harbison, 1986; Nath et al., 2014a; Nath et al., 2014b). Mangrove sediments may therefore be considered an appropriate setting for studying historical activities in a catchment (Defew et al., 2005; Marchand et al., 2011; McCaffrey and Thomson, 1980; Serrano et al., 2011), particularly when baseline levels can be defined (Miola et al., 2016; Nienhuis, 1986; Serrano et al., 2011).

Spatial distributions of heavy metal contaminants originating from point sources have previously been constructed using sediments from mangroves (Harbison, 1984). For instance, Machado et al. (2016) estimated Hg fluxes into sediments in an estuary in Brazil. The Hg flux increased during rapid industrialization in the 1950s and decreased after emission control in the 1980s. Sanders et al. (2014b) revealed that mangrove sediments are also efficient in accumulating nutrients as a result of enrichment of the catchment. Here, we use a mangrove sediment core from Coffs Creek estuary (Coffs Harbour, Australia) to assess the heavy metal and nutrient depositional history in the catchment.

Coffs Creek estuary runs through the center Coffs Harbour, on the north coast of New South Wales, Australia. Coffs Harbour has a humid subtropical climate with seasonal variations in rainfall (wet summers and dry winters). Average yearly rainfall is 1699 mm with an average of 142 precipitation days and a yearly average of 63% relative humidity (BOM, 2017). Coffs Creek estuary is wave-dominated and intermittently infilled. The catchment area is small relative to the surrounding catchments and comprises the urban and residential center of Coffs Harbour (Fig. 1). Residential and agricultural land comprise 66% of the 25 km^2 catchment area (Ryder et al., 2012). The mangrove forests of Coffs Creek are dominated by *Avicennia marina* and have an area of 20.07 ha (Brown et al., 2016). The main anthropogenic pressures on Coffs Creek estuary include elevated sediment and nutrient loads in runoff and stormwater, increased concentrations of pesticides, herbicides and potential sewage inputs from on-site systems and overflows from the city sewage system (Ryder et al., 2012).

The history of Coffs Harbour includes agriculture and urban expansion. From ~1950 banana cultivation dominated in the basin (Yeates, 1990) and aerial spraying of bananas began in 1958 (Hedditch, 2014). More recently, blueberry agriculture has taken the place of some banana cultivation, accompanied by steady population increase and intense urban development. Agricultural substances of particular concern in Coffs Harbour are arsenic (As) from pesticides and phosphorus (P) from fertilizers. In addition, copper (Cu) fungicides are also used in blueberry, banana, and avocado farms in in this region (Van Zwieten et al., 2007). An increase in traffic through Coffs Harbour commenced in the 1950s as a result of the construction of a major highway, Pacific

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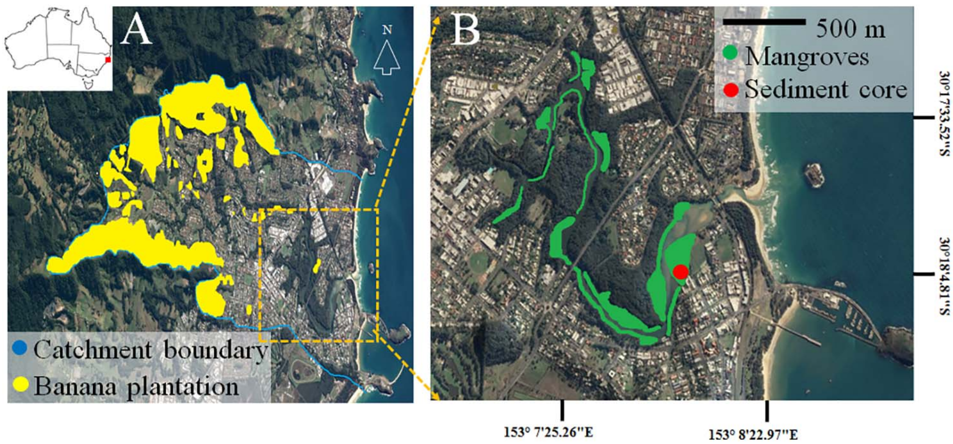

Fig. 1. A. Map of Coffs creek catchment (outlined in blue) with land historically used for banana plantations (yellow). Plantation area data provided by Coffs Harbour City Council (CHCC, 2016a, b). The catchment area is 25 km^2 and comprises the major urban center of Coffs Harbour. Residential and agricultural use makes up 66% of the catchment area (Ryder et al., 2012). B. Coffs Creek lower estuary with mangroves (green). Areal extent of Coffs Creek mangroves is 20.07 ha (Brown et al., 2016). Sediment core location indicated by red dot. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Highway (Yeates, 1990). Furthermore, a small airport is located approximately 5 km from Coffs Creek.

To investigate possible pollution in the estuary, a 50 cm long sediment core was collected from the upper tidal mangroves near the estuary mouth (30°18′3.63″S, 153° 8′2.23″E, Fig. 1) with a 5 cm diameter Russian peat auger on March 16, 2016. The sediment core was sectioned into 2 cm intervals. Sediments were dried to estimate the dry bulk density (DBD) (Brown et al., 2016). The content of As, Cu, lead (Pb), nickel (Ni), zinc (Zn), mercury (Hg), aluminum (Al) and P were measured at each 2 cm interval. Metals were extracted from sediments using 1:3 HNO_3/HCl acid digestion and an APHA Inductively Coupled Plasma Mass Spectrometer (ICPMS). Sediment reference materials were digested (AGAL 12) with every batch (sourced from National Measurement Institute) to confirm the recovery of the digest. To confirm accuracy and precision of the instrument we analyzed certified reference materials after the calibration and monitored drift by re-analyzing our mid-point standards every 20 samples and routinely use internal standards Sc, Ge, Rh and Ir. Nitrogen (N) concentration were determined in a Thermo Finnigan Model Delta Plus XP with analytical precision of N = 0.1%.

Enrichment factors (EF) were calculated to distinguish natural and anthropogenic sources of heavy metal and nutrients in Coffs Creek mangrove sediment. Enrichment factors compare preindustrial baseline levels to more recent sediment elemental content. Metals and nutrients were normalized to naturally abundant metals that are often unrelated to anthropogenic sources, such as Al (Abrahim and Parker, 2008; Weiss et al., 1999). Enrichment factors between 0.5 and 1.5 indicate natural fluctuations related to normal geological weathering, while a value of EF > 1.5 or patterns of increasing EF from 1 indicate anthropogenic sources (Zhang and Liu, 2002). To calculate enrichment factor, we normalized metal content to Al (Abrahim and Parker, 2008; Miola et al., 2016), as follows:

$$EF = \frac{\frac{[metal_x]}{[Al_x]}}{\frac{[metal_{baseline}]}{[Al_{baseline}]}}$$

where EF is enrichment factor and $[metal_x]$ is content of desired element at depth x and $[Al_x]$ is content of Al at depth x and $[metal_{baseline}]$ and $[Al_{baseline}]$ are baseline contents. The baseline content was defined as the concentration found in the bottom subsample of each core (Abrahim and Parker, 2008).

Radionuclide dating (^{210}Pb) was used to determine the sediment age and accumulation rates (SAR) (Appleby and Oldfield, 1992). Sediment intervals were combined and homogenized in 4 cm intervals for dating due to limited mass. Combined intervals were dated up to 40 cm depth. Five to eight grams of sediment were packed in vials and sealed with epoxy resin for at least 21 days to allow for ^{222}Rn to establish secular equilibrium between ^{226}Ra and its granddaughter ^{214}Pb. ^{210}Pb activity was determined by measuring the 46.5 keV gamma peak in a Canberra high-purity germanium (HPGe) well gamma detector. ^{226}Ra activity was determined by averaging peaks from the daughters ^{214}Pb and ^{214}Bi (295.2 keV, 351.9 keV and 609.3 keV) (Moore, 1984). The ^{210}Pb and ^{226}Ra activities were calculated by multiplying the counts per minute by a correction factor that includes the gamma ray intensity and detector efficiency determined from standard calibrations. Excess ^{210}Pb activity was calculated by subtracting the supported ^{210}Pb (i.e., ^{226}Ra activity) from the total ^{210}Pb activity. Excess (unsupported) ^{210}Pb was used to determine ages of sediment intervals using the Constant Initial Concentration (CIC) method described by Appleby and Oldfield (1992). Age of sediments is calculated by the following equation:

$$\text{Sediment age} = \text{Year of collection} - \left(\frac{\text{average depth of sediment interval}}{\text{SAR}}\right)$$

Heavy metal and nutrient accumulation rates are defined as amount of material entering the sediment per unit area per unit time ($\mu g\ m^{-2}\ yr^{-1}$):

MAR = $[metal_x] * SAR * DBD$ where $[metal_x]$ is trace metal content in sediment sample, SAR is sediment accumulation rate, and DBD is dry bulk density of the sample. Metal accumulation rates (MAR) were calculated for each metal and each 2 cm interval in the mangrove core.

The excess ^{210}Pb ($^{210}Pb_{ex}$) vertical profile revealed a log-linear decay (Fig. 2) with counting errors below 10%. The slope shows a significant correlation between $^{210}Pb_{ex}$ and depth (R^2 = 0.65; n = 5; $p < 0.05$) which allows an estimate of the sedimentation rates using the CIC method. The CIC dating method assumes that the sediment accretion rate has not varied during the encompassed time span (Appleby and Oldfield, 1992). According to the ^{210}Pb CIC dating method, the SAR is 0.51 cm yr^{-1}. The sediments from the bottom interval of the core date back to 1928.

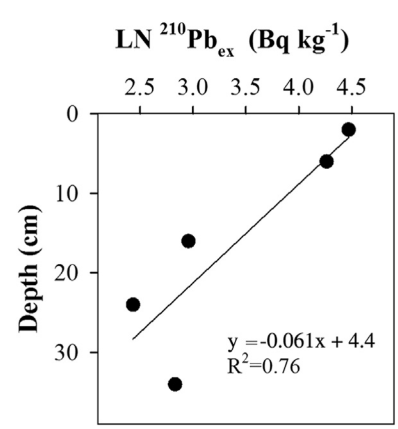

Fig. 2. Excess ^{210}Pb activity (Bq kg^{-1}) plotted against sediment depth (cm).

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