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How significant is atmospheric metal contamination from mining activity adjacent to the Tasmanian Wilderness World Heritage Area? A spatial analysis of metal concentrations using air trajectories models

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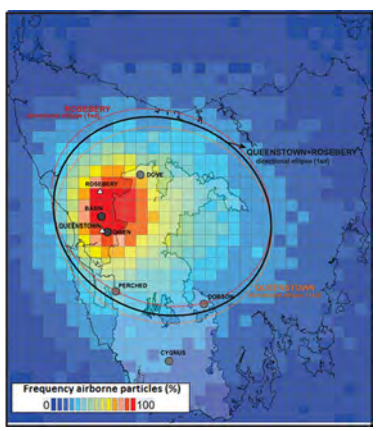
HIGHLIGHTS

- Airborne metal contamination from historical mining was investigated in Tasmania.
- The largest contamination occurred ca. 1930 when open-cut mining commenced.
- The HYSPLIT model explains the atmospheric distribution of metal in the environment.
- The metal contamination hotspot is in the northwest region of the World Heritage Area.
- Cu and Pb enrichment factors are among the highest reported worldwide.

GRAPHICAL ABSTRACT



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ABSTRACT

This study investigated metal contamination from historical mining in lakes in the Tasmanian Wilderness World Heritage Area (TWWHA) and surrounding region. The largest increase in sedimentation and metal contamination occurred ca. 1930 when open-cut mining commenced and new mining technology was introduced into the region. The geochemical signal of lake sediments changed from reflecting the underlying geology and lithology to that reflecting mining activities. The HYSPLIT air particle trajectory model explains metal distribution in the lakes, with those in the northwest region closest to the mines having the highest metal contamination. Lake metal concentrations since mining activities commenced are in the order: Owen Tarn > Basin Lake > Perched Lake > Lake Dove > Lake Dobson > Lake Cygnus, with Perched Lake and Lakes Dove, Dobson and Cygnus in the TWWHA. Metal contamination affected centres up to 130 km down-wind of mining sites. Enrichment factors (EF) for Pb, Cu, As and Cd are >1 for all lakes, with Owen Tarn and Basin Lake having very high EFs for Cu and Pb (98 and 91, respectively). Pb, Cu, As and Cd concentrations are above the Australia/New Zealand lower

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Enrichment factor
Extremely severe enrichment

sediment guidelines, with Pb, Cu and As above the high guidelines in Owen Tarn and Basin Lake. This study demonstrated the legacy of metal contamination in the TWWHA by mining activities and the consequences of a lack of execution of environmental regulations by past governments in Tasmania.

1. Introduction

Mining has been a key factor in the economic development of Tasmania, Australia, with numerous abandoned mine sites that are still contaminating soils, rivers, lakes and estuaries (Fig. 1; Augustinus et al., 2010). This is of environmental concern in a wide geographical area beyond the immediate vicinity of mine sites, as particulate emissions released to the atmosphere by mining operations can be transported over long distances by atmospheric circulation (Suvarapu and Baek, 2017). Here, we use computer modelling of air particle trajectories and lake sediment contamination measurements to develop and test a model of airborne contamination transport from historic mining activities in western Tasmania.

The west coast of Tasmania is characterised by folded and faulted geology containing several ore bodies that were exploited when the British arrived in Australia in the late 1700s. Principal among these are the major mining centres developed around the closely spaced Mt Lyell and Mt Read ore deposits, in Queenstown and Rosebery, respectively.

Analyses of sediment and water from Macquarie Harbour, downstream from the Queenstown region, indicates a dramatic increase in metal and metalloid concentrations (hereafter collectively referred as metals) in the harbour resulting from contamination of the Queen and King Rivers by the Mt Lyell mine (Augustinus et al., 2010; Carpenter et al., 1991; Eriksen et al., 2001; Stauber et al., 2000; Teasdale et al., 2003). Further afield, an increase in metals in isolated catchments downwind from both Queenstown and Rosebery reveal the same trends in metal contaminants through the period of intensive mining and smelting operations, suggesting transportation of metal contaminants by wind from mining centres (Harle et al., 2002).

While the effects of mining on the environment around the Queenstown-Rosebery region are relatively well recognised, e.g.

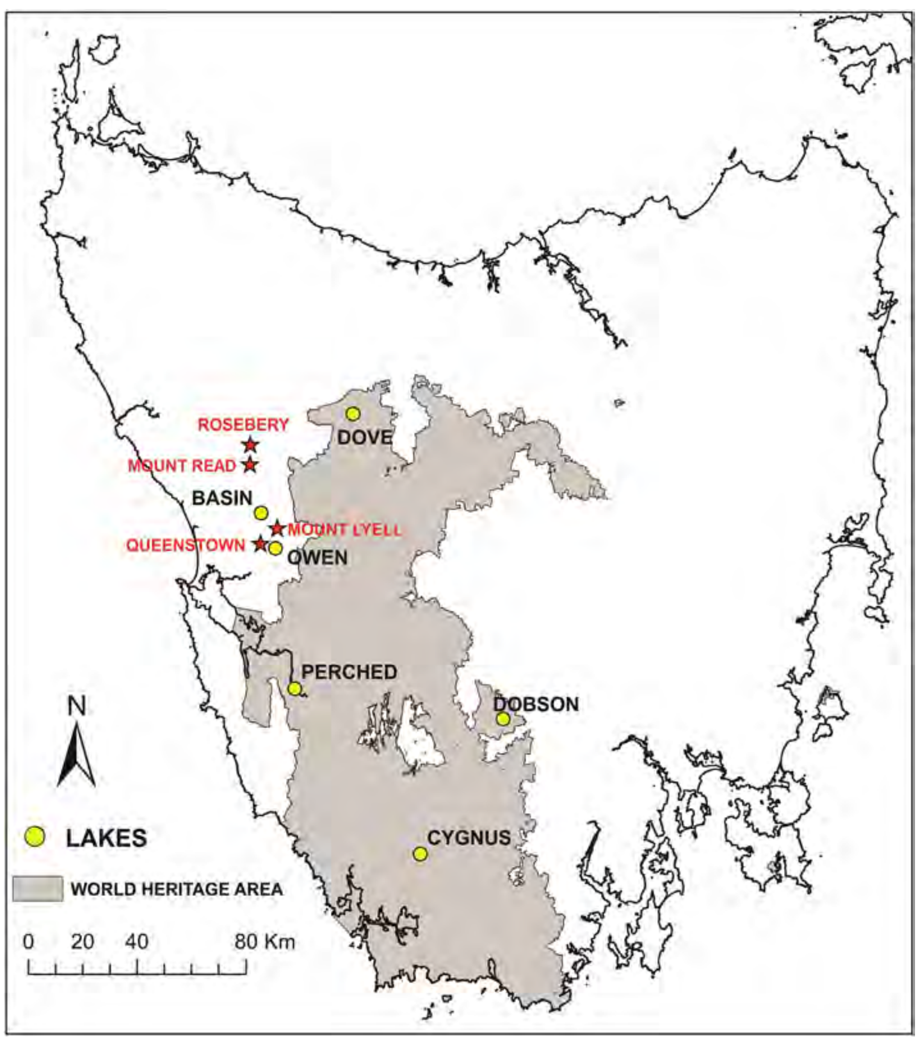

Fig. 1. Map of Tasmania, Australia, with the Tasmanian Wilderness World Heritage Area in grey. The yellow circles indicate the six lakes in this study. The red stars indicate the three mining centres in the area: Queenstown, Mount Lyell and Mount Read, Rosebery.

localised deforestation and downstream impacts on aquatic ecosystems (De Blas, 1994; Harle et al., 2002; Hodgson et al., 2000; Kozlov and Zvereva, 2006), there has been no attempt to understand the spatial distribution of airborne metal contaminants from Queenstown and Rosebery. This is important because the western boundary of the Tasmanian Wilderness World Heritage Area (TWWHA) lies just 11 and 12 km from both Queenstown and Rosebery, respectively, in the prevailing wind direction.

The atmospheric distribution of metal contaminants involves a complex interplay of environmental factors, climate and local meteorological characteristics. The principal environmental factors affecting atmospheric transport of metals include, but are not limited to, precipitation, temperature, air movement and pressure (Fang et al., 2005; Pacyna et al., 2009; Suvarapu and Baek, 2017). All of these must be taken into consideration when assessing atmospheric metal distribution and deposition into the environment.

A useful tool to understand the interplay of climate factors on metal distribution is the Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT) model (NOAA, 2018). HYSPLIT produces forward trajectories that, when combined with satellite images (from NASA's MODIS satellites), can calculate air particle trajectories over a set period and, thus, the direction atmospheric contamination has travelled (Kusumaningtyas and Aldrian, 2016). Despite its apparent usefulness and value, this model has never been applied to assess airborne contamination from historical mining sites in Tasmania to understand the potential effects of airborne contamination on the environment.

In this study, we assess the extent of metal contamination in the TWWHA and surrounding areas using sediment cores from six freshwater lakes. In particular, we applied the HYSPLIT model and statistical analyses to establish the main chemical and physical factors affecting the airborne distribution of metals in these lakes. Furthermore, we compared lake sediment metal concentrations with the Australia/New Zealand (ANZECC/ARMCANZ, 2000) sediment guidelines to assess the past and current health of the local environments. Ultimately, the study was undertaken to inform the scientific community and the public about the legacy of metal contamination within the TWWHA to support government initiatives in establishing appropriate regulations and policies to protect the environmental values of this wilderness area.

2. Material and methods

2.1. Regional description

Western Tasmania is a mountainous area underlain predominantly by intensely folded and faulted Cambrian and pre-Cambrian quartzite rocks and conglomerate units that are intersected with highly mineralised volcanic belts (Corbett and Solomon, 1989). The area includes >4000 lakes and tarns, mostly of glacial origin, ranging from highly acidic, dystrophic lakes to ultra-oligotrophic clear water lakes (Hodgson et al., 2000). The west coast receives high orographic rainfall produced by air masses rising over mountains (Gentilli, 1972; Sturman and Tapper, 2006). The rainfall reaches a maximum of 3400 mm and there is an annual temperature range of 3–21 °C, with a mean annual temperature of 11 °C at sea level, and 6 °C at 1000 m altitude (Langford, 1965). The climate is dominated by the prevailing zonal westerlies that latitudinally migrate through the seasonal cycle, with west to south-westerly airflow dominant in the austral winter and west to north westerly airflow dominant in the austral summer.

2.2. Historical setting

Tasmania has been occupied by humans for ca. 40,000 years (Cosgrove, 1999), with Tasmanian Aborigines responsible for maintaining an essentially open landscape through the use of fire (Fletcher and Thomas, 2010). Despite the arrival of the British in Tasmania in the late 1700s, it was not until the late 19th century with the arrival of mineral prospectors that western Tasmania was exposed to exploitation for deposits of gold, silver, lead, zinc and copper. Subsequently, major mining and smelting operations were established and concentrated at several centres around Queenstown and Rosebery (Fig. 1).

Discharge of tailings, slag, toxic metals and acid drainage into the Queen River that runs through Queenstown, and downstream to the King River and ultimately into Macquarie Harbour, has eliminated all but the most robust forms of aquatic life in these waterways (Hodgson et al., 2000). Today, these towns are located along the boundary of the TWWHA, thus, there is a high probability that areas within the TWWHA have experienced some degree of long-range metal contamination and ecological change from historic mining activities (Harle et al., 2002; Hodgson et al., 2000).

Mineral exploration commenced in the 1880s, however, it was not until the end of 1920s, with the advent of automation and changed work practices, that mining activities expanded from underground to open-cut. This was attributed to favourable copper prices and advances in transport (Rae, 1994).

The mining boom in Queenstown-Rosebery saw a downturn in the 1980s due to the low price of copper and activities were reduced to two mining companies: Copper Mines of Tasmania (CMT) in Queenstown which has now been active for 100 years, and MMG Rosebery, active in Rosebery since 1936.

2.3. Site selection and sediment core collection

Sites were chosen to provide an adequate spatial coverage to characterize the aerial transport of metals from mining sites and to document spatial differences in the deposition of metals within the TWWHA. Given the aim of assessing atmospheric transport of particles and metal deposition in lakes, we targeted lakes with small catchments to avoid major geochemical influence from the drainage basins. A total of six lakes were identified as suitable for these analyses, and a total of six cores (one per lake) were collected from the deepest point of the lakes. A 25 m-resolution digital elevation model (DEM) was used to analyse lake catchment morphologies from where the catchment area for each lake was derived using the suite of Hydrology Tools (Arc Hydro) in ArcGIS 10.3 (ESRI, 2015). This approach allowed us to map flow direction and stream paths based on aspect and slope for each cell of the DEM. The catchment boundaries were delineated, and the surface was calculated using the same program.

Sediment collection was conducted in two periods:

Collection 1: Sediment cores from lakes in the TWWHA (Dove Lake, Lake Cygnus, Lake Dobson and Perched Lake) were collected in 2000 using a gravity corer and hammer driven piston corer (Neale and Walker, 1996).

Collection 2: Sediment cores from lakes bordering the TWWHA (Lake Basin and Owen Tarn) were collected in 2011 and 2015, respectively, using a Universal Corer.

2.4. Sediment dating

Lead-210 (^{210}Pb) samples were processed at the Australian Nuclear Science and Technology Organisation (ANSTO) using alpha spectrometry and following methods described by Harrison et al. (2003). Each dried sediment sample (2 g) was spiked with Polonium-209 (^{209}Po) and Barium-133 (^{133}Ba) tracers. Each sample was then leached with hot nitric and hydrochloric acids to release polonium and radium. Polonium was autoplated onto silver disks after adding the reducing agent hydroxylammonium chloride. Radium and barium were isolated by co-precipitation and collected as colloidal micro-precipitates of barium sulphate on fine membrane filter papers. The activities of ^{210}Po on the silver disks and ^{226}Ra on the membrane filters were determined by alpha spectrometry. Each membrane filter was also counted by gamma spectrometry to measure the ^{133}Ba tracer activity. Chemical yield recoveries of ^{210}Po and ^{226}Ra were calculated using the recoveries of ^{209}Po and ^{133}Ba tracers, respectively. Unsupported ^{210}Pb activity for each sample was calculated from the activity of ^{210}Po (the proxy for total ^{210}Pb) minus the ^{226}Ra activity (the proxy for supported ^{210}Pb).

The ^{210}Pb dating models Constant Initial Concentration (CIC) (Pennington et al., 1976; Robbins and Edgington, 1975) and Constant Rate of Supply (CRS) (Appleby and Oldfield, 1978) were used to determine sediment ages and mass accumulation rates for sediment cores with dry bulk density data available. A modified CIC ^{210}Pb dating model as described by Brugam (1978) was used to determine CIC ages and sedimentation rates for those sediment cores where dry bulk density data were not available.

2.5. Geochemical analyses

All samples were transported from the field to the laboratory at ANSTO or at the University of Canberra and stored at 4 °C in a cool room. Samples were manually mixed and transferred to a clean glass vial, covered with parafilm and placed in a FreeZone Plus 6 freeze-drier (Labconco, Kansas City, MO) and lyophilized at −50 °C for 48 h.

Given the substantial gap between the analysis of samples from collection 1 and 2 (ca. 15 years), the methodologies differ slightly due to the technology and procedures in use at the time. Similar results for certified reference materials indicate the applicability and comparability of both procedures.

Samples from collection 1 (Lake Dove, Perched Lake, Lake Dobson and Lake Cygnus): Approximately 0.5 g of dried sediment was weighed into a tetrafluormethaxil closed digestion vessel (Ethos Milestone) and 3 mL of sub-boiled nitric acid, 1 mL of sub-boiled hydrochloric acid, 0.1 mL of 50% w/v high purity hydrofluoric acid (Merck, Suprapur) and 3 mL of deonised water added. Each vessel was capped and placed in a Milestone MLS 1200 Mega microwave cavity, heated to 180 °C for 25 min, and then held at 180 °C for 15 min before being cooled to room temperature and diluted with 30 mL of deonised water. One milliliter of the digest was transferred to an 8 mL centrifuge tube and 4 mL of ICP-MS internal standard added (^{6}Li, ^{45}Sc, ^{89}Y, ^{103}Rh, ^{115}In, ^{185}Re, and ^{209}Bi).

Metal concentrations in sediments were measured by an inductively coupled plasma mass spectrometer (ICP-MS) and inductively coupled plasma atomic emission spectrometer (ICP-AES). Mixed standard working solutions in the 500 to 0.001 μg/mL range and continuous calibration verification solutions were measured at the same time as samples. Internal standard and suppression solutions (In, Rh, Rb) were prepared and added to the sample via on-line addition. Certified reference materials, National Research Council of Canada (NRCC) sediment SRMS MESS-3 and PACS-1 were also analysed and measured values were in agreement with certified values (Supplementary Table 1).

Sediment samples from collection 2 (Owen Tarn and Basin Lake): approximately 1 g of sediment was weighed into a 60 mL polytetrafluoroacetate (PFA) closed digestion vessel (Mars Express), and 2 mL of concentrated nitric acid (Aristar, BDH, Australia) and 1 mL of 30% concentrated hydrochloric acid (Merck Suprapur, Germany) added (Telford et al., 2008). Each PFA vessel was then capped, placed into an 800 W microwave oven (CEM model MDS-81, Indian Trail, NC, USA), and samples heated at 120 °C for 15 min. The digests were cooled to room temperature and diluted to 50 mL with deionised water (Sartorius). The tubes were then centrifuged at 5000 rpm for 10 min. One milliliter of the digest was transferred into a 10-mL centrifuge tube and then diluted to 10 mL with ICP-MS internal standard (Li^{6}, Y^{19}, Se^{45}, Rh^{103}, In^{116}, Tb^{159} and Ho^{165}). Digests were stored (0–5 °C) until analysis. Samples were analysed using an ICP-MS (PerkinElmer DRC-e) with an AS-90 autosampler (Maher et al., 2001). The certified reference NIST-2710 Montana Soil was used as a control to check the quality and traceability of metals. Measured concentrations were in agreement with certified values (Supplementary Table 1).

2.6. The Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT)

Wind trajectories from the mining sites were calculated using a map of the frequencies of HYSPLIT trajectories (Stein et al., 2015). A map with the average circulation of air masses over Tasmania during the period 1961–1990 for particles released at 42°S and 145.5°E (Queenstown, Tasmania) and 41.78°S and 145.5°E (Rosebery, Tasmania) was created using ~1 million data points corresponding to the position of hourly-resolved HYSPLIT forward-trajectories, overlayed with 10 × 10 km grid cells. The 1961–1990 period represents the full extent of the data available from the Australian Bureau of Meteorology.

Hourly-resolved meteorological data for calculating the HYSPLIT trajectories were derived from NOAA ARL NCEP/NCAR Reanalysis FTP (ftp://arlftp.arlhq.noaa.gov/pub/archives/reanalysis). The number of occurrences per grid cell was extracted in ArcMap 10.3 and relative frequencies calculated. Red indicates grid cells with a higher occurrence of air masses travelling from Queenstown/Rosebery. A directional ellipse was derived using the 'Directional Distribution: Standard Deviational Ellipse' function in ArcMap 10.3. This tool creates an elliptical polygon centred on the mean for all features. The orientation of the ellipse indicates the average direction of flow during the chosen time window and spatial scale. One standard deviation was chosen to cover approximately 68% of all input feature centroids.

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 sediments (Abrahim and Parker, 2007). 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, which may have substantial variations in the sediment (i.e. clay rich to sand ratios).

The EF method normalises the measured metal concentration with respect to a sample reference element such as iron (Fe) or aluminium (Al) (Cevik et al., 2009). In this approach the Fe or Al is considered to act as a "proxy" for the clay content. In this study, as Fe atmospheric deposition in lakes are known to have been altered by mining activities, we used Al as it was the element with least change through the profiles.

The EF was calculated using the average contamination for the years comprising the peak in mining contamination (1930 to 1980), following the equation:

$$EF = (Mx/Alx)/(Mb/Alb)$$

where *Mx* and *Alx* are the average metal and aluminium concentrations, respectively, for the mining period between 1930 and 1980. *Mb* and *Alb* are metal and aluminium background concentrations, respectively.

The lower metal concentrations in the bottom of the cores were interpreted as sediment deposited before the beginning of mining activities in 1880. From these results, natural background heavy metal values for the six lakes was proposed based on the average of pre-mining trace element concentrations.

2.8. Statistical analyses

All analyses were performed using the R Statistical Software (R Development Core Team, 2008) and the respective libraries used in particular analyses are cited.

To reveal differences in the metal concentrations among lakes and mining phases, we conducted a permutational multivariate analysis of variance (PERMANOVA) based on Euclidean distances (adonis, vegan package 2.5-1 https://cran.r-project.org/web/packages/vegan/index.html) using the function vegdist to find the dissimilarities. Lakes and phases were included as fixed factors, and metal concentrations were given as a matrix from where vegan calculated pairwise distance to find the dissimilarities.

Principal Component Analysis (PCA) was used to explore the similarity of metal concentration in the lakes before and after mining activities in the region (dudi, ade4 package https://cran.r-project.org/web/packages/ade4/index.html). A multiple regression with backward-stepwise selection was performed to identify the main drivers of metal deposition in the sediments. Metal concentrations were log transformed to comply with the assumptions of linearity, normality and homoscedasticity. Before running the multiple regression, predictor correlations were checked to avoid problems for parameter estimation and potentially leading to the wrong identification of relevant predictors of the statistical model (Dorman et al., 2013). The predictors tested were: catchment size, precipitation, atmospheric temperature, distance from the mining site and frequency of particles passing over the lakes, calculated from wind directions and speed in HYSPLIT trajectories (Section 2.5). If a correlation was >0.7, then one of the predictors was removed.

3. Results and discussion

3.1. Sediment dating

^{210}Pb dating results are shown in Supplementary Table 2 and Supplementary Fig. 1. The CIC and CRS ^{210}Pb dating model results were in close agreement in most lakes except for Lake Cygnus and Owen Tarn. The unsupported ^{210}Pb activities from these cores exhibited non-monotonic profiles, thus the use of the CRS model was more appropriate (Supplementary Table 2).

The largest variations in sedimentation rates for sediment cores closer to the mines were recorded in the 1930s (Supplementary Table 3). At this time, the open-cut mine commenced in the region and new technology such as stamper and mills arrived in the region. This change in mining methods and technology increased sedimentation rates in these lakes as a result of increased atmospheric inputs. Details are discussed in a separate publication.

3.2. Background metal concentrations and spatial distribution

Patterns of metal deposition in sediments have changed dramatically since the start of mining activities (Table 1 and Supplementary Table 3). Sediment metal concentrations differed significantly between lakes (PERMANOVA: F model $= 104.1$; $r^2 = 0.729$; $P < 0.001$; 999 permutations) and between mining periods (PERMANOVA: F model $= 9.5$; $r^2 = 0.153$; $P < 0.001$; 999 permutations). PCA of metal concentrations (Fig. 2A–B) illustrates the changes in metal concentrations (Axis 1) and their dramatic change since mining activities commenced.

Pre-mining metal concentrations were in the order of: Lake Dove > Perched Lake > Basin Lake > Lake Dobson > Owen Tarn > Lake Cygnus. Metal concentrations since mining activities commenced are in the order of: Owen Tarn > Basin Lake > Perched Lake > Lake Dove > Lake Dobson > Lake Cygnus (Table 1, Supplementary Table 2). These results demonstrate that mining activities have caused a shift in the geochemical signals of sediments in the lakes, from signals reflecting the specific geology and lithology to an association with mining activities.

The PCA performed using metal concentrations measured in sediments dated from before and after the 1880s clearly indicates that the most proximal sites to the mining centres (Owen Tarn and Basin Lake) have the highest metal concentrations since mining. Lake Dove and Perched Lakes, with the highest background concentrations, decreased in the rank of metal concentrations since mining activities commenced (Fig. 2A–B).

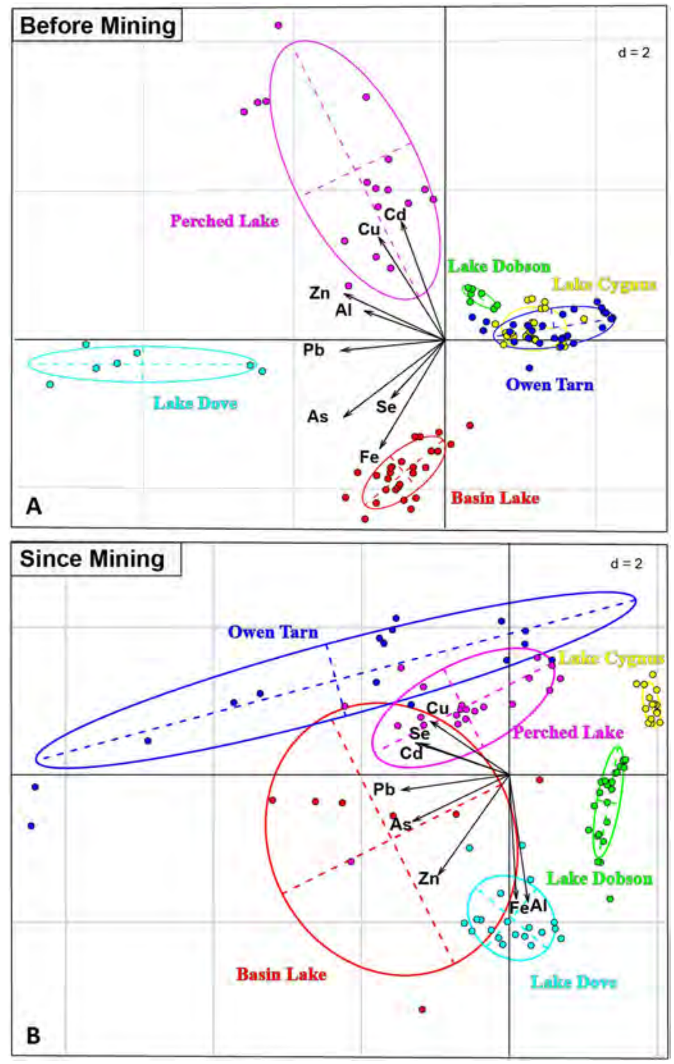

Fig. 2. Principal Component Analyses of metal concentrations (Cu, Se, Cd, Pb, As, Zn, Fe and Al) in sediments of the four lakes in the Tasmania Wilderness World Heritage Area (Perched Lake and Lakes Dove, Dobson and Cygnus) and closer to the mining centres (Basin Lake and Owen Tarn). A) before mining and B) since mining activities commenced.

The majority of effort in determining the impact of mining contamination on aquatic environments in western Tasmania has focussed on waterborne contamination down-stream from mines (Augustinus et al., 2010; Carpenter et al., 1991; Dawson, 1996; De Blas, 1994; Eriksen et al., 2001; McQuade et al., 1995; Stauber et al., 2000; Teasdale et al., 2003), with airborne contamination receiving comparatively little attention (Harle et al., 2002). Our study reveals that metal contamination can influence sites up to 130 km down-wind of mining sites, with Lake Cygnus in the TWWHA displaying clear evidence of contamination. These results indicate that most of the TWWHA area has potentially been affected by airborne contamination from the Queenstown-Rosebery mines (Fig. 3). We thus urge a concerted effort to understand the environmental and ecological consequences of this contamination in the TWWHA.

3.3. Drivers of metal spatial distribution

Predictors of metal atmospheric distribution in the TWWHA are given in Table 2 and Figs. 3 and 4. Table 2 also summarises the main geographical and climatological information for each lake that were considered to be the main factors influencing metal distribution in lakes across the TWWHA.

Table 1
Metal concentrations in lake sediments within the Tasmanian Wilderness World Heritage Area (Lakes, Dove, Cygnus, Dobson and Perched) and near the mining centres (Owen Tarn and Basin Lake).

Lake	Mining	Al	Fe	Pb	Cu	Zn	As	Se	Cd
ID	Phases	Concentration (mg/kg)							
Owen Tarn	Before mine (1880)	1639	769	10	5	3	3	1.7	0.2
Owen Tarn	Mining peak (1930 to 1980)	3336	1311	492	220	51	50	7.6	1.4
Owen Tarn	Fold-increase	1.0	0.7	46	43	19	14	3.6	6.4
Basin	Before mine (1880)	4185	19,077	7	4	6	6	4.5	0.0
Basin	Mining peak (1930 to 1980)	8680	26,838	388	203	44	63	7.4	0.6
Basin	Fold-increase	1.1	0.4	58	44	6	10	0.7	55.8
Dove	Before mine (1880)	39,337	15,366	74	9	50	9	3.3	0.3
Dove	Mining peak (1930 to 1980)	55,179	16,986	248	47	78	33	3.4	0.6
Dove	Fold-increase	0.4	0.1	2.4	4.3	0.6	2.8	0.0	1.0
Cygnus	Before mine (1880)	12,292	2741	2	2	3	0.6	0.3	0.1
Cygnus	Mining peak (1930 to 1980)	14,662	4382	5	12	6	0.8	2.3	0.1
Cygnus	Fold-increase	0.2	0.6	1.1	6.4	1.1	0.3	7.3	0.0
Dobson	Before mine (1880)	15,783	10,567	1	9	19	0.5	BDL	BDL
Dobson	Mining peak (1930 to 1980)	16,214	12,929	13	9	39	0.8	BDL	0.4
Dobson	Fold-increase	1.0	1.2	11	1.1	2.1	1.6	N/A	N/A
Perched	Before mine (1880)	10,362	3505	8	18	27	2.5	4.3	1.3
Perched	Mining peak (1930 to 1980)	18,360	6760	118	71	37	6.4	8.5	2.3
Perched	Fold-increase	0.8	0.9	14	2.9	0.3	1.5	1.0	0.7

BDL = below detection limit.

Predictors (Table 2) were checked for between-predictors correlations to select the predictors to run the statistical model. The factors distance, precipitation and frequency had a correlation >0.7 and were, therefore, removed from the model and the HYSPLIT-derived frequency of particles was used. This decision was based on knowledge that the HYSPLIT frequency model takes into consideration environmental variables and distance in its calculation. The final list of predictors for the model was therefore: catchment size, temperature and frequency of the particles.

The HYSPLIT frequency of particles model (Fig. 5) successfully explained most of the metal atmospheric transport and metal deposition into the lakes (Table 3). The significance of the HYSPLIT model on metal distribution indicates that this model provides an effective predictive tool for the spread of airborne pollutants in the landscape. The decline in metal concentration over distance is indicative of atmospheric dispersion of the particles, resulting from mining activity.

The high precipitation rate within the TWWHA area suggests that wet deposition is an important factor in metal deposition into the environment. Although lakes with small catchment areas were only considered in this study, catchment size was a significant factor only for the major elements Fe, Al and Zn. This indicates that metals deposited in these lakes were mainly a result of atmospheric metal deposition rather than catchment leaching (Table 3).

3.4. Enrichment factors

To evaluate the extent of the historical metal contamination affecting the TWWHA lake sediments, EFs were calculated for the period 1930–1980, where 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). All lakes had at least one of the metals with sediment concentrations showing moderate enrichment (EF > 3) since mining commenced (Table 4).

The EF values demonstrate significant metal contamination in the TWWHA. Mining contamination has reached distances as far as 130 km as demonstrated by the EF values > 1 for Lake Cygnus, the furthest lake from Queenstown and Rosebery in this study (Table 4, Fig. 3). The effect of metal contamination distribution in the entire TWWHA and surrounding area can be visualised in Fig. 6, which demonstrates the significant increase of metal inputs since mining started.

The EF values demonstrate that, from the metals measured in this study, As, Cd, Cu, Pb and Zn are the elements of most concern in the region. Owen Tarn and Basin Lake had the most significant metal enrichment in sediments. In Owen Tarn, specifically, Cu and Pb were 90 times higher relative to the background values (Table 4). This is of major concern as Pb and Cd bioaccumulate in the bodies of aquatic and soil

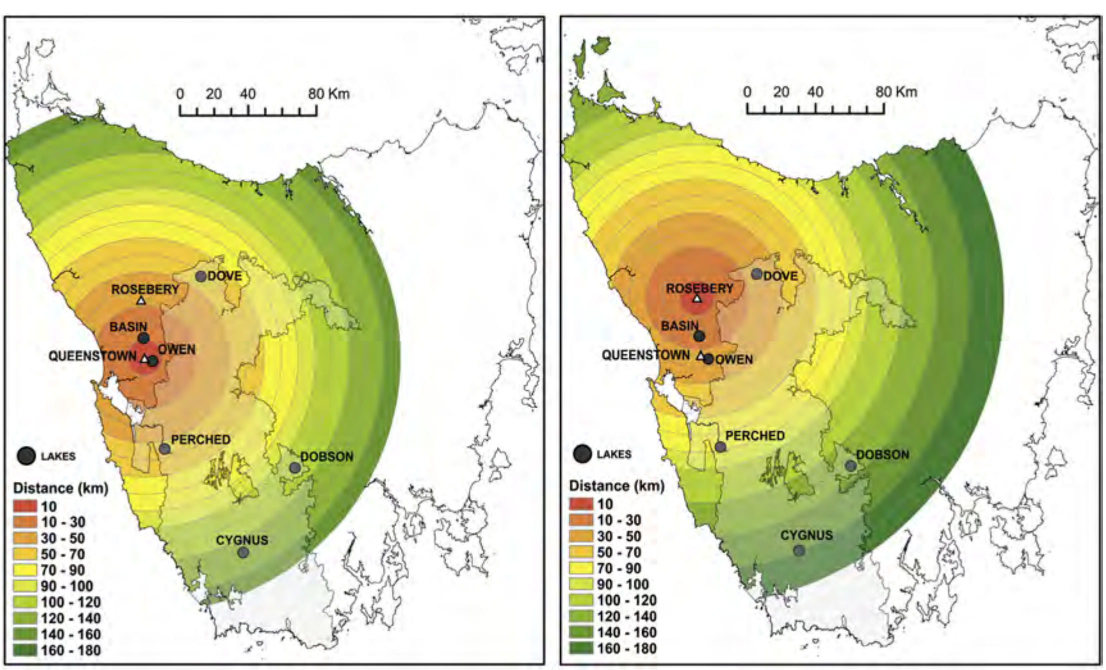

Fig. 3. Distance of studied lakes from: (A) Queenstown mining site and (B) Rosebery mining site. Tasmanian Wilderness World Heritage Area in grey. Maps developed in ArcMap 10.3.

Table 2
Tasmanian Wilderness World Heritage Area and surrounding lakes and their attributes: catchment size (km^2), geographic coordinates, annual precipitation (mm), annual temperature (°C), and distance from the mining sites in both Queenstown and Rosebery.

Lake name	Core depth analysed (cm)	Catchment size (km^2)	Longitude	Latitude	Total annual precipitation (mm)	Mean annual temperature (°C)	Distance from (km)	
							Queenstown	Rosebery
Owen Tarn	48	0.2	145.609434	−42.0997	2816	8.8	5	36
Basin Lake	37	0.9	145.54829	−41.9808	3128	9.6	11	22
Dove Lake	37	5.3	145.962222	−41.6575	2706	9	58	38
Perched Lake	10	0.2	145.686163	−42.5648	2150	10.6	55	88
Lake Dobson	10	1	146.617778	−42.6719	1443	8.1	109	133
Lake Cygnus	10	0.3	146.241944	−43.1183	1974	8.9	128	160

organisms (Cresswell et al., 2015; Lanctôt et al., 2017; Storelli, 2008; Zheng et al., 2007). Even small concentrations of these metals can affect body functions of aquatic organisms (Hodgson et al., 2000).

The Pb and Cu EFs of 91 and 98, respectively, recorded in Owen Tarn are among the highest reported in the scientific literature. These results are comparable to highly contaminated places such as in the Kurang River in Pakistan, subjected to heavy metal contamination from urbanisation and discharge of untreated domestic effluents (EF Pb = 4.46, EF Cu = 12) (Zahra et al., 2014), the Shur River in Iran receiving inputs from copper mining (EF Pb = 118.42, Cu = 264.1) (Karbassi et al., 2008), and the Lot River France receiving inputs from mining and smelting activities since the late 19th century (EF Pb = 10, EF Cu = 5) (Audry et al., 2004).

EFs for As are of concern in Owen Tarn and Basin Lake. Cd in Owen Tarn and Basin Lake are also significantly higher, with Cd in Basin Lake yielding an increase of 25-fold from background concentrations (indicating severe enrichment) (Table 4).

Although Se concentrations increase in most lakes, only Owen Tarn has severe enrichment while Perched Lake has moderate enrichment. Therefore, Se concentration increases in these two lakes should be taken into consideration in further studies.

The extremely high enrichment of these elements in the TWWHA and surrounding area supports the need to investigate the effects of mining contamination on aquatic organisms in western Tasmania, given that 15,842 km^2 (one fifth of the island) is Wilderness World Heritage Area.

3.5. Comparison of metal concentrations and ANZECC/ARMCANZ (2000) sediment quality guidelines

The Australian and New Zealand interim sediment quality guidelines (ISQGs) (ANZECC/ARMCANZ, 2000) comprise two sediment guideline concentrations: (1) ISQG-Low concentrations and (2) ISQG-High concentrations.

ISQG-Low concentrations is the threshold limit above which possible adverse biological effects in aquatic organisms may occur. The ISQG-High concentration is a threshold limit above which adverse biological effects are expected to occur frequently in aquatic organisms.

Table 5 shows the ratios of maximum concentration to sediment quality guideline values, being the concentration of a given metal in the sediment divided by that of the ISQG-low and ISQG high values. Results show that for all lakes other than Lakes Dobson and Cygnus, at least two out of Pb, Cu, As and Cd are above the respective ISQG-Low threshold limit. Of particular concern are Pb and As concentrations in Owen Tarn and Basin Lake, and Pb in Lake Dove, which are above the ISQG-High threshold limit.

In Australia, there are no selenium guidelines for sediments. The Screening QuickReference Tables (SQuiRTs) developed by the National Oceanic and Atmospheric Administration (Buchman, 2008) were, therefore, used to assess Se contamination in sediments. Although SQuiRTs screening values are intended for preliminary screening purposes only, Owen Tarn has shown a Se concentration 11 times higher than the SQuiRTs screening values, indicating severe contamination

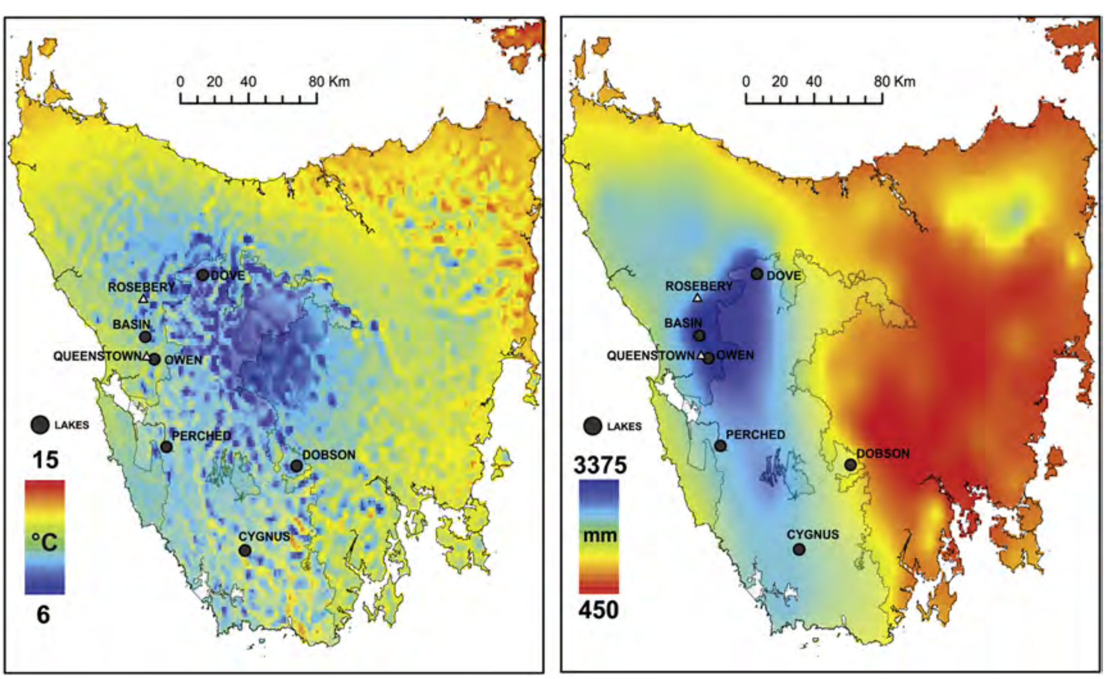

Fig. 4. (A) Atmospheric temperature (degrees Celsius) and (B) precipitation (mm) in the Tasmanian Wilderness World Heritage Area lakes (i.e. Lakes Dove, Perched, Dobson and Cygnus), and lakes near Queenstown (Owen Tarn) and Rosebery (Basin Lake) for the period 1961–1990. Data from Australian Bureau of Meteorology, maps developed in ArcMap 10.3.

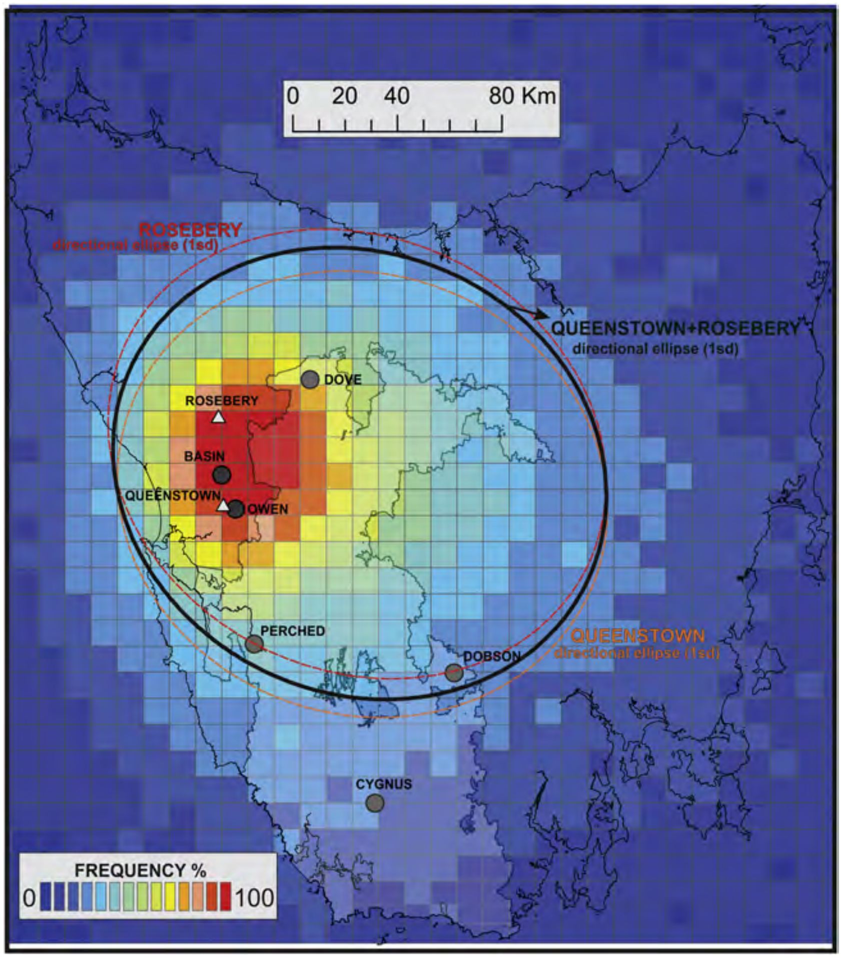

Fig. 5. The Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT) forward trajectories calculations demonstrating air parcel trajectories and directions of atmospheric particles and associated metals in the Tasmanian Wilderness World Heritage Area. This map represents the average circulation of air masses over Tasmania during the period 1961–1990 for particles released at Queenstown (42°S, 145.5°E) and Rosebery (41.78°S, 145.5°E). 1sd is one standard deviation.

and a likelihood of adverse biological affects in the area. The Se concentrations in Owen Tarn (up to 17 mg/kg in the 1950s) is higher than concentrations reported in the sediments of Belews Lake, North Carolina, a lake heavily contaminated by Se in wastewater released from a coal-fired electric generating facility during 1974–1985 (Lemly, 1997). In Belews Lake, Se concentrations of 4 to 12 mg/g in sediments were high enough to cause severe reproductive failure and teratogenic deformities in fish. It is likely that Owen Tarn organisms might be facing health issues due to Se contamination.

Table 3
Linear model results (R^2 and p value) for environmental factors influencing metal atmospheric transport and metal deposition in sediments of the four lakes in the Tasmanian Wilderness World Heritage Area (Perched Lake and Lakes Dove, Dobson and Cygnus) and two lakes closer to the mining centres (Basin Lake and Owen Tarn).

Metal	Predictors					Statistics	
	Distance	Precipitation	Size	Temperature	Frequency	R^2	P
Al	Removed	Removed	***	***	***	0.87	***
Fe	Removed	Removed	**	NS	NS	0.09	**
Pb	Removed	Removed	NS	NS	***	0.51	***
Cu	Removed	Removed	NS	NS	***	0.67	***
Zn	Removed	Removed	***	NS	**	0.16	***
As	Removed	Removed	NS	NS	***	0.38	***
Se	Removed	Removed	NS	NS	***	0.19	***
Cd	Removed	Removed	NS	***	***	0.13	**

NS = not significant.
* $P < 0.05$.
** $P < 0.01$.
*** $P < 0.001$.

The EF and sediment guidelines indicate that the northwest side of the TWWHA has been severely contaminated (Tables 4 and 5), and most likely have generated adverse biological effects in aquatic organisms. This is of great concern considering that contamination in organisms takes place through bioaccumulation from sediments to plants (Schneider et al., 2015) and its subsequent movement through trophic levels to animals and humans (Schneider et al., 2018). No study testing the health of aquatic organisms has been conducted in the area.

Studies in other areas of western Tasmania (De Blas, 1994; Humphrey et al., 1997; Keele, 2003; Rae, 2005) have shown metal concentrations in food web organisms above guideline limits proposed by the World Health Organisation (WHO, 1993). In Owen Tarn, a change in diatom composition from oligotrophic to those more characteristic of dystrophic, acidic lake waters, and a decline in species richness occurred in response to mining activities (Hodgson et al., 2000). It was also found that valve deformations in *Eunotia* species were a response to chemical stress (Hodgson et al., 2000). A study of metal bioaccumulation and toxicity of aquatic organisms within the TWWHA is highly recommended.

3.6. Government regulations and inconsideration

This study demonstrates the atmospheric extent of deposition of metals in the TWWHA from past mining activities. During Tasmania's prosperous mining phase, mining companies were not subject to the same environmental regulations as the present day.

Table 4
Enrichment factors of metals in the sediments of the four lakes in the Tasmania Wilderness World Heritage Area (Perched Lake and Lakes Dove, Dobson and Cygnus) and two lakes closer to the mining centres (Basin Lake and Owen Tarn).

Lake	As	Cd	Cu	Fe	Pb	Se	Zn
Owen Tarn	30.2	14.8	97.7	3.4	91	8.7	26
Basin Lake	5.4	27.4	21.9	0.7	28.4	0.8	3.4
Lake Dove	2.4	0.3	1	0.4	7.5	0.2	0.7
Lake Cygnus	1.2	0.7	6.1	1.2	1.7	0.1	1.6
Lake Dobson	1.9	45	1.3	3.1	10.2	N/A	3.2
Perched Lake	2.2	1.6	2.8	1.5	11	3.3	1
	EF < 1	EF < 3	EF 3–5	EF 5–10	EF 10–25	EF 25–50	EF > 50

Footnote: white = no enrichment, very light grey = minor enrichment, light grey = moderate enrichment, mid grey = moderately severe enrichment, dark grey = severe enrichment, very dark grey = very severe enrichment, and black = extremely severe enrichment. N/A = not applicable, concentration below detection limit.

Tasmania implemented and integrated environment protection legislation in 1973, when the *Environment Protection Act 1973* (comprising air, water and noise pollution, and waste management) was put in place. Even though Tasmania was one of the first states to have environment protection legislation in Australia (Bingham, 1992), mining companies were allowed to operate under exemptions that were granted by the government of the day. The argument supporting exemptions was that the cost of installing equipment to comply with emission standards would be such that the mine would have to close (De Blas, 1994).

Had regulations been strictly followed, it is probable that the metal contamination in the TWWHA would only be less severe and would have left a minor legacy of metal contamination. The high historical metal concentrations in lake sediments reported in this study leads to the question of how to and who should deal with the legacy of environmental problems arising from long running or discontinued activities,

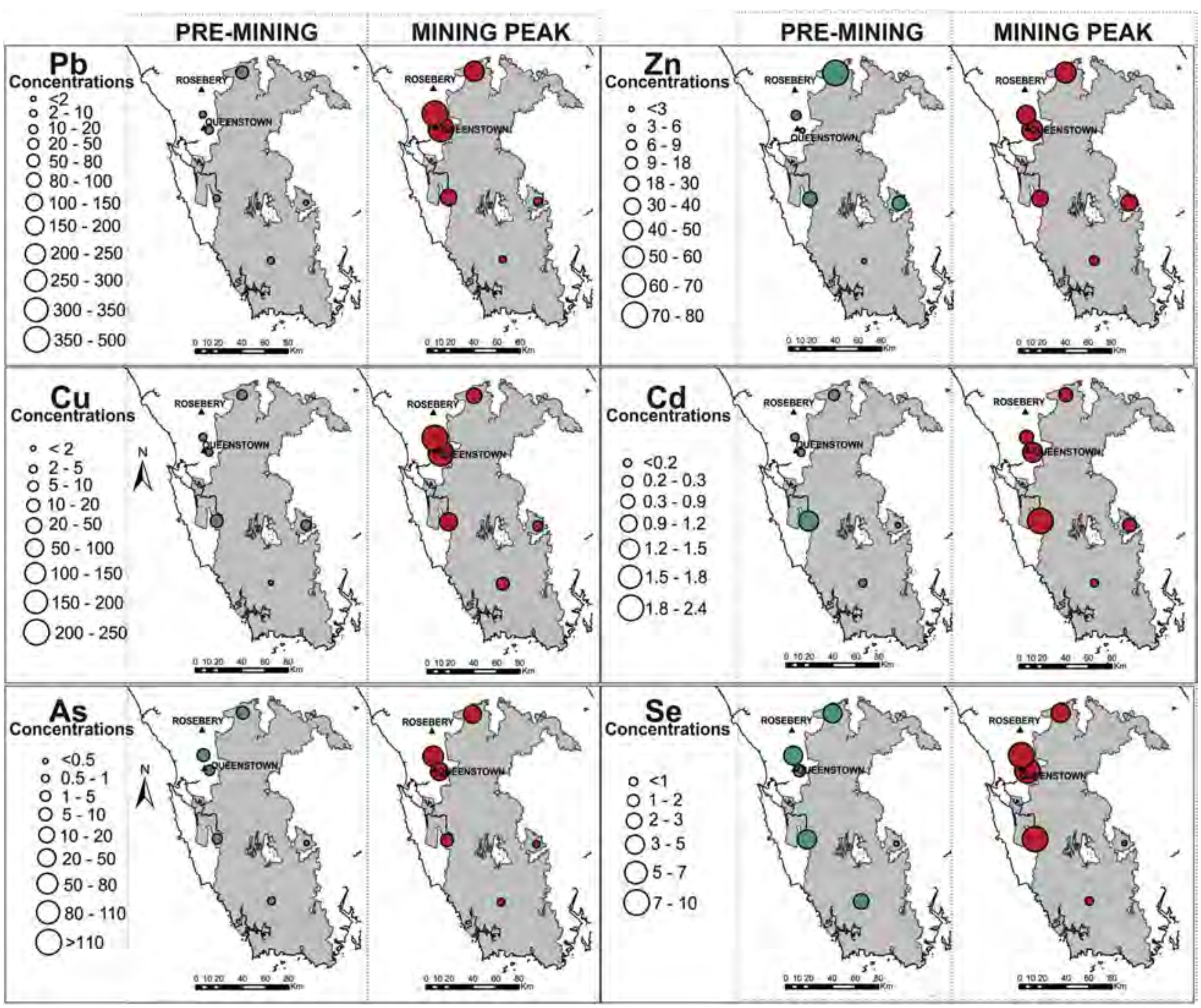

Fig. 6. Metal concentrations (mg/kg) in the four lakes in the Tasmanian Wilderness World Heritage Area (Perched Lake and Lakes Dove, Dobson and Cygnus) and two lakes closer to the mining centres (Basin Lake and Owen Tarn) pre-mining and during its peak (1930–1980).

Table 5
Ratios of maximum concentration of sediments (average concentrations from 1930 to 1980) from lakes in this study to ANZECC/ARMCANZ (2000) sediment quality guideline values. Metal concentrations highlighted in yellow indicate that the metal concentrations are above guidelines values.

Lakes	Pb		Cu		Zn		As		Cd		Se
	ISQG-Low	ISQG-High	ISQG-Low	ISQG-High	ISQG-Low	ISQG-High	ISQG-Low	ISQG-High	ISQG-Low	ISQG-High	SQuiRTs
Owen Tarn	18.4	4.2	6.0	0.7	0.6	0.3	8.1	2.3	2.2	0.3	11.6
Basin Lake	10.8	2.4	3.5	0.0	0.3	0.1	6.8	1.9	0.5	0.1	0.4
Lake Dove	6.2	1.4	0.9	0.0	0.5	0.2	1.9	0.5	0.5	0.1	0.6
Lake Cygnus	0.2	0.0	0.2	0.0	0.0	0.0	0.1	0.0	0.1	0.0	0.1
Lake Dobson	0.4	0.1	0.2	0.0	0.3	0.1	0.1	0.0	0.5	0.1	0.6
Perched Lake	2.9	0.6	1.4	0.0	0.7	0.3	0.5	0.1	3.2	0.5	2.5

which in earlier times had no environmental management protocols in place or lacked legal compliance to guidelines.

4. Conclusions

This study demonstrates that historical metal concentrations in lake sediments can assist in interpreting the extent and severity of metal contamination in pristine areas. While independent studies and governmental reports have focused on the environmental effects of mining contamination in the King River and Macquarie Harbour, this study demonstrates that the atmospheric transport of metals has caused contamination to sites outside the mining catchment areas.

Atmospheric metal contaminates from mining activities in Queenstown-Rosebery in Tasmania have contaminated most of the TWWHA area and have significantly altered the natural geochemical signal of lakes. The precipitous increase in metal contamination from the 1930s, due to the start of open-cut mining and introduction of new technology, demonstrates the importance of considering historical records when interpreting metal contamination.

The HYSPLIT forward trajectories particle model has been demonstrated to be a useful tool to track past metal contamination from airborne sources, explaining most of the metal atmospheric transport and metal deposition into the lakes of the TWWHA. Sediment EF values > 90 (classified as extremely severe enrichment) and metal concentrations above ISQG-High guidelines indicate that metal contamination might be posing health risks to aquatic organisms and humans feeding on them. Lakes closest to Queenstown and Rosebery, just outside the TWWHA, recorded some of the highest EFs for Pb and Cu reported in the scientific literature, while all lakes other than Lakes Dobson and Cygnus had at least one metal above ISQG-low threshold values. Further investigation of metal bioaccumulation in ecosystems of the TWWHA is warranted starting in the northwest where the metal contamination is highest.

Although mining activities have decreased significantly in the region, the metals deposited in the sediments are constantly remobilised by redox reactions, wind, catchment leaching and activities of microorganisms in the sediment. The environmental contamination in the TWWHA, therefore, is not a past issue and justifies current attention.

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2018.11.241.

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