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Recent environmental change and trace metal pollution in World Heritage Bathurst Harbour, southwest Tasmania, Australia

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Abstract Bathurst Harbour in World Heritage southwest Tasmania, Australia, is one of the world's most pristine estuarine systems. At present there is a lack of data on pollution impacts or long-term natural variability in the harbor. A *ca.* 350-year-old ^{210}Pb-dated sediment core was analysed for trace metals to track pollution impacts from local and long-range sources. Lead and antimony increased from AD 1870 onwards, which likely reflects remote (i.e. mainland Australian and global) atmospheric pollution sources. Variability in the concentrations of copper and zinc closely followed the history of mining activities in western Tasmania, which began in the AD 1880s. Tin was generally low throughout the core, except for a large peak in AD 1989 ± 0.5 years, which may be a consequence of input from a local small-scale alluvial tin mine. Changes in diatom assemblages were also investigated. The diatom flora was composed mostly of planktonic freshwater and benthic brackish-marine species, consistent with stratified estuarine conditions. Since mining began, however, an overall decrease in the proportion of planktonic to benthic taxa occurred, with the exception of two distinct peaks in the twentieth century that coincided with periods of high rainfall. Despite the region's remoteness, trace metal analyses revealed evidence of atmospheric pollution from Tasmanian and possibly longer-range mining activities. This, together with recent low rainfall, appears to have contributed to altering the diatom assemblages in one of the most pristine temperate estuaries in the world.

Keywords Diatoms · Trace metals · ^{210}Pb · Palaeoecology · Estuaries · Australia · Tasmania

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Introduction

Projected climate changes that cause shifts in rainfall, a decline in seawater pH, and rising water temperatures, represent a major threat to estuarine and marine biodiversity (IPCC 2007). The biota of mid-latitude

Southern Hemisphere coastal ecosystems are particularly vulnerable because there are no landmasses further south, except for a few small sub-Antarctic islands and Antarctica, which can provide refugia (Barrett and Edgar 2010). Relatively pristine estuaries are rare globally, particularly in temperate areas, as many are already heavily degraded by human activities (Brush 2001), including local and long-range pollution. For southeast Australia (including Tasmania), European land management practices have caused unprecedented environmental changes since the AD 1800s (Dodson and Mooney 2002). This makes relatively pristine estuaries particularly valuable ecosystems. Understanding their natural variability and the implications of pollution and climate change are essential if their natural state and biodiversity are to be maintained.

To accomplish this requires knowledge of past ecosystem variability, as this provides a context for understanding recent observed changes and predicting what may happen in the future. A lack of baseline data and long-term monitoring, however, makes it difficult to do this, particularly in remote areas. For example, few studies have examined the effects of Australia's rapid industrialisation on trace metal pollution in remote locations (Marx et al. 2010).

A palaeoecological approach has the potential to address this problem (Smol 2008). Palaeoecological methods to assess changes in estuarine systems have previously been used to infer long-term human impacts related to salinity changes (Dick et al. 2011; Haynes et al. 2011; Ryves et al. 2004; Wachnicka et al. 2013), nutrient enrichment (Ellegaard et al. 2006; Logan et al. 2011; Weckström 2006), the effects of land-use change (Sloss et al. 2011; Taffs et al. 2008), alterations to estuarine openings (Saunders et al. 2007, 2008) and impacts of mining (Hollins et al. 2011; McMinn et al. 2003). Other applications include investigating sea-level change (Horton et al. 2006), determining natural baselines for management (Kauppila et al. 2005) and identifying 'near-pristine' estuaries as potential reference sites (Logan and Taffs 2011).

Bathurst Harbour (43°20′S, 146°10′E, Fig. 1) has the most pristine estuarine catchment in southern Australia. Despite Bathurst Harbour and the Port Davey area being more than three times the area of Sydney Harbour (178 vs. 55 km^2), it is less well known and has been the subject of very limited scientific study. Located within the Tasmanian Wilderness World Heritage Area (WHA), Bathurst Harbour forms part of the United Nations Educational Scientific and Cultural Organisation Biosphere Reserve, and is the only large estuarine system in southern Australia without significant human impact in its catchment (Edgar et al. 2010). It has highly distinct flora and fauna with high conservation value. There are no conspicuous introduced taxa and numerous endemic species exist, including many deep-sea species present at anomalously shallow depths (Edgar et al. 2010; Reid et al. 2008). At present, however, there are no baseline data on pollution impacts or past environmental changes. We therefore applied a multiproxy palaeoecological approach, using: (1) selected trace metals to investigate pollution from remote and Tasmanian mining activities; and (2) diatom and particle size analyses to identify natural ecosystem variability in Bathurst Harbour over the last *ca.* 350 years.

Site description

Bathurst Harbour is at the head of the Port Davey–Bathurst Harbour estuarine system, which is a prominent ria in southwest Tasmania (Fig. 1) formed by the postglacial inundation of a large river valley and its associated alluvial plain. It is a partially mixed estuary with a halocline that varies from 0 m (i.e. absent) during extended dry periods in summer to about 6 m during peak rainfall periods in winter (Edgar et al. 2010). The two layers differ markedly in salinity, temperature, chemical composition, pH and density (Edgar and Cresswell 1991). The bottom waters are brackish-marine and have a stable temperature from 13 to 16 °C year round, whereas the surface waters are relatively fresh and temperatures vary from 17 to 20 °C in summer and 9 to 12 °C in winter (Edgar and Cresswell 1991).

There are three major rivers leading into Bathurst Harbour and a long (12 km) narrow channel connecting it to Port Davey and the Southern Ocean (Barrett and Edgar 2010). It has an area of 46 km^2, a catchment of 2,000 km^2 and a relatively flat bottom that is 6–8 m deep (Mackey et al. 1996; Reid et al. 2008).

The geology of southwest Tasmania is Precambrian quartz-rich conglomerate, sandstone and siltstone, with micas and bands of altered impure siltstone (Reid

Fig. 1 Location and aerial photo (taken by K. Saunders) of Bathurst Harbour and other sites mentioned in the text

et al. 2008). Weathering does not provide many nutrients, which, together with high rainfall, means the soil in the catchment is generally low in nutrients, leached and thin. Previous studies found that concentrations of trace metals such as copper, cadmium and nickel were comparable with levels in open ocean water (Mackey et al. 1996). Vegetation cover is dominated by buttongrass moorlands and sedge grasses with *Eucalyptus* and temperate rainforests restricted to sheltered valleys and drainages (Reid et al. 2008). The dominance of buttongrass and peat in the catchment means the surface water layer contains a high concentration of tannin and other humic leachates. Humic substances supply few nutrients and substantially reduce light penetration. This means Bathurst Harbour has a thin photic zone (2–3 m, Reid et al. 2008), which results in a combination of deeper-water marine biota and organisms more typical of temperate shallow-water environments. As a result, the estuary is very low in nutrients and has low aquatic productivity.

The climate of southwest Tasmania is mostly wet and cool: mean annual precipitation is 2,400 mm, mean annual maximum temperature is 13.9 °C, mean annual minimum temperature is 8.6 °C. Mean annual wind speed is 33 km h^{-1} and is predominantly from the northwest (50 %) and west (15 %, BOM 2012).

Although currently nearly uninhabited, the area was discovered by Europeans in AD 1815. From the early AD 1830s to the 1870s, small ‘bay whaling’ settlements of up to ten workers were established in nearby Port Davey (Julen 1974). Near the end of the AD 1800s, commercial fishing developed in the region. During the AD 1900s, the only commercial activities were very limited tourism and small-scale fishing and mining. A tin deposit beside Melaleuca Lagoon, a tributary of Bathurst Harbour, first opened in AD 1935 and remained very small-scale. From AD 1941–2007 only one to three miners worked there, and the population in the entire catchment of Bathurst Harbour did not exceed 20 people in the twentieth century (Edgar et al. 2010).

Bathurst Harbour is 150 km southeast from Queenstown, part of the major mining region in Tasmania. This means that even though the mining operations are not within the catchment of Bathurst Harbour, they are upwind of the harbor, which likely receives input from the mining operations via atmospheric deposition. This, however, has not yet been established.

Materials and methods

Sediment core collection

A 35-cm-long sediment core was collected from the centre of Bathurst Harbour (43°20′35.2″S, 146°11′1.7″E, Fig. 1) at 7 m water depth in March 2007 using a Glew gravity corer. The sediment core was sectioned at 0.5-cm intervals, on-site, and transported to the University of Tasmania, Hobart, Tasmania, where samples were stored in the dark at 4 °C until analysis.

Chronology

A sediment chronology was established using ^{210}Pb dating (Appleby and Oldfield 1978). Unsupported ^{210}Pb activities were measured in bulk sediment samples at the Australian Nuclear Science and Technology Organisation (ANSTO) Institute for Environmental Research, using alpha spectroscopy following methods described by Harrison et al. (2003) and McMinn et al. (1997). The ^{210}Pb ages were calculated using the constant rate of supply (CRS) model (Appleby 2001). Errors on CRS ages were calculated by propagating uncertainties in the excess ^{210}Pb inventory. The CRS model was selected for age and mass accumulation rate calculations at Bathurst Harbour, because no significant catchment disturbance or changes to hydrology have occurred, and the majority of ^{210}Pb deposition is likely to have been atmospheric and stable over time (Appleby 2001, 2008). Given the constant mass accumulation rate from 10 to 15 cm, we felt justified in estimating the sedimentation rate to the base of the sediment core by extrapolation.

Grain size

The grain size distribution was measured at 0.5-cm intervals from 0 to 14 cm (except 9.5, 11 and 13 cm) and 1.5- or 2-cm intervals from 15 to 35 cm using a Malvern Mastersizer S Laser Particle Size Analyser. Approximately 0.2 g of bulk wet sediment from each sample was dispersed in water and pumped through a measurement chamber in the laser particle analyser. Measurements were done in duplicate. The particle size distribution of solids with a diameter in the range 0.05–880 μm was determined and split into three groups: fine (clay) particles (<2 μm), medium (silt) particles (2–63 μm) and large (sand) particles (>63 μm).

Trace metals

Trace metal analyses were carried out on sub-samples from the same depths as grain size (except 6, 7, 8 and 10.5 cm). Each sub-sample was dried at 40 °C and ground to obtain a homogenous representative sample. Between 0.02 and 0.3 g of sample was digested in a closed-vessel microwave digestion unit following the USEPA 3051 method. A small amount of hydrofluoric acid (l mL) was added in addition to nitric and hydrochloric acid prior to microwave digestion. A reagent blank and a standard reference material (PACS-1 or MESS-3) were prepared and measured with each batch of samples. Measurements of total As, Co, Cu, Ni, Pb, Sb, Sn and Zn were made using inductively coupled plasma mass spectrometry (ICP-MS). Reagent blanks showed negligible concentrations of measured elements and standard reference material results were within ±20 % of certified values.

Diatoms

Diatom samples were analysed at 0.5-cm intervals throughout the sediment core. Samples were prepared using standard methods (Battarbee et al. 2001). Where possible, at least 300 valves were counted per sample, using phase contrast and oil immersion at 1,000× magnification on a Zeiss Z20 light microscope. The relative abundance of all species (including unidentified forms) was recorded as a percentage of the total number of valves counted (Battarbee et al. 2001). Taxonomy was principally based on reference datasets for coastal Tasmanian and southeast mainland Australian water bodies (Saunders 2010) and sub-Antarctic Macquarie Island lakes and ponds (Saunders et al. 2009), with additional references to other Tasmanian (Hodgson et al. 1997; Vyverman et al. 1995), mainland Australian (John 1983; Sonneman et al. 2000), sub-Antarctic (Van de Vijver et al. 2002) and Antarctic (Roberts and McMinn 1998) literature. All taxa were photographed and are archived with K. Saunders. A final species list was developed including only those species occurring with >1 % relative abundance.

Diatom preservation was poor below 6 cm and as many diatoms as possible were counted below this depth. All samples containing >150 valves were included in the interpretation. Samples (depths 15, 20, 35 cm) containing <150 were not.

Because of the diverse diatom flora and poor preservation, the interpretation was based on a qualitative assessment of changes. The ratio of planktonic to benthic taxa was calculated and diatom zones were determined by optimal partitioning (Birks and Gordon 1985) using constrained hierarchical clustering and the broken stick method (Bennett 1996). This was performed in R (R Development Core Team 2009) using the add-on packages Rioja (Juggins 2012) and Vegan (Oksanen et al. 2007). All stratigraphic diagrams were developed in C2 version 1.4 (Juggins 2007).

Results

Chronology

Table 1 summarises the ^{210}Pb results. ^{210}Pb activity reached background at 15–15.5 cm (AD 1859 ± 6) and the linear sedimentation rate was fairly constant over the datable section of the core (Fig. 2). Dates within the ^{210}Pb-dated period are given with errors (± years), whereas extrapolated dates below 15 cm are approximate and preceded by '*ca.*'

Grain size

The sediment consisted of homogenous, organic, fine-grained material throughout. The sediment was principally composed of silt (mid)-size particles and to a lesser extent sand (coarse)-size particles, with <5 % contribution from clay (fine)-size particles (Fig. 3).

Trace metals

The trace metal results showed different stratigraphic trends (Fig. 4). Co fluctuated throughout the sediment core and was significantly ($p < 0.05$) correlated to sand and silt (Fig. 3) ($r = -0.61$ and 0.62, respectively). Cu, Pb, Sb, As and Zn were all significantly correlated with one another ($p < 0.05$). All had low concentrations from the base of the core to 14 cm. From 14 to 9.5 cm all increased. From 9.5 to 1 cm, Pb, Sb and Zn remained relatively stable at higher concentrations than in the lower part of the sediment core, whereas Cu continued to increase, reaching a peak at 4.5 cm. All the trace metals decreased from 4.5 to 1 cm and increased in the surface sample (Fig. 4). Ni did not have the same initial increase from 9.5 cm, but also peaked at 4.5 cm. Sn remained at relatively low concentrations throughout the sediment core, although there was a slight increase at 9.5 cm and large peak at 2.5 cm (Fig. 4).

Table 1 The unsupported ^{210}Pb activities measured in the sediment core and calculated CRS ages

Depth (cm)	Cumulative dry mass (g cm^{-2})	Total ^{210}Pb (mBq/g) or (Bq/kg)	Unsupported ^{210}Pb (mBq/g) or (Bq/kg)	Dry bulk density (g/cm^3)	Calculated CRS ages (years)	Mass accumulation rates (g cm^{-2}/y)	Calendar age (years AD)
0.25 ± 0.25	0.07 ± 0.07	353.3 ± 13.7	349.7 ± 13.8	0.304	2.0 ± 0.1	0.036 ± 0.001	2005
1.25 ± 0.25	0.34 ± 0.07	324.2 ± 11.0	315.0 ± 11.0	0.241	7.3 ± 0.3	0.048 ± 0.002	2000
2.25 ± 0.25	0.61 ± 0.07	359.1 ± 8.1	363.4 ± 8.2	0.293	13.6 ± 0.4	0.045 ± 0.001	1993
2.75 ± 0.25	0.76 ± 0.07	333.2 ± 10.9	333.0 ± 11.0	0.293	17.7 ± 0.5	0.043 ± 0.001	1989
3.25 ± 0.25	0.90 ± 0.07	321.2 ± 6.1	322.7 ± 6.3	0.288	22.2 ± 0.6	0.041 ± 0.001	1985
3.75 ± 0.25	1.05 ± 0.07	258.4 ± 9.1	252.8 ± 9.2	0.281	26.6 ± 0.8	0.039 ± 0.001	1980
4.25 ± 0.25	1.18 ± 0.07	277.9 ± 7.9	278.0 ± 8.2	0.59	31.1 ± 0.9	0.038 ± 0.001	1976
5.25 ± 0.25	1.41 ± 0.07	202.7 ± 8.6	195.0 ± 8.7	0.205	39.3 ± 1.1	0.036 ± 0.001	1968
5.75 ± 0.25	1.53 ± 0.07	188.2 ± 4.2	188.6 ± 4.4	0.274	43.5 ± 1.2	0.035 ± 0.001	1964
6.75 ± 0.25	1.81 ± 0.07	125.7 ± 3.5	120.9 ± 3.7	0.279	53.0 ± 1.4	0.033 ± 0.001	1954
7.75 ± 0.25	2.09 ± 0.07	126.4 ± 3.2	123.9 ± 3.3	0.281	63.5 ± 1.7	0.033 ± 0.001	1943
8.75 ± 0.25	2.35 ± 0.07	106.5 ± 3.6	100.9 ± 3.7	0.249	76.8 ± 2.0	0.031 ± 0.001	1930
9.25 ± 0.25	2.49 ± 0.07	80.4 ± 2.2	78.8 ± 2.3	0.297	84.4 ± 2.2	0.030 ± 0.001	1923
10.3 ± 0.25	2.77 ± 0.07	46.7 ± 2.5	37.8 ± 2.8	0.252	97.3 ± 2.7	0.028 ± 0.001	1910
11.8 ± 0.25	3.17 ± 0.07	31.0 ± 1.7	23.5 ± 1.9	0.288	113.6 ± 3.4	0.028 ± 0.001	1893
12.3 ± 0.25	3.31 ± 0.07	24.0 ± 0.7	20.6 ± 0.8	0.273	119.4 ± 3.7	0.028 ± 0.001	1888
13.8 ± 0.25	3.69 ± 0.07	17.1 ± 1.1	10.0 ± 1.3	0.235	133.9 ± 4.8	0.028 ± 0.001	1873
14.3 ± 0.25	3.82 ± 0.07	20.2 ± 0.7	10.1 ± 1.2	0.272	138.3 ± 5.2	0.028 ± 0.001	1869
14.8 ± 0.25	3.96 ± 0.07	17.1 ± 0.6	9.0 ± 1.0	0.277	143.7 ± 5.8	0.028 ± 0.001	1863
15.3 ± 0.25	4.08 ± 0.07	17.7 ± 1.6	6.2 ± 2.0	0.226	148.1 ± 6.3	0.028 ± 0.001	1859
16.3 ± 0.25	4.33 ± 0.07	10.7 ± 0.6	2.8 ± 1.0	0.281	*157.8*		*1849*
20.3 ± 0.25	5.31 ± 0.07	17.2 ± 1.9	7.5 ± 2.3	0.210	*196.3*		*1811*
25.3 ± 0.25	6.42 ± 0.06	16.2 ± 1.9	5.8 ± 2.2	0.231	*244.4*		*1763*
30.3 ± 0.25	7.71 ± 0.06	19.1 ± 1.5	7.6 ± 1.8	0.284	*292.5*		*1714*
33.3 ± 0.25	8.57 ± 0.06	15.5 ± 1.3	0.6 ± 1.9	0.294	*321.3*		*1686*

Dates from 15 cm to the base of the core are in italics and estimated based on extrapolation of CRS-derived ages above 15 cm. Cumulative dry mass and dry bulk density are calculated according to Appleby (2001) using moisture content and an estimated sediment density of 2.5 g cm^{-2}. Unsupported ^{210}Pb is corrected to core collection date (13 March 2007)

Diatoms

Diatom preservation was relatively poor throughout the sediment core and it was not possible to count 300 specimens in all samples. Less than 150 specimens were identified at 15, 20 and 35 cm, and fewer than 200 specimens were identified at 10, 12, 13, 18–19, 24–25 and 28–29 cm. Thus, poor preservation was accounted for when interpreting the diatom record.

A total of 174 diatom species were identified and optimal partitioning divided the core into three zones: (1) 35–20 cm; (2) 20–8.5 cm; and (3) 8.5–0 cm. The dominant (≥10 % maximum relative abundance) species were *Aulacoseira distans*, *Aulacoseira distans* var. *nivalis*, *Cocconeis neothumensis*, *Cocconeis scutellum*, *Cocconeis* sp. 1, *Coscinodiscus* sp. 1, *Cyclotella bodanica*, *Grammatophora macilenta*, *Pinnularia rabenhorstii* var. *rabenhorstii*, *Pinnularia rabenhorstii* var. *subantarctica* and *Rhabdonema arcuatum* (Fig. 5). Of these, *A. distans* var. *nivalis* occurred with the greatest relative abundance (max. 81.7 % at 3.5 cm, AD 1980 ± 0.8 years). The other planktonic taxa occurred with <20 % maximum relative abundance and fluctuated throughout the sediment core.

G. macilenta and *R. arcuatum* were the most abundant benthic taxa and were present throughout. They were most abundant from the base to 20 cm

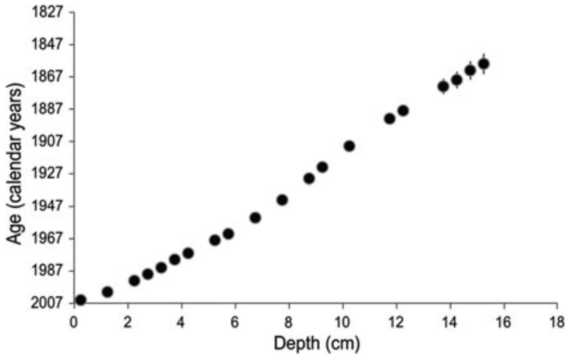


Fig. 2 CRS-derived ^{210}Pb age-depth model for the Bathurst Harbour sediment core

(Fig. 5). *P. rabenhorstii* var. *rabenhorstii* and *P. rabenhorstii* var. *subantarctica* were the most abundant benthic freshwater taxa. There was a clear transition between the two at 20 cm and a virtual disappearance of both from 13.5 cm to the surface of the core. The relative abundance of *C. scutellum* steadily increased from the base of the core to a peak at 21 cm, after which it occurred in very low relative abundance from 20 to 4.5 cm. From 4.5 cm to the surface of the core it steadily increased, together with *Cocconeis* sp. 1 and *C. neothumensis* (Fig. 5).

The proportion of planktonic to benthic species fluctuated throughout the sediment core and was mostly a result of changes in the relative abundance of *A. distans* var. *nivalis*. The benthic diatoms were mostly brackish-marine taxa, but there was an increase in freshwater benthic taxa from 26 to 14 cm, which was primarily as a consequence of an increase in *P. rabenhorstii* var. *rabenhorstii* and *P. rabenhorstii* var. *subantarctica* (Fig. 5).

Discussion

Trace metals

Atmospheric pollution from mining is inferred from the changes in Pb, Sb, Ni, As, Cu, Zn and Sn (Fig. 4). The concentrations of all were low prior to AD 1869 ± 5.2 years. After AD 1869 ± 5.2 years, concentrations of all trace metals increased, but the timing of the initial increase differed for each trace metal species, which may reflect the relative balance of remote (Pb and Sb), versus Tasmanian (As, Cu, Ni and Zn) and possibly local (Sn) sources (Fig. 4).

Pb and Sb increased from AD 1873 ± 4.8 years onwards (Fig. 4). This predates the start of mining in western Tasmania, and suggests these metals are derived from remote (mainland Australian and/or global) sources. The timing coincides with the onset of lead mining in mainland Australia in the AD 1840s and the intensification of lead production after the late AD 1880s, following the discovery of the Broken Hill deposits, >1,300 km north of Bathurst Harbour (Fig. 1). Previous work in the Australian Alps demonstrated metal enrichment in peat profiles was also closely tied to Australian mining and production records (Marx et al. 2010). Additionally, global deposition of these metals has previously been identified in remote regions elsewhere, including the Himalaya (Lee et al. 2008), Antarctica (Van de Velde et al. 2005) and the Arctic (Bindler et al. 2001).

Extensive mining occurred in western Tasmania with the arrival of Europeans. Bathurst Harbour is located downwind of many of the major mining sites, with 50 % of the winds coming from the northwest (BOM 2012). The rise in As, Cu, Ni and Zn occurred after Pb and Sb. Mines began operating in western Tasmania in the AD 1880s (Harle et al. 2002). The largest mine, Mt. Lyell, opened in AD 1893 (Fig. 1), coinciding with an increase in As, Cu and Zn (Fig. 4) in the Bathurst Harbour record. In the AD 1950s, open-cut mining dramatically expanded, leading to large quantities of metal-laden dust (Harle et al. 2002). This is seen in the sediment record as corresponding increases in As, Cu, Ni and Zn, with Ni peaking in AD 1964 ± 1.2 and As, Cu and Zn peaking in AD 1968 ± 1.1 years (Fig. 4).

The impacts of mining on Tasmanian estuarine (Seen et al. 2004) and lake sediments (Harle et al. 2002; Hodgson et al. 2000) have been demonstrated previously. For example, Seen et al. (2004) found that heavy metal profiles in sediments in northern Tasmania (Tamar Estuary, near Launceston, Fig. 1) had a strong correlation with mining activities and industrialisation. They showed a gradual increase in heavy metals occurred from the early AD 1900s onwards, which increased to a broad peak after World War Two as a consequence of accelerated industrialisation in the Launceston region. Decreases in metal concentrations occurred in the late twentieth century, likely as a consequence of reduced industrial activity and/or the implementation of pollution control measures (Seen et al. 2004).

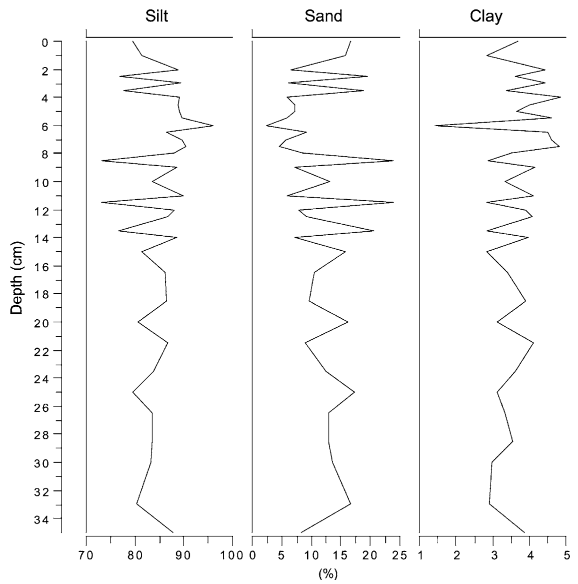


Fig. 3 Grain size distribution in the Bathurst Harbour sediment core

Harle et al. (2002) showed reductions in pollen diversity together with rapidly increasing concentrations of trace metals in sediments from Lake Dora, western Tasmania (<20 km from Queenstown). The timing of this change coincided with the escalation of open-cut mining from the AD 1950s–1970s, when significant deforestation occurred because of logging and pollution from smelters. Mining production declined in the AD 1970s, which likely led to the decrease in As, Cu and Zn from the AD 1970s onwards in Bathurst Harbour. In particular, there was a sharp decrease in Cu and Zn after AD 1993 ± 0.4 years, which may be a consequence of the closure of Mt. Lyell in AD 1994 (Harle et al. 2002).

Changes in Sn were also observed in the Bathurst Harbour sediment core. There was a slight increase from AD 1887 ± 3.7 years onwards to a new baseline concentration reached by the start of the twentieth century (Fig. 4). The main feature of the Sn record, however, is the large peak in AD 1989 ± 0.5 years. Very small-scale tin mining started in the catchment of Bathurst Harbour, near Melaleuca Inlet in AD 1936 and continues today (Fig. 1). Given the sudden increase and decrease of Sn, the source may have been local in origin and caused by a one-off event or 'slug' of Sn entering the Harbour. Further measurements and sediment cores from Bathurst Harbour are needed to determine the cause of this rise.

Diatoms

The diatom analysis provided a record of natural environmental changes spanning the last *ca.* 350 years. The dominant taxa identified were primarily freshwater planktonic species (except *Coscinodiscus* sp. 1) and brackish-marine benthic taxa, except *P. rabenhorstii* var. *rabenhorstii* and *P. rabenhorstii* var. *subantarctica*. This likely reflects the different diatom composition of the fresh surface waters vs. the estuarine brackish waters in Bathurst Harbour. Many of the dominant taxa were also more characteristic of sub-Antarctic water bodies rather than coastal Tasmanian/mainland Australian water bodies. In particular, *A. distans*, *A. distans* var. *nivalis*, *C. neothumensis*, *P. rabenhorstii* var. *rabenhorstii* and *P. rabenhorstii* var. *subantarctica* were previously found to be abundant in Macquarie Island lakes and ponds (Saunders et al. 2009), and common in other sub-Antarctic samples (Van de Vijver et al. 2002). They have not, however, been previously identified in east coast Tasmanian or mainland southeast Australian coastal water bodies.

A. distans and *A. distans* var. *nivalis* are freshwater planktonic species common in sub-Antarctic lakes and ponds with low conductivity and slightly acidic to circum-neutral pH (Saunders et al. 2009; Van de Vijver et al. 2002). *A. distans* is also widespread in Tasmanian alpine lakes (Vyverman et al. 1995).

P. rabenhorstii var. *rabenhorstii* and *P. rabenhorstii* var. *subantarctica* are freshwater benthic species and commonly found in soil samples influenced by sea spray (Van de Vijver et al. 2002). *C. neothumensis* has been commonly found in Macquarie Island lakes with higher salinity and nutrients (Saunders et al. 2009). It has also been recorded on other sub-Antarctic islands on semi-wet mosses and in small lakes with circum-neutral pH and high conductivity (Van de Vijver et al. 2002), and in brackish, relatively high-nutrient environments of the Baltic Sea (Weckström and Juggins 2006).

In contrast, *C. scutellum* and *G. macilenta* were common in many brackish and marine sites, respectively, in eastern Tasmanian and mainland southeastern Australian coastal water bodies (Saunders 2010). They, however, have not been found in Macquarie Island or alpine Tasmanian lakes.

R. arcuatum is a marine species (Guiry and Guiry 2012) that has previously been identified in coastal sediments near King George Island, Antarctica (Al-

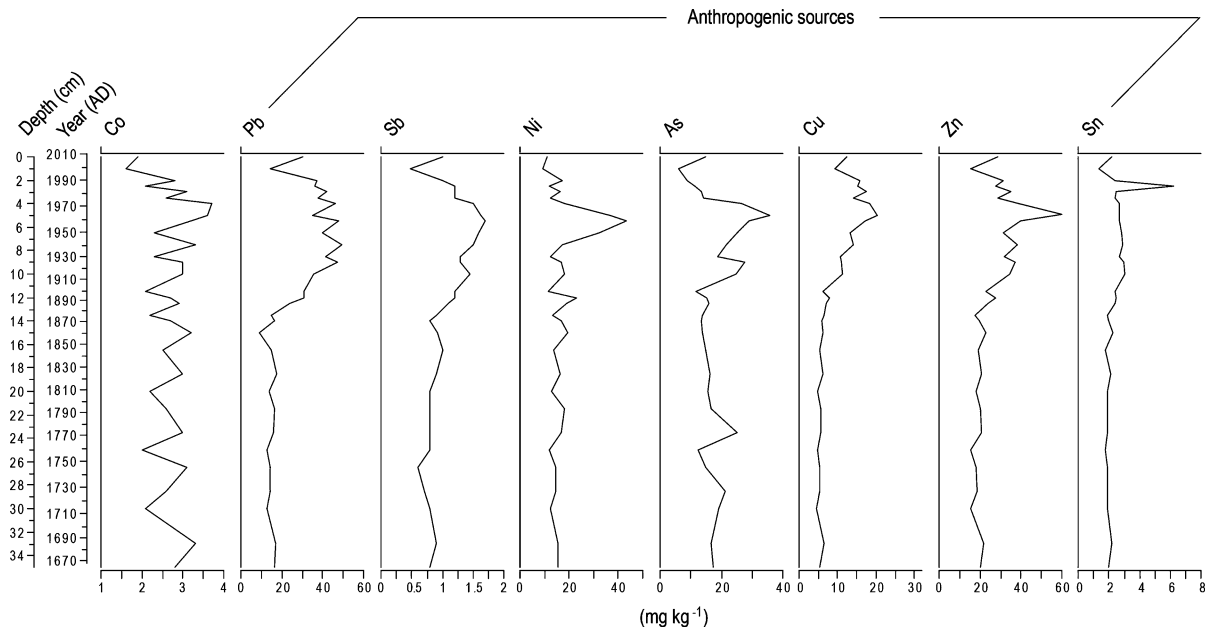


Fig. 4 Summary of trace metal analyses in the Bathurst Harbour sediment core

Handal and Wulff 2008), whereas *C. bodanica* is a freshwater species characteristic of relatively low-nutrient environments (Bradbury et al. 2002).

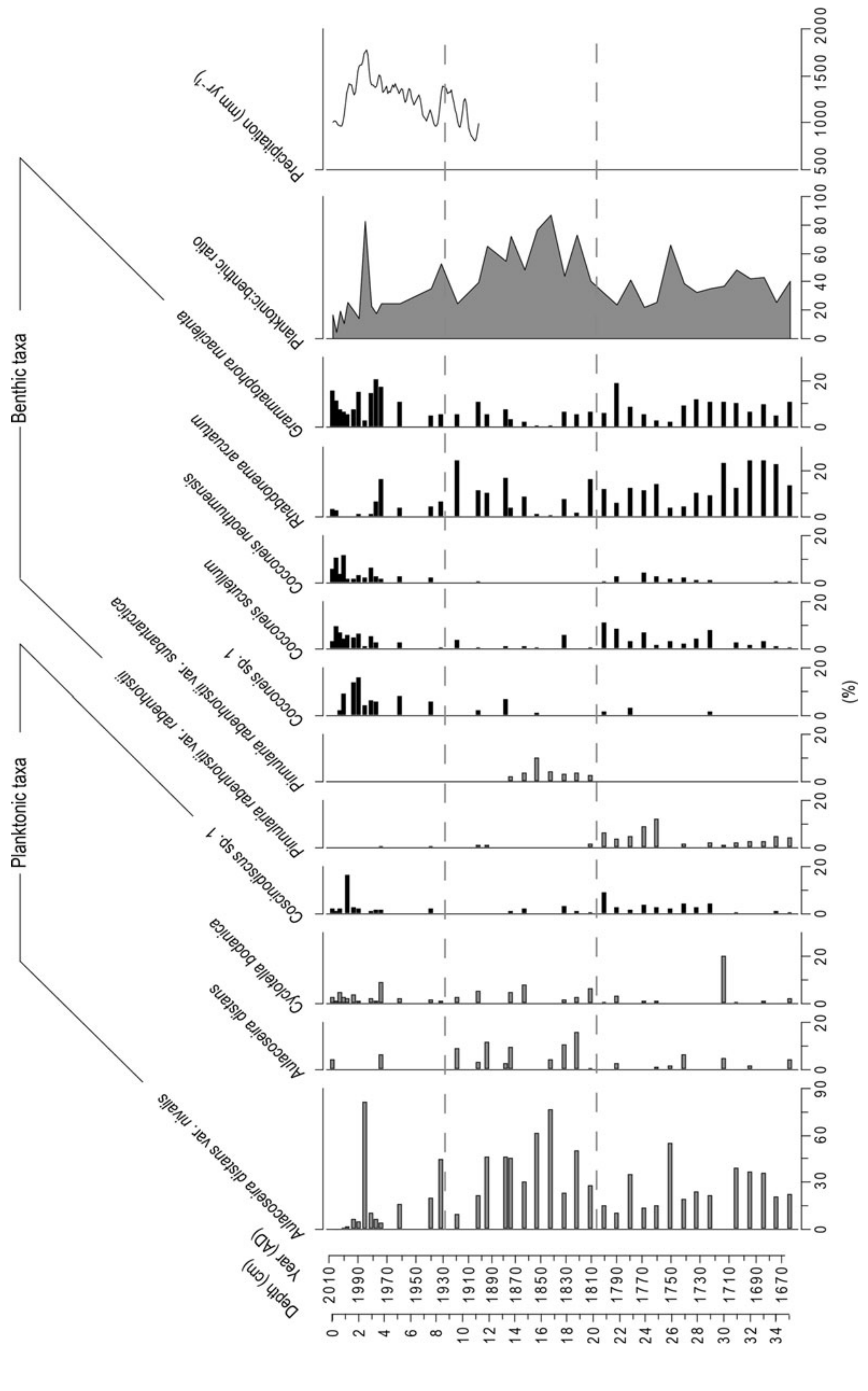
Based on optimal partitioning, there were three main phases of diatom assemblage variability: *ca.* AD 1685–1810 (35–20 cm), *ca.* AD 1810–1930 (20–8.5 cm) and AD 1930–present (8.5–0 cm, Fig. 5). We link these phases to a combination of atmospheric pollution impacts and changes in the precipitation and stratification regime.

From *ca.* AD 1685–1810, a combination of planktonic *A. distans* var. *nivalis* and benthic *R. arcuatum* and *G. macilenta* dominated the diatom flora (Fig. 5). This suggests the benthic environment had relatively high salinity, which is also implied by the presence of *C. scutellum* and *C. neothumensis*, as these taxa have brackish-marine preferences. The freshwater *P. rabenhorstii* var. *rabenhorstii* was also present, but in low (<5 %) relative abundance. Overall, the diatom flora suggests the halocline was likely maintained throughout this period.

The period from *ca.* AD 1800–1930 was characterised by a transition from *P. rabenhorstii* var. *rabenhorstii* to *P. rabenhorstii* var. *subantarctica*, and generally lower relative abundances of brackish-marine taxa *C. scutellum*, *C. neothumensis*, and at times, *R. arcuatum* and *G. macilenta* (Fig. 5). The reason for the transition between the two *Pinnularia* species is not clear, as they have similar ecological preferences in terms of salinity, nutrients and pH (Saunders et al. 2009; Van de Vijver et al. 2002), but may be a consequence of another factor such as temperature, light and/or oxygen concentration.

The proportion of planktonic taxa, which is mostly accounted for by fluctuations in *A. distans* var. *nivalis*, increased at the start of this period and peaked *ca.* AD 1840, before declining (Fig. 5). The dominance of freshwater planktonic taxa suggests extended periods of stratification, a thicker lens of surface fresh water and/or reduced light penetration below the stratified layer. As rainfall and riverine inputs are necessary to maintain stratification (Edgar et al. 2010) and the planktonic forms were dominated primarily by taxa found on Macquarie Island (*A. distans* var. *nivalis*) and alpine Tasmanian lakes (*A. distans*), this suggests the majority of the nineteenth century may have been relatively wet and cool in southwest Tasmania. This is supported by other records from the region, as this period was relatively wet in northwestern Tasmania (Saunders et al. 2012) and cool in western Tasmania and New Zealand (Cook et al. 2000; Xiong and Palmer 2000).

From AD 1873 ± 4.8 years onward, *P. rabenhorstii* var. *rabenhorstii* and *P. rabenhorstii* var. *subantarctica*


Benthic taxa
Planktonic taxa
Precipitation (mm yr-1)
Planktonic:benthic ratio
Grammatophora macilenta
Rhabdonema arcuatum
Cocconeis neothumensis
Cocconeis scutellum
Cocconeis sp. 1
Pinnularia rabenhorstii var. subantarctica
Pinnularia rabenhorstii var. rabenhorstii
Coscinodiscus sp. 1
Cyclotella bodanica
Aulacoseira distans
Aulacoseira distans var. nivalis
Year (AD)
Depth (cm)
(%)

◀**Fig. 5** Dominant diatom taxa (species ≥10 % maximum relative abundance) in the Bathurst Harbour sediment core and record of precipitation from AD 1900 (BOM 2012). Diatom species plotted in grey are freshwater taxa. All data are presented as percentage values (except precipitation, which is in mm year^{-1}). *Grey dashed lines* indicate zones as determined by optimal partitioning of the diatom data (see text for details). (Color figure online)

virtually disappeared, whereas the proportion of benthic taxa with higher salinity preferences increased (Fig. 5). From *ca.* AD 1930 onward, there was an increase in benthic brackish-marine taxa, virtually no benthic freshwater taxa and an overall decrease in planktonic taxa, apart from a brief period in the AD 1980s (Fig. 5). The peak in planktonic taxa was primarily a consequence of a peak in *A. distans* var. *nivalis*, and coincided with a peak in rainfall (Fig. 5).

Potential impacts of precipitation variability and atmospheric pollution on diatom assemblages in Bathurst Harbour

Precipitation is known to be important for maintaining the halocline in Bathurst Harbour (Edgar et al. 2010). Precipitation alone, however, probably does not explain all of the changes in the diatom assemblages during the twentieth century. In particular, while rainfall generally increased, the proportion of planktonic taxa decreased.

The main decline in planktonic taxa began *ca.* AD 1930. This coincided with an increase in Ni and further increases in As, Cu and Sb. The impact of trace metal pollution on diatom communities, particularly a reduction in planktonic taxa, has been documented (Cattaneo et al. 2008; Ruggiu et al. 1998; Tuovinen et al. 2012). The establishment and increase in mining activity in western Tasmania may have caused enough atmospheric pollution to impact Bathurst Harbour's diatom assemblages.

Although the trace metal concentrations reported for recent decades are relatively low compared to established guidelines for Australian and New Zealand sediments (Table 2, ANZECC 2000), the mean concentration of each trace metal increased after the onset of mining (i.e. post-AD 1870). In particular, the mean concentrations of Cu and Pb in the sediments pre- and post-AD 1870 more than doubled, while the mean concentration of Zn nearly doubled (Table 2). The maximum concentrations of Cu, Pb, Sn and Zn nearly tripled post-AD 1870 and the maximum concentration of Ni exceeded current Australian and New Zealand sediment quality guidelines, while Pb almost did (Table 2, ANZECC 2000). The maximum concentration of As, both pre- and post-AD 1870, exceeded

Table 2 Pre-1870 and post-1870 mean, minimum and maximum concentrations of trace metals measured in sediments from Bathurst Harbour, together with ANZECC (2000) Interim Sediment Quality Guidelines (ISQG)

Metal	Pre-1870 concentration (mg kg^{-1})			Post-1870 concentration (mg kg^{-1})			ISQG-low[a]	ISQG-high	Common sources
	Mean	Min.	Max.	Mean	Min.	Max.			
Antimony	0.8	0.6	1.0	1.2	0.5	1.7	2	25	Discharge from petroleum refineries, fire retardants, ceramics, electronics, solder
Arsenic	16.7	12.4	**25.1**	18.6	6.1	**35.7**	20	70	Erosion of natural deposits
Copper	5.5	4.4	6.4	12.7	6.2	20.3	65	270	Erosion of natural deposits
Lead	14.7	9.1	17.4	35.5	14.1	49.2	50	220	Erosion of natural deposits
Nickel	15.2	11.9	19.3	18.9	9.1	**43.4**	21	52	Present in some ground waters as a consequence of dissolution from nickel ore-bearing rocks
Tin	2.0	1.8	2.3	2.7	1.3	6.2	N/A	N/A	Naturally-occurring. Anthropogenic sources include coal and wood combustion, waste incineration and sewage sludge
Zinc	19.3	15.2	22.8	31.7	15.6	60.6	200	410	Naturally-occurring. Industrial sources or toxic waste sites associated with mining, coal and waste combustion and steel processing

Low and high concentrations are listed. Numbers in bold highlight metals that exceed trigger value concentrations

[a] Trigger value concentrations

current Australian and New Zealand sediment quality guidelines. The reason for relatively high concentrations prior to AD 1860 is not known. Since the peak in metal concentrations in the AD 1960s, the overall concentrations have decreased in recent decades and most trace metals, except Cu and Zn, have nearly returned to their pre-mining (i.e. baseline) concentrations (Fig. 4). This likely reflects a combination of reduced mining activity and/or improved adherence to environmental pollution guidelines (Seen et al. 2004).

Since trace metal concentrations began to decrease, however, there has been no corresponding increase in the proportion of planktonic taxa. Instead, the planktonic:benthic ratio became lower than at any time in the last *ca.* 350 years (Fig. 5). Previous studies found that a reduction in pollution from mining did not result in complete recovery of diatom communities (Greenaway et al. 2012). This may also be the case in Bathurst Harbour. In addition, rainfall since AD 1980 has decreased steadily, which has likely contributed to the decline in planktonic taxa. Further studies of the links between diatom species and environmental variables (Vyverman et al. 1995), and long-term monitoring of the biology and hydrology of the estuary are required to fully understand these changes.

Nevertheless, the data from this and other studies of Bathurst Harbour (Edgar and Cresswell 1991) suggest that any reduction in rainfall poses a risk to Bathurst Harbour's aquatic ecosystems because less rainfall will cause less riverine runoff, a reduction in the depth of the halocline and increased light penetration through the brackish, tannin-stained surface layer (Edgar et al. 2010).

Current climate projections for Tasmania indicate that annual rainfall may decrease by less than 5 % across Tasmania, but there may be a more than 15 % decrease on a seasonal basis (Grose et al. 2010). Importantly for Bathurst Harbour, the biggest decline in rainfall is predicted to be in western and central Tasmania in summer (Grose et al. 2010). The summer is when stratification is weakest (Edgar et al. 2010) and further declines in rainfall have the potential to extend periods of reduced stratification, creating unfavourable conditions for the natural aquatic ecosystems in Bathurst Harbour. It is not known, however, at what threshold these reductions in rainfall would result in further declines or seasonal disappearances of planktonic taxa. Further work, including analyses of diatoms in a longer core and the development of detailed rainfall reconstructions, is needed to address this issue and to better understand the impact of rainfall variability on stratification and the response of the diatom community.

In addition, although mining activities have generally declined and better environmental guidelines have been implemented, mining remains an important part of Tasmania's economy. Some small-scale exploration and development of relatively short-term (<10 years) extraction projects has occurred recently (MRT 2012), so further mining impacts cannot be ruled out. If mining activity and associated atmospheric pollution were to increase, this may compound the effect of reduced rainfall on the diatom assemblages in Bathurst Harbour.

Conclusions

This study provides insights into both long- and short-range pollution from mining activities and the importance of precipitation for maintaining stratification and natural ecosystem variability in one of the most pristine estuaries in southern Australia. Trace metal analyses showed that concentrations of Cu, Pb, Sn and Zn nearly tripled post-AD 1870 as a result of Tasmanian mining activities and that past maxima in Ni exceeded current Australian and New Zealand sediment quality guidelines. Diatom analyses indicated stratification has likely persisted in Bathurst Harbour for at least the last *ca.* 350 years and the majority of taxa are more characteristic of the sub-Antarctic than coastal Tasmanian and southeast mainland Australian coastal water bodies. The proportion of planktonic taxa declined after the onset of mining, with the exception of two peaks in the twentieth century, which coincided with high rainfall periods. Since the peak in mining activity in the AD 1960s, concentrations of trace metals have decreased, yet the proportion of planktonic taxa continued to decline and recently reached its lowest values in the last *ca.* 350 years. This is likely a consequence of low rainfall measured since the AD 1980s, and suggests that further decreases in rainfall, particularly in summer when stratification is already weakest, have the potential to substantially change the balance between freshwater and brackish influences on Bathurst Harbour's natural ecosystem.

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