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Abstract
The Hunter and Latrobe Valleys have two of the richest coal deposits in Australia. They also host the largest coal-fired power stations in the country. We reconstructed metal deposition records in lake sediments in the Hunter and Latrobe Valleys to determine if metal deposition in freshwater lakes have increased in the region. The current regulatory arrangement applied to metal emissions from coal-fired power stations in Australia are presented, discussing their capacity to address future increases in metal deposition from these sources. Sediment records of spheroidal carbonaceous particles (SCPs), a component of flyash, were also used as an additional line of evidence to identify the contribution of industrial activities related to electricity generation to metal deposition in regions surrounding open-cut coal mines and coal-fired power stations. Sediment metal concentrations and SCP counts in the sedimentary records, from the Hunter and Latrobe Valleys, both indicated that open-cut coal mining and the subsequent combustion of coal in power stations has most likely resulted in an increase in atmospheric deposition of metals in the local region. In particular, the metalloids As and Se showed the greatest enrichment compared to before coal mining commenced. Although the introduction of bag filters at Liddell Power Station and the decommissioning of Hazelwood Power Station appear to have resulted in a decrease of metal deposition in nearby lakes, overall metal deposition in the environment is still increasing. The challenge for the years to come will be to develop better regulation policies and tools that will contribute to reduce metal emissions in these major electricity production centres in Australia.

1. Introduction

Metal contamination has markedly increased in water bodies proximal to where coal-fired power stations have been constructed and operated (Meij and te Winkel, 2007; Schneider et al., 2014a). Many studies in Europe (Barbante et al., 2004), North America (Donahue et al., 2006; Ouellet and Jones, 1983) and Japan (Han et al., 2002) have demonstrated a significant increase in metal contamination in depositional environments, however, little is known about how metal concentrations have increased in the Australian environment since the commissioning of open-cut coal mines and associated coal-fired power stations. Identifying historical trends in metal concentrations in Australia since the commissioning of coal-fired power stations is, therefore, critical for informing policy for the future management of coal-fired power stations in the country.

Coal mining has defined Australian’s economy through providing a critical energy supply to Australia and other coal-dependent economies, particularly in Asia (Cottle and Keys, 2014). Coal is central to both Australia’s domestic energy requirements (accounting for 62% of total electricity generated in 2016-2017 DEE, 2018) and also to the national economy as it represents the largest Australian export commodity (Geoscience Australia, 2019a). Australia is currently the biggest net exporter of coal internationally with 32% of global exports (389 Mt out of 1,213 Mt total), and is the fourth-highest producer, with 6.9% of global production (503 Mt out of 7,269 Mt total) (IEA, 2017).

The mining and subsequent combustion of coal generates significant environmental challenges. These activities release metals and metalloids (hereafter collectively referred to as metals) to the environment both via the dust produced during the extraction of coal (open-cut coal mines), as well as during the combustion process (Tang et al., 2012). Metal emission rates are, therefore, dependent on the total amount of metal present in the coal, the amount and method of coal mined and combusted and the type of pollution control devices employed within power stations (Gade, 2015).

Coal-fired power stations are ageing in Australia and it is rare for modern devices to control particle (and consequently metal) emissions to be retrofitted (Schneider and Sinclair, submitted). By 2030, around half of the 24 coal-fired power stations in Australia will be over 40 years old, with some stations having operated for nearly 60 years (Stock, 2014). The most modern metal emission control devices used in these power stations are filter bags in New South Wales (NSW), and electrostatic precipitators in Victoria (VIC).

Once metals are released to the atmosphere, they return to the surface environment by both wet and dry depositional processes. As soils and lake sediments are two important sinks for deposited metals (Yang et al., 2016), lake sediments are widely used to reconstruct metal contamination histories (Schneider et al., 2015a). Historical records of regional trends in atmospheric metal deposition are usually derived from high altitude lake sediments, where atmospheric deposition is the only input of anthropogenic metals into the aquatic system

(Fitzgerald et al., 2005; Yang et al., 2010). In Australia, due to the low mean annual runoff, perennial (e.g. permanent lakes) water bodies are rare. Artificial dams and reservoirs, however, have been constructed since 1857 (Chanson, 1999) and the sediments of these reservoirs provide a unique opportunity to trace changes in metal concentrations over time from atmospheric and catchment sources. In addition, the sediments can indicate the extent of metal exposure to biota in aquatic systems.

Little information on past levels of industrially-derived air pollution is available in Australia, and the few studies published have mainly focussed on estuaries (Lafratta et al., 2019; Schneider et al., 2014a, 2014a, 2015b, 2015c; Serrano et al., 2016) . Furthermore, we still have little understanding of how various management practices in coal-fired power stations (e.g., retrofitting of pollution control devices) have affected the deposition of metals in the environment.

This study investigates the historical metal and metalloid contamination records of dams in the Hunter and Latrobe Valleys, to assess if metal concentrations in these environments have increased since the establishment of coal-fired power stations and associated open-cut coal mining. Temporal metal contamination from both atmospheric and catchment sources are measured in sediment cores collected from dams adjacent to open-cut coal mines and power stations. Spheroidal carbonaceous particles (SCPs), a component of fly-ash only derived from the high combustion temperature of coal and oil fuels (Rose, 2001), are also measured in sediments as supporting evidence for metal sources. Finally, we discuss the current regulatory arrangement applied to metal emissions from coal-fired power stations in Australia, and their capacity to address future increases in metal deposition from these sources.

2. Methods

In Australia, coal mining occurs mainly in Queensland, New South Wales and Victoria (Mohr et al., 2011). The major coal deposits in these areas have resulted in the largest coal-fired power stations being developed in these regions, including the Hunter and Latrobe Valleys (Figure 1). We selected dams that are proximal to coal-fired power stations in these two valleys to study the temporal trends in metal deposition and the relationship between these trends and the establishment and operations of coal-fired power stations (Figure 1).

There are two types of coal mined in Australia. Bituminous black coal, with varying volatile matter and ash content, is about 250 million years old. This type of coal is mostly mined in the state of New South Wales (in the Sydney-Gunnedah Basin) (NSW Planning and Environment, 2019), and supplies coal-fired power stations in the Hunter Valley (Table 1). Power generation in the state of Victoria is mainly in the Latrobe Valley, (Table 1) and uses brown coals from the Gippsland Basin (Australia Government Regional Assessment Program, 2019). Lignite brown coal is about 15–50 million years old and has a high water content (Australia Government Regional Assessment Program, 2019; Nelson, 2007).

2.1. The Hunter Valley

Sediment cores were collected from Lake Glenbawn (32.0940° S, 150.9891° E), situated approximately 30 km northwest of Liddell and Bayswater coal-fired power stations and open-cut coal mines (Figure 1, Table 1). No coal-mining activities exist within the Lake Glenbawn catchment area.

Sediment cores were collected from a large punt in May 2018 using a 50 cm length polycarbonate tube mounted in a gravity corer .The gravity corer and tube was lowered 30 metres using an onboard davit and winch to the lake floor, where it penetrated the sediment.

Disturbance of the core water-sediment interface was minimal and maximum penetration into the sediment was 32 cm. The davit and winch were then used to slowly bring the sediment core back to the surface, where it was sliced into 1 cm segments, stored in Ziplock bags and transported to the Australian National University cold room for storage at 4° C.

Lake Glenbawn was completed in 1958 by the New South Wales Water Conservation & Irrigation Commission to supply water for irrigation and flood mitigation. With a catchment area of 1,226 km^2, the dam was formed by blocking the Hunter River, a sand and cobble-bed river that drains to the eastern coast of NSW (Erskine and Livingstone, 1999). Its catchment is dominated by Triassic sediments to the south, and Devonian and Carboniferous rocks to the northeast (Chessman et al., 1997). These sediments have eroded over millennia to generate rich alluvial plains underlain by shallow aquifers closely associated with the river. The main effect of Lake Glenbawn on the Hunter River has been a reduction in the size and frequency of small to medium flows, and the maintenance of steady low flows when the river might otherwise be dry (Hancock and Boulton, 2005).

Rainfall for the Muswellbrook (27 km south of Lake Glenbawn) averages 600 mm per year (Stone et al., 2018). As regional variation in metal deposition is dependent on precipitation rate (Sorensen et al., 1994), metal atmospheric emissions from Liddell and Bayswater power stations can travel long distances as the low rainfall reduces wet deposition.

2.2. Latrobe Valley

The Latrobe Valley is the major power production in Victoria, producing electricity from the combustion of predominantly brown coal. The valley produces approximately 85% of the state’s electricity and also supplies some electricity to New South Wales and Tasmania (Weller et al., 2011). Coal-fired power plants located in the Latrobe Valley include

Hazelwood Power Station (decommissioned in 2017), Loy Yang Power Stations A & B, Yallourn Power Station and Yallourn Power Station (Table 1, Figure 1).

A sediment core was taken from the Traralgon Railway Conservation Reserve (38°12'35.35"S, 146°31'47.32"E), in Traralgon, VIC (Figure 1). The field work campaign was conducted in April 2018, and sediment samples were collected using a piston core fitted with a 1 m polypropylene tube. A 43 cm core was retrieved at 3 m water depth. The core was sliced into 1 cm, stored in Ziplock bags and transported to the Australian National University cold room for storage at 4º C.

The Reserve has an area of 0.295 km^2 and is largely made up of varied bush and grassland. The dam (Traralgon Reservoir) was built in the centre of the Reserve in 1883 for the original purpose of providing water for steam trains (Latrobe City, 2019). As for Lake Glenbawn, Traralgon Railway Reservoir does not have any coal-mining activities within its catchment.

The reservoir is in the Traralgon Creek catchment which is a deeply incised valley. The catchment area for Traralgon Reservoir is small, approximately 1.5 km^2, and is located at the headwaters of the creek. There are no springs supplying water to the Reserve and water for the reservoir has its origin though runoff from a farmland gully, on the southern boundary (Figure 1). The reservoir outlet is through the dam weir, flowing through a gully in the reserve, into Traralgon Creek and then the Latrobe River. The mean annual rainfall (mm) for years 1984 to 2019 is 735.5 mm (Australian Bureau of Meteorology, 2019).

Local open-cut coal mines (Figure 2), which supply coal for local power stations and for export, also contribute to increased metal deposition in both the Latrobe and Hunter Valleys. Most of the coal mines in Australia are open-cut, which uses large-scale mechanisation which releases dust and gases to the atmosphere (Dhar, 1994).

2.3. Precipitation

Precipitation data were sourced from the Australian Bureau of Meteorology (BOM, 2019a). Observations from Scone Airport (1926 to 2018), 13 km west of Lake Glenbawn, and Morwell airport (1985 to 2018), 5 km west of Traralgon Railway Reservoir, were used in this study.

The precipitation ratio was calculated by averaging annual rainfall for 3 years (the year before, the current year and the year after the average age in a given sediment layer) and then dividing by the long-term average. Values above 1 represent periods where rainfall was above the long-term average. Values less than 1 represent periods where rainfall was below the long-term average.

2.4. Spheroidal carbonaceous particles (SCPs)

Sediment samples were analysed for SCPs using sequential treatments of nitric, hydrofluoric and hydrochloric acids to remove organic, siliceous and carbonate fractions respectively (Rose, 1994). A known fraction of the final suspension in water was then evaporated onto a coverslip, mounted onto a glass slide, and the number of SCPs were counted using a light microscope at 400 times magnification. Standard criteria for SCP identification followed Rose (2008) and concentrations were calculated as the number of particles per gram dry mass of sediment (gDM^{-1}). SCP fluxes were calculated as the product of SCP concentration and bulk dry sediment accumulation rate (number of particles per cm^2 per year; cm^{-2} yr^{-1}). Analytical blanks and SCP reference materials (Rose 2008) were included with all sample digestions. The detection limit for the technique is typically less than 100 gDM^{-1} and calculated concentrations generally have an accuracy of c. ± 45 gDM^{-1}.

2.5. Grain size analyses

All sections of the cores were analysed for sediment grain size by first sieving to a particle size of <2000 mm (i.e. gravel-free). Grain size analyses were performed using a Malvern Mastersizer 2000 with a wide angle detection system between 0.1 and 2,000 μm. Samples were dispersed in water and ultrasonicated for 30 s to break up agglomerated particles. A total of three measurements were made for each sample and the average used as the final value. Particle size is expressed as volume mean diameter. Calculations are presented in Supplementary Information.

2.6. Metal analyses

Sediment digestions were performed using 0.2 g of freeze-dried material, to which 2 mL of concentrated nitric acid (Aristar, BDH) and 1 mL of 30% concentrated hydrochloric acid (Merck Suprapur, Germany) were added, following the method of Telford et al. (2008). More details are presented in Supplementary Information.

Samples were analysed using inductively coupled plasma mass spectrometry (PerkineElmer DRC-e) with an AS-90 autosampler (Maher et al., 2001). The certified reference materials NIST- 2710a (Montana Soil I) and NIST- 8704 (Buffalo River sediment) were used as controls to check the quality and traceability of metals. Measured concentrations were in agreement with certified values (Supplementary Table 1). The Ecochemistry Laboratory confidence limit for metal concentration analyses is 0.01µg/g dry mass. Sediment flux was calculated as the cumulative dry mass (g/cm^3) times sedimentation rate (cm/yr) times metal concentration (mg/kg) and are reported as µg/yr/m^2.

2.7. Enrichment factor (EF)

The calculation of a normalized enrichment factor (EF) for metal concentrations above uncontaminated background levels enables an estimation of anthropogenic inputs of metals to lake bed sediments (Abrahim and Parker, 2008). The EF calculation seeks to reduce the variability of metal concentrations associated with fluctuations in clay/sand ratios and is a convenient tool for plotting geochemical trends across large geographic areas and within lakes, which may have substantial variations in the sediment (i.e. clay to sand ratios).

The EF method normalises the measured metal concentration with respect to a sample reference element which is used as a “proxy” for the clay content (Cevik et al., 2009). In this study, Al and Mg were chosen as reference elements to calculate the EF in Lake Glenbawn and Traralgon Reservoir, respectively. These elements were chosen because they are the major elements least affected by emissions from coal-fired power stations and coal mining and because they are relatively conservative tracers of the natural metal-bearing phases in sediments (Schiff and Weisberg, 1999; Wilcke et al., 1998). Iron content in Australian coal is known to be high (Ali et al., 2017; Borsaru et al., 1993; Ilyushechkin et al., 2011), and therefore was not used for normalisation.

The EF calculation method is presented in the Supplementary Information. An EF < 1= no enrichment, EF 1-3 = minor enrichment, EF 3–5 = moderate enrichment, EF 5–10 = moderately severe enrichment, EF 10–25 = severe enrichment, EF 25–50 = very severe enrichment, and EF > 50 = extremely severe enrichment (Cevik et al., 2009).

2.8. Sediment chronologies

Samples for lead-210 (^{210}Pb) dating were processed at the Australian Nuclear Science and Technology Organisation (ANSTO) by alpha particle spectrometry, following methods described by Harrison et al. (2003). Details on this method are presented in Supplementary Information.

The age-depth models used in this study were obtained by the *Plum* model (Aquino-López et al., 2018). This approach uses a Statistical-Bayesian framework to obtain sediment chronologies. *Plum* uses an autoregressive gamma process, presented by Blaauw and Christen (2011), together with an assumption of a constant supply of ^{210}Pb to the sediment to infer an age-depth function. More information on the Plum package is provided in the Supplementary Information.

2.9. Statistical Analyses

All analyses were performed using R Statistical Software (R Development Core Team, 2008). The respective libraries for the particular analyses are cited below. The calculation of catchment area was conducted using a Digital Elevation Model (DEM), which derives stream networks based on terrain elevation using the R package OpenSTARS (Kattwinkel and Szocs, 2018).

The sediment age-depth models were obtained using the *Plum* model (Aquino-López et.al 2018), which has a corresponding R-package that can be obtained at https://github.com/maquinolopez/Plum. Stratiplots with metal profiles plotted against age were produced using the R package analogue (Simpson, 2007). Principal Component Analysis (PCA) of log-transformed data (ln x) was used to explore the similarity of metal concentration in the lakes before and after coal-fired power stations were commissioned.

PCA plots were produced using the function prcomp with the metal scores indicated by ggbiplot arrows (Vincent, 2011).

3. Results and Discussion

3.1. Age depth model and sedimentation rate

Age-depth models from Lake Glenbawn and Traralgon respectively, with age 0 equivalent to the collection dates in 2018, are shown in Supplementary Figures 1 and 2. Unsupported ^{210}Pb activities in the Traralgon Reservoir sediment core decreased with depth between 0 and 20 cm (Supplementary Table 2A-B). Between 20 and 40 cm depth, the total ^{210}Pb activities are constant (i.e., no longer decreasing with depth). Supported ^{210}Pb was estimated from the concentrations of ^{226}Ra using Plum to obtain the age-depth model (Supplementary Figure 2).

In Lake Glenbawn, the hydrological system and subsequent sedimentation rate was modified in 1957, when the Hunter River was dammed. For this reason, the age-depth model was constructed only down to 28 cm (the total core length was 34 cm). The sedimentation rate in Lake Glenbawn generally had a mean of 0.5 cm/yr, with changes directly after the construction of Lake Glenbawn (Supplementary Figure 1). No major sedimentary changes have occurred in Lake Glenbawn after the dam reaction time (the period between the onset of the change or disruption and the beginning of channel adjustment) (Supplementary Figure 1). This finding is supported by a previous study in Lake Glenabawn (Erskine and Bell, 1982), which reported a short reaction time at this dam. A short reaction time leads the river channel to reach equilibrium relatively fast, minimising erosion of the sediment above the dam and favouring a stable channel morphology (Pizzuto, 2002). The short reaction time is supported by the geological nature of the upper Hunter River, which is a bedrock-controlled channel with coarse materials (Erskine and Bell, 1982).

The low sedimentation rate in Lake Glenbawn is also partially a result of extensive operations by the NSW Soil Conservation Service on soil conservation that resulted in a progressive decrease in the area affected by soil erosion within the catchment (Erskine and Bell, 1982). In addition, deforestation by clearing and ringbarking in the Hunter Valley occurred mainly in the last century and before 1949 (Erskine and Bell, 1982). Given the stable nature of Lake Glenblawn catchment, erosion and catchment inputs is unlikely to have overridden the historical signatures of atmospheric metal deposition in this lake.

Traralgon Reservoir sediment rate results also demonstrates the unlikely occurrence of major erosion events in the catchment, as this dam has a small catchment size (1.5 km^2) and water in the reservoir originates from a gully runoff less than 500 m away. Sedimentation rate in this core is around 0.5 cm/yr at the bottom and 1 cm/yr in the middle-top of the core (Supplementary Figure 2).

No previous study is available on the effects of dam construction on the sedimentation rate of Traralgon Railway Reservoir. Our core, which covers the period back to 1940, indicates a fairly constant sedimentation rate after dam construction with little evidence to suggest that there have been major hydrologic shifts in this reservoir (e.g., drought or floods) as indicated by the grain size analyses presented below. This dam, therefore, is unlikely to be significantly affected by land-use changes in the catchment.

3.2. Metal concentrations

3.2.1. Lake Glenbawn

In this study, the major elements, Al and Mg, have been used as indicators of catchment erosion inputs, as both are typical lithogenic elements that are released by weathering of the local bedrock (Price et al., 1999; Wilcke et al., 1998). Iron, commonly used as a normalising agent, has not been used in this study as it is known to occur in relatively high concentrations in Australian coal (Ali et al., 2017; Borsaru et al., 1993; Ilyushechkin et al., 2011), and consequently could bias our interpretation of catchment erosion. Aluminium and Mg have a very distinctive deposition rate, independent of the effect of coal mine and coal-fired power station activities (Figure 3A-B) and illustrate that atmospheric transport and deposition has not significantly contributed to Al and Mg concentrations in the sediments of these two lakes.

Aluminium and Mg concentrations in the sediment have increased since the construction of Lake Glenbawn and the dam's reaction time (Figure 3A), Supplementary Table 3). These results are consistent with previous studies showing that lithogenic elements reflecting the geology of the lake catchment increase in bed sediments after dam construction (Hahn et al., 2018). No major erosion, however, has occurred in the Lake Glenbawn catchment once the dam reaction time was over, demonstrated by the constant grain-size of the sediment (Figure 3A). Had major erosion occurred in the area, a change in grain-size would likely be detected as a result of the introduction of newly transported material (Laceby et al., 2017; Singer and Anderson, 1984).

Metal concentrations and fluxes (Co, Cu, Zn, As, Se and Pb) in the Lake Glenbawn sediment core have increased from the 1970s (Figure 3A-B, Supplementary Table 3), corresponding with the commissioning of the Mount Arthur open-cut coal mine in 1968 and the Liddell coal-fired power station in 1971. Metal concentrations reached their maxima in approximately 1985, which corresponds to the commissioning of the Bayswater coal-fired power station. A decrease in metal concentrations is observed at ~1990, which corresponds to Liddell power station being retrofitted with bag filters. Unlike Bayswater, Liddell's power plant bag filters were not fitted at the time of its construction. The decrease in metal concentrations at the time of retrofitting bag filters suggests this has contributed to the decline in metal emissions and deposition in Lake Glenbawn.

From 2000 onwards, increased sediment metal concentrations and metal fluxes are likely a result of both the large expansion of the open-cut works at the Muswellbrook and Mount Arthur coal mines (Bioregional Assessments, 2015; Idemitsu, 2019) and increased electricity demand, that required greater quantities of coal to be burnt (Figure 3B).

The lack of correspondence between Al and Mg concentrations (indicators of catchment erosion) and Co, Cu, Zn, As, Se and Pb concentrations (indicators of anthropogenic inputs) suggests that metal deposition in the dam and its catchment are predominantly from atmospheric inputs.

Spheroidal carbonaceous particle (SCP) concentrations in the sediment began to increase in the 1970s, corresponding to the commissioning of Liddell Power Station in 1971. Overall, SCP concentrations increase in the sediments of Lake Glenbawn. The decline in SCP concentrations in 1990 could be due to the retrofitting of bag filter particulate control devices to Liddell Power Station at that time. In addition, a decline in SCP concentrations is seen in the period when coal consumption declines, between 2008 and 2014 (DEE, 2018). Both declines in SCPs, however, are based on single data points and therefore should be interpreted with caution.

The results of the PCA further suggests that coal-mining and subsequent burning in the Hunter Valley are strong contributors to metal concentrations in the sediments of Lake Glenbawn (Figure 4). Sediment metal concentrations from before the commissioning of coal-fired power stations are in the low metal concentration zone of Axis 1 (PCA1), while sediment samples from after coal-fired power station commissioning are located in the high metal concentration zone. The most recent samples are closer to the high concentration zone, corresponding to higher consumption and burning of coal due to increased demand for electricity (Figure 3 and 4).

Aluminium and Mg are clustered separately from the other metals in the biplot shown in Figure 3. Aluminium and Mg are indicative of normal erosion processes of soil in the catchment. In contrast increases in metals commonly found in higher concentrations in coal ash and coal dust suggest that the catchment soils have been contaminated since the establishment of coal mining and burning activities through atmospheric deposition. In the Upper Hunter Valley the major sources of air particulate metals are coal mining activities, power generation, biomass burning and top soils (Hibberd et al., 2016), however, biomass burning and top soils are considered to be relatively minor sources of metals (Smith et al., 2011). Thus, it is suggested that coal mining and consecutive burning for electricity production are the main drivers of increase in metal concentrations in Lake Glenbawn.

3.2.2. Traralgon Railway Reservoir

Metal concentrations associated with coal mine activities increased in the bed sediments of Traralgon Railway Reservoir after 1964 (Figure 5A), which coincides with the timing of the commissioning of the Hazelwood power station (the first coal-fired power station in the Latrobe Valley region). Metal concentrations then further increased, correlating with the commissioning of other coal-fired power stations and open-cut coal mines in the Latrobe Valley i.e. Yallourn in 1975, Loy Yang A in 1984 and Loy Yang B in 1993 (Figure 5A).

Sediment metal concentration profiles in the Traralgon Reservoir core are similar to the trend in annual brown coal consumption rates in Australia. The data presented in Figure 5B is for brown coal consumption for the entire country, which we assume is representative of coal consumption in Victoria because 92% of the electricity produced by burning brown coal in Australia occurs in that state (Geoscience Australia, 2019b).

As for Lake Glenbawn, sediment Mg and Al concentrations and fluxes in Traralgon Railway Reservoir have greater variability in the core than Co, Cu, Zn, As, Se and Pb concentrations and fluxes, demonstrating a minor influence of catchment runoff to the input of metals (Figure 5A-B) in this reservoir. Furthermore, grain-size is fairly constant through the core, indicating no major run-off or erosion has occurred in this lake over the period covered by the core (Figure 5A). Spheroidal carbonaceous particle concentrations follow the general trend of metal concentrations associated with coal-related activities, higher concentrations occurring in the most recent years. These high concentrations are possibly the result of a fire in the Hazelwood open cut coal mine in 2014 (Lord, 2014), but more data are necessary to confirm this.

As for Lake Glenbawn, the PCA plot for the Traralgon Railway Reservoir core (Figure 6) further supports our hypothesis that coal mining and subsequent burning in power stations have resulted in increased metal concentrations. Metal concentrations in sediments from before the commissioning of coal mines and coal-fired power stations are in the low metal concentrations zone of Axis 1 (PCA1) (Figure 6), while samples related to the time after the commissioning of coal-fired power stations are in the high metal concentration zone of Axis 1 (PCA1). In addition, similar to the biplot for Lake Glenbawn, lithogenic elements (Al and Mg) are clustered separately from the other metals (except for Se). This suggests that the metal trends (Cu, Co, Pb, Zn, As) in the bed sediments of Traralgon Railway Reserve are not driven by catchment run-off or erosion. Therefore, it is unlikely that climate or agricultural development are influencing the metal trends in the sediments of these lakes.

3.3. Enrichment factors (EF)

EF values in the sediments of Lake Glenbawn increased after the dam was established and coal-fired power stations were commissioned in 1971 (Figure 7, Supplementary Table 3). By 2018, all metals associated with coal had minor enrichment (EF = 1-3), except Se which was moderately enriched (EF = 5-10).

Traralgon Railway Reservoir sediments were more enriched in anthropogenic metals than Lake Glenbawn, with all metals having a minor enrichment factor (EF = 3-5), Zn, Se and Pb reaching a moderately severe enrichment factor (EF = 5-10) and As reaching a severe enrichment factor (EF = 10-25) (Figure 8; Supplementary Table 3).

For both Glenbawn and Traralgon, As and Se concentrations have undergone the greatest enrichment and this is probably due to As and Se being present in high concentrations in Australian coal (Dale, 2006) as well as being more volatile than other metals (Schneider et al., 2016). Elsewhere, these elements have been found to exceed environmental guideline levels for sediments in water bodies near coal-fired power stations (Schneider et al., 2014a, 2015b).

The greater EF values found for metals in Traralgon Railway Reservoir can be explained by two factors. Firstly, Traralgon Railway Reservoir is predominantly downwind from the power stations (Figure 2), while Lake Glenbawn is predominantly upwind (Figure 2). Therefore, although Lake Glenbawn is positioned to capture some metal emissions from open-cut mines and from Liddell and Bayswater Power Stations, it is not as well positioned as Traralgon Railway Reservoir. Secondly, power stations in the Latrobe Valley do not have bag filters installed as pollution control devices unlike in the Hunter Valley (Table 1). The lack of bag filters would result in greater metal emissions and consequently higher EF values in the Latrobe Valley.

Baghouses have been used extensively in Canada, Europe, Japan, and the United States because they are efficient at dust collection (Tavoulereas and Charpentier, 1995; Zhao et al., 2014). The advantage of bag filters over the electrostatic precipitators (ESPs) used in the Latrobe Valley power stations is that bag filters have a more stable dust collection efficiency, and a collection efficiency above 99%, even for particles in the 0.05 to 1.0 µm range (Tavoulereas and Charpentier, 1995).

In general, sediment metal concentrations and SCP counts have increased and continue to increase in both Lake Glenbawn and Traralgon Railway Reservoir sediments. This result runs contrary to trends in other developed nations where studies have shown that metal emissions have been declining in Europe and North America over the last two decades due to changes in fuel use (e.g. from coal to gas), better regulations and implementation of better pollution control devices and practices (Fitzgerald et al., 2018; Rühling et al., 1992). In Asia, where regulation and mitigation strategies for reducing airborne metal contaminants are limited, metal emissions are still increasing (Amann et al., 2013; Klimont et al., 2013; Kurokawa et al., 2013). A similar pattern of continuous increase of metal emissions is found for Lake Glenbawn and Traralgon Railway Reservoir regions.

3.4. Comparison of metal concentrations and ANZG (2018) sediment quality guidelines

The Australian and New Zealand Sediment Quality Guidelines (ANZG, 2018) established the default guidelines value (DGV) as indicative of concentrations below which there is a low risk of unacceptable ecosystem effects occurring. Together, with other lines of evidence, these guidelines should be used to protect aquatic ecosystems. While these guidelines provide DGV for Cu, Zn, As and Pb, no Se guideline is available for sediments in Australia. We therefore use the Screening QuickReference Tables (SQuiRTs) developed by the National Oceanic and Atmospheric Administration (NOAA) (Buchman, 2008) to assess Se contamination in sediments.

Table 3 shows the DGV threshold limit and the maximum concentration in sediments of Lake Glenbawn and Traralgon Railway Reservoir. Metal concentrations in sediments of these two lakes are still below the DGV. This is an encouraging result but action is required to prevent exceedances in the future, as metal concentrations have significantly increased in both lakes since the commissioning of coal-fired power stations.

Of particular concern is the increase in Cu and As concentrations in Lake Glenbawn and Se concentrations in both Lake Glenbawn and Traralgon Railway Reservoir, which have significantly increased from background values (Table 3) and are now close to the ANZG (2018) DGV threshold limits (Table 4). These elements have already been shown to be an environmental issue in other Australian lakes located next to coal-fired power stations, for example, in Lake Macquarie and Lake Wallace, where Se and As concentrations in sediments and sea food were found to be above the dietary levels considered to induce detrimental health effects to the environment (Jasonsmith et al., 2008; Schneider et al., 2018, 2014b).

If we assume that the same coal mining and coal burning rates and climate conditions that have happened over the past 10 years continues, then most metal concentrations in Glenbawn and Traralgon sediments will reach the ANZG threshold guideline value within the next 13 years (Table 4). However, these estimations should be interpreted with caution, as conditions are likely to change particularly if Australia decides to invest more in green energy technologies and production.

3.5. Regulation of metal air emissions from coal-fired power stations in Australia

Metal concentrations in sediments of Lake Glenbawn and Traralgon Railway Reservoir are within the Australian and New Zealand DGV sediment quality guidelines. However, these concentrations have risen significantly since the commissioning of local coal-fired power stations in the 1960s and are likely to continue increasing if metal emissions are not curbed. Given the potential harm of metal emissions to human and environmental health, it is prudent to ensure that air quality regulations are adequate to reduce future atmospheric metal depositions from coal-fired power stations. Here we discuss the capacity of existing regulations to achieve this aim. These existing regulations in Australia are summarised in Figure 9.

The federal government oversees the National Environmental Protection Measures (NEPMs) and the National Pollutant Inventory (NPI). Neither NEPMs nor the NPI impose direct limits on emissions at coal-fired power stations. The NEPMs are aspirational limits, designed to provide guidance to the States and Territories on how to manage and regulate ambient air quality. Consequently, state governments have significant discretion to decide how and in what ways they implement NEPMs (NEPC, 2019). The NPI is an internet database administered by the Commonwealth Department of Agriculture, Water and the Environment that provides publicly available information on the types and quantities of pollutants emitted to the environment by industrial and commercial sources (DEE, 2017).

The responsibility for regulating metal emissions from coal-fired powered stations rests with the States and Territories. This has resulted in different regulatory approaches between States. We address below the experiences of NSW and Victoria, two states in Australia that are heavily dependent on coal-fired power stations for electricity generation, and within which the Hunter and La Trobe Valleys are located, respectively.

In NSW, the *Protection of the Environment Operations (POEO) Act* 1997 and *Protection of the Environment (Clean Air) Regulation 2010* is administered by the NSW Environment Protection Authority (EPA), and addresses the NEPMs. Under this act, the EPA issues Environmental Protection Licences (EPLs) for individual coal-fired power stations, including Bayswater and Liddell (EPA NSW, 2018). While the POEO Act provides pollutant emission limits, EPLs allow for limits tailored to the circumstances of individual power stations.

In Victoria, EPA Victoria is responsible for regulating air emissions from coal-fired power stations under the *Environment Protection Act 1970* and *Environment Protection Act 2017*. It issues pollution control licences for Loy Yang A, Loy Yang B and Yallourn. Power stations in Victoria require a licence that sets emission limits. These power stations face prosecution for breaches of the *Environment Protection Act 1970* (see also the planned changes in the *Environment Protection Amendment Act 2018*).

Overall, the state and federal regulatory frameworks have three issues that are relevant to future efforts to reduce emissions. These are: (i) the coverage of metals in the regulations, (ii) the regulation of management technologies, and (iii) independent validation of monitoring data.

The coverage of metals in regulations

At the national level, current NEPMs include only carbon monoxide, nitrogen dioxide, ozone, sulphur dioxide, lead and particle matter (PM_{10} and $PM_{2.5}$) (NEPC, 2019). However, the regulations for metals varies between states. In NSW, metals are regulated through the *Protection of the Environment (Clean Air) Regulation 2010*. This sets concentration limits for emissions of Sb, As, Cd, Pb, Hg, Be, Cr, Co, Mn, Ni, Se, Sn and V. These limits are aggregated across multiple activities, except for Cd and Hg which are addressed individually for scheduled activities, including coal-fired power stations.

In contrast, EPA Victorian licences currently only set limits for carbon monoxide, chlorine compounds, fluorine compounds, oxides of nitrogen, sulfur dioxide, sulfur trioxide and particulates (EPA VIC, 2017), although they do need to monitor and report on As and Hg under required under the NPI.

Regulation of management technology implementation

There has been installation of different remediation technologies in different states. For example, fine particle bag filters have been installed in power stations in NSW whilst Victorian power stations have employed electrostatic precipitation (which is a less effective remediation technology (Tavouleréas and Charpentier, 1995)).

In addition, the state regulatory frameworks do not specifically regulate the installation of air pollution remediation technologies. Instead, regulations impose concentration limits and leave it to individual companies/sites to determine what technology they use to comply with these limits. For example, section 128 of the NSW POEO states that activities or plants cannot exceed the standard of concentration provided in the Clean Air Regulation schedules. The rationale is that such performance standards allow for (i) individual sites to choose the technology to suit their individual circumstances, and, (ii) the development of innovative responses.

Independent validation of monitoring data

Finally, monitoring data of atmospheric emissions from coal-fired power stations reported to the NPI lacks independent validation or auditing, beyond reviews by government agencies. This has led to concerns about the completeness of the data being reported. A related issue is that the NPI uses *indicative* metal concentrations for brown and black coals, rather than *actual* metal concentrations, thus generating uncertainty about the data reported. For example, an assessment of NPI data on mercury reported in 2004-2005 found lower than expected Hg emissions from coal-fired power stations being reported due as a result of the use of indicative metal concentrations (Nelson, 2007).

4. Conclusion

Studies assessing atmospheric metal emissions and deposition in the environment are lacking in Australia. In this study, sediments from dams proximal to open-cut coal mines and power stations provided important insights into the fate of atmospheric metal emissions and evidence of the degree to which metal concentrations have changed relative to background levels since the commissioning of coal mines and power stations in the Hunter and Latrobe Valleys.

This study highlights the value of dam sediments as a record of past contamination in dry areas such as mainland Australia. Increases in metal concentrations in these dams correlated with the timing of the commissioning of coal mines and coal-fired power stations and the change in technologies employed in the Hunter and Latrobe Valleys. In particular, the significant Enrichment Factors of As and Se make these two elements of particular concern and they should be the focus of metal emission control activities in the two regions.

While the metal concentrations in sediments of these two dams are still under the Interim Sediment Quality Guidelines ISQG ANZECC/ARMCANZ (2018), the steady temporal increase of metal contamination calls for action to avoid reaching a level of environmental health concern. The trends in metal emissions from the power stations near Lake Glenbawn and Traralgon Railway Reservoir exhibit the opposite trend to metal emission trends in other developed countries worldwide. In these countries, atmospheric metal emissions are decreasing, likely due to the shift from coal combustion to green technologies.

The current steady increase in metal emissions from coal-fired power stations in Australia provides grounds for a review of current regulatory approaches. This review indicates that current regulations for Se, As and other metals differ across jurisdictions, with different levels of emissions reductions, and may in some cases provide only partial coverage, particularly in Victoria. In addition the review indicates that there is room for future regulations to encourage the adoption of best available technologies to further limit Se, As and other metals emissions. Finally, further levels of independent checks on metal emissions data from coal-fired power stations would enhance the reliability of data reported to the NPI.

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Figure and Table Captions:

Figure 1 – Map of Australia showing the locations of Traralgon Railway Reservoir (Latrobe Valley) in Victoria (VIC) and Lake Glenbawn (Hunter Valley) in New South Wales (NSW).

Sediment sampling sites in Traralgon Railway Reservoir (bottom left) and Lake Glenbawn (bottom right). The location of the open-cup coal mines and power stations in relation to the lakes are indicated by red arrows.

Figure 2 – Hunter Valley, NSW: location of open-cut mines and power stations. On the right are wind roses showing the frequency of occurrence of wind speed and direction for 9am and 3pm annually, Scone Airport, 14 km west of Lake Glenbawn. Latrobe Valley, VIC: Location of Yallourn, Loy Yan and Hazelwood power stations and respective coal mines in relation to Traralgon Railway Reservoir in the Latrobe Valley (VIC). On the right are wind roses showing the frequency of occurrence of wind speed and direction for 9am and 3pm annually, Latrobe Valley airport, 5 km west of Traralgon Railway Reservoir. For both Hunter and Latrobe Valley images, the percentage of calm conditions is represented by the size of the centre circle - the bigger the circle, the higher is the frequency of calm conditions. Each branch of the rose represents wind coming from that direction, with north to the top of the diagram. The branches are divided into segments of different thickness and colour, which represent wind speed ranges from that direction. Speed ranges of 10km/h are used in these wind roses. The length of each segment within a branch is proportional to the frequency of winds blowing within the corresponding range of speeds from that direction. From: Australian Bureau of Meteorology (BOM, 2019b).

Figure 3 – Lake Glenbawn core data: A) sediment metal concentration (mg/kg), spheroidal carbonaceous particles (SCP) concentration (g/DM) and Specific Surface Area (SSA) (m^2/g); B) sediment metal flux ($ug/yr/m^2$), spheroidal carbonaceous particles (SCP) flux ($n^o/cm^2/yr$), precipitation rate (average annual rainfall for 3 years divided by past 100 years average) and black coal consumption in New South Wales (kt) (source: Australian Energy Statistics, Table P DEE, 2018). Note that prior to 1957 most sediment layers are part of the Hunter river bed sediment and chronology was not considered from this depth down the core.

Figure 4 - Principal Component Analyses for metal concentrations in Lake Glenbawn sediment cores. Dates in blue refers to the time before coal-fired power stations were commissioned, while dates in orange refer to the time after coal-fired power stations were commissioned. Numbers next to dots are the year for a given sediment layer.

Figure 5 – Traralgon Railway Reservoir sediment core data: A) sediment metal concentration (mg/kg), spheroidal carbonaceous particles (SCP) concentration (g/DM) and Specific Surface Area (SSA) (m^2/g); B) sediment metal flux ($ug/yr/m^2$), spheroidal carbonaceous particles (SCP) flux ($n^o/cm^2/yr$), precipitation rate (average annual rainfall for 3 years divided by past 100 years average), and black coal consumption in New South Wales (kt) and brown coal consumption in Australia (kt) (source: Australian Energy Statistics, Table P DEE, 2018).

Figure 6 - Principal Component Analyses for metal concentrations in Traralgon Railway Reservoir. Dates in blue refers to the time before coal-fired power stations were commissioned, while dates in orange refer to the time after coal-fired power stations were commissioned.

Figure 7 – Enrichment factor of metals plotted against years in sediments of Lake Glenbawn, NSW.

Figure 8 – Enrichment factor of metals plotted against years in sediments of Traralgon Railway Reservoir, NSW.

Figure 9 - Summary of key federal and state regulations relevant to atmospheric metal emissions from coal-fired power stations.

Table 1 – Commissioning date, electrical capacity, coal type used and pollution control devices in coal-fired power stations of the Latrobe and Hunter Valley.

Power Station	**Location**	**Commissioned**	**Electrical capacity (MW)**	**Coal used**	**Pollution Control Device**
Bayswater	Hunter Valley	1985	2640	bituminous	Bag filter (NPI, 2019a).
Liddell	Hunter Valley	1971	2000	bituminous	Bag filter (retrofitted in 1990) (NPI, 2019b).
Loy Yang A	Latrobe Valley	1984	2210	lignite	Electrostatic precipitator (NPI, 2019c).
Loy Yang B	Latrobe Valley	1993	1,070	lignite	Electrostatic precipitator. (NPI, 2019d).
Yallourn	Latrobe Valley	1975	1,480	lignite	Electrostatic precipitator. (NPI, 2019e).

Table 2 – List of main open-cut coal mines near Lake Glenbawn and Traralgon Railway Reservoir and coal annual production (tonnes).

		Co	**Cu**	**Zn**	**As**	**Se**	**Pb**
		(mg/kg)					
Glenbawn	**Background**	18	28	84	4.3	BCL	6.9
	2018	30	59	137	14	1.6	13
	Fold-increase	1.6	2.1	1.6	3.3	NA	1.9
Traralgon	**Background**	3.7	2	9.7	0.15	BCL	3.5
	2018	18	23	133	5.6	1.1	22
	Fold-increase	4.9	11	14	37	NA	6.3

BCL = Below Confident Limit of 0.01 mg/kg dry mass.

NA = not applicable, not measure or not available.

Table 3 – Background and modern (2018) metal concentration in Lake Glenbawn and in Traralgon Railway Reservoir, with fold-increase in concentrations per metal.

	Cu	**Zn**	**As** **(mg/ kg)**	**Se**	**Pb**
ANZG default guideline values	65	200	20	2	50
Lake Glenbawn, Hunter Valley	59	137	14	1.6	13
Traralgon Railway Reservoir, Latrobe Valley	24	133	6	1.3	25

Table 4 – Metal concentration default guideline value threshold limit from the ANZG (2018)) sediment quality guideline, and maximum concentration of sediments in Lake Glenbawn and Traralgon Railway Reservoir.

Study site	**Metal (mg/kg)**	**ANZG guidelines**	**2008**	**2018**	**Fold increase past 10 years**	**Nº years to reach guideline threshold concentration***
Glenbawn	**Cu**	65	56	59	1.1	10
	Zn	200	125	137	1.1	13
	As	20	11.5	14.2	1.2	11
	Se	2	1.2	1.6	1.3	10
	Pb	50	11.1	12.8	1.2	33
Traralgon	**Cu**	65	10.49	22.66	2.2	13
	Zn	200	60.64	132.85	2.2	7
	As	20	1.73	5.64	3.3	11
	Se	2	0.55	1.12	2.0	9
	Pb	50	17.25	22.13	1.3	18

* calculations assumed the exactly same coal burning rates and climate variables as per the last 10 years.

Author contribution statement

L Schneider and S. Haberle conceived the original idea for this project. L. Schneider, A. Lintern and S. Haberle planned and performed field work in the Latrobe Valley. L. Schneider, W. Maher, J. Potts, D. Sinclair and C. Holley performed field work in the Hunter Valley. L Schneider and W. Maher performed ICPMS analyses and interpretation of the data. N. Rose and L. Schneider performed analyses of spheroidal carbonaceous particles. W. Maher and N. Rose verified the analytical methods used in this study. A Zawadizki and A. Lintern performed ^{210}Pb dating analyses, and together with M. A. Lopez have interpreted the dating data. M.A. Lopez developed the age-depth model. D Sinclair and C. Holley researched the pertinent regulatory framework on metal emissions in Australia and discussed the regulatory section of this manuscript. All authors discussed the results and contributed to the final manuscript.

Conflict of Interest

No conflict of interest is declared for this study and final manuscript.

Graphical abstract

Highlights

Metal contamination was assessed in two major coal production centres in Australia

Metal concentrations increased with commissioning of coal mining and power generation

SCP appeared in sediments at the time coal fired power stations were commissioned

Metal emissions are still increasing in Australia, following the trend in Asian countries

Our results provide grounds for a review of current regulatory approaches in Australia