Lead and zinc dust depositions from ore trains characterised using lead isotopic compositions

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Graphical Abstract

Textual Abstract

Elevated lead and zinc concentrations in remote environments can be traced to uncovered transport of ore concentrates from mining operations.

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Environmental Impact Statement

This paper examines lead and zinc deposits from a long history of transporting uncovered ore concentrates that have caused contamination along hundreds of kilometres of train lines. These transport corridors pass through pristine environments and local towns resulting in elevated concentrations of the toxic metal lead. Dangerous levels of lead have been found in drinking water supplies in these towns and our study reveals dusts in houses to contain lead and zinc ore. This shows that the impact of mining operations can be detected well beyond acceptable limits. This study has widespread implications to any mining operations or indeed, any operations where hazardous materials are transported and can cause losses into the environment.

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Lead and zinc dust depositions from ore trains characterised using lead isotopic compositions

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This study investigates an unusual source of environmental lead contamination - the emission and deposition of lead and zinc concentrates along train lines into and out of Australia's oldest silver-lead-zinc mine at Broken Hill, Australia. Transport of lead and zinc ore concentrates from the Broken Hill mines has occurred for more than 125 years, during which time the majority was moved in uncovered rail wagons. A significant amount of ore was lost to the adjoining environments, resulting in soil immediately adjacent to train lines elevated with concentrations of lead (695 mg/kg) and zinc (2230 mg/kg). Concentrations of lead and zinc decreased away from the train line and also with depth shown in soil profiles. Lead isotopic compositions demonstrated the soil lead contained Broken Hill ore in increasing percentages closer to the train line, with up to 97 % apportioned to the mined Broken Hill ore body. SEM examination showed ceiling dusts collected from houses along the train line were composed of unweathered galena particles, characteristic of the concentrate transported in the rail wagons. The loss of ore from the uncovered wagons has significantly extended the environmental footprint of contamination from local mining operations over an area extending hundreds of kilometres along each of the three train lines.

Introduction

Mining in Broken Hill, New South Wales (NSW) commenced in 1884 after the discovery of the world's largest lead-zinc-silver ore body in 1883[1]. From 1886 until 1897 smelting of the ore was conducted on site with up to 28 smelters in operation[2]. Due to a lack of local fuel sources for the smelters, it became necessary to conduct smelting operations outside of Broken Hill. The South Australian Government responded quickly to the opportunity and constructed a train line in 1885[3] in order to transport the raw ore to the new Port Pirie smelter. South Australian Railways opened their train line for operation in June 1887 to connect Adelaide and Port Pirie in South Australia (SA) to Cockburn on the NSW/SA border close to Broken Hill[4]. As the NSW government refused the construct a rail link from Cockburn to Broken Hill[3], the privately owned Silverton Tramway Company (STC) train line was subsequently opened for operation in January 1888[1-3] to connect the SA train line at Cockburn to trains coming from Broken Hill, via Silverton (Figure 1). Following completion of the train line connections between Broken Hill and Port Pirie, and the start of smelting operations in Port Pirie in June 1889, ore was transported to Port Pirie for smelting via rail on the STC and South Australian Railways narrow gauge (1067 mm) train line in uncovered wagons[5]. By 1897 all smelting operations were moved to Port Pirie[1, 2]. As of the end of June 1964, the Silverton train line had transported 38,747,602 tonnes of ore concentrate from Broken Hill[2].

In 1970, a new standard gauge train line (1435 mm) connecting Broken Hill to Port Pirie via Triple Chance (Figure 1) enabled a greater tonnage of ore to be carried on this route. However, not all ore concentrates were sent to Port Pirie for smelting, as lead and zinc concentrates were also sent eastward via rail to either Rozelle in Sydney, NSW for shipment overseas or Cockle Creek, NSW for further processing[2]. The standard gauge Broken Hill to Sydney train line was opened in October 1927[1] and 594,498 tonnes of lead and zinc concentrate had been transported via this line during the period from 1947 to the end of June 1964[2], although these ore wagons were covered with tarpaulins.

Ore was loaded into the wagons wet at Broken Hill, however, it would dry rapidly while in the shunting yards and also during transport (summer temperatures exceed 40°C[6]). The result was that, as the uncovered ore was moved between Broken Hill and Port Pirie, it was subjected to dispersal along the train line, either by wind, train movement or through gaps in the wagons[5]. A survey of the Broken Hill to Port Pirie railway corridor was conducted in 1986 to investigate lead losses from ore trains as part of the Port Pirie Lead Survey[5]. Forty-five transects of surface soils (0-5 cm) across the train lines were sampled between Broken Hill and Port Pirie, including both the old narrow gauge (Silverton line) and new standard gauge train lines. The study showed significant elevation of soil lead levels (over 21,000 mg/kg), immediately adjacent to the train tracks, with lead concentrations decreasing with distance away from the tracks[5]. The railway corridor survey also investigated the difference between the old and new train lines where they deviated from each other and found a noticeable increase in lead concentration along the old rail corridor. This was to be expected given that the new standard gauge line had only been subject to 15 years of ore dust deposition at the time of the study in 1986[5], compared to more than 80 years of ore movements along the old narrow gauge line.

In 1996 in a delayed response to the Broken Hill railway corridor survey, the NSW Environment Protection Authority (NSW EPA) required that all ore wagons be covered to prevent ore dust being blown from the wagons into the surrounding environment[7]. In 1997, fiberglass covers were introduced for ore wagons carrying lead and zinc ores to eliminate product loss and reduce the environmental risk of ore wagon dust emissions[8, 9].

While the Broken Hill railway corridor survey in 1986 characterised the extent of lead contamination in surface soil along the Broken Hill to Port Pirie train line, there is limited published literature on the loss and impact of heavy metal ore minerals during rail transport. Lead and zinc dust emissions from uncovered ore wagons from the Red Dog Mine in the Yukon Territory showed extensive contamination of the city of Skagway, where the trains passed, with elevated levels of lead along the transport routes of up to 28,000 mg/kg[10]. Emission losses from the haulage of ore has been occurring since ore was first transported, with lead and zinc contamination identified adjacent to Roman roads from as early as the 2nd-4th century[11].

This study examines the effect of industrial processes on the environment in and around the lead and zinc ore mining town of Broken Hill, Australia. Although the effects of lead exposure on young children in Broken Hill has been well documented for over 20 years[12], the source and cause of childhood blood lead exposures has not been clarified accurately. In 1993 Woodward-Clyde[13] completed a report on the environmental lead problem in Broken Hill and concluded "the primary source of this lead is the ore body, arising from erosion of the surface due to geological processes, and the mining and mineral processing activities over the past 100 years". This unfounded 'belief' about the natural source and cause of the lead problem has stymied a fully integrated strategy to deal with emissions from the Broken Hill mining operations. Indeed, many residents in Broken Hill still believe that the environmental lead problem is predominantly due to lead occurring naturally in the soils and dusts around the city[14].

Therefore, this study examines the role of ore transport as a contributing source of industrial contamination on the environment in and around Broken Hill city, and importantly to the environment outside of mining operations. The impact from ore transport on Broken Hill train lines is evaluated by measurement of lead and zinc concentrations in soils and dusts. Lead isotopic composition and scanning electron microscopy analyses are used to confirm the source and origin of the environmental contamination.

Methods

Field sampling methods

Soils were collected along transects perpendicular to train lines at varying distances up to 150 m from the line and at varying depths (0-2 cm; 2-10 cm; 10-20 cm; 20-30 cm; 30-40 cm; 40-50 cm) using established methods[15]. Soil samples collected at depth were intended to provide local background values (total metal concentrations and lead isotopic composition) compared to the more recently contaminated surface samples. Soil samples (n=104) were collected along four train line transects in 2009, 2011 and 2012.

Soil samples (n=36) along the standard gauge Broken Hill to Port Pirie train line were collected 31 km SW of Broken Hill at the turn off to Triple Chance Mine in October 2009 (referred to as Triple Chance). Soil samples (n=18) along the old narrow gauge train line were collected on the Silverton Tramway Company train line 10 km NW of Broken Hill in September 2011. The Broken Hill to Sydney train line was sampled in two locations, just outside the limits of Broken Hill city (referred to as Broken Hill (n=18)) in September 2011 and at The Gorge 27 km east along the train line (n=32) in September 2012.

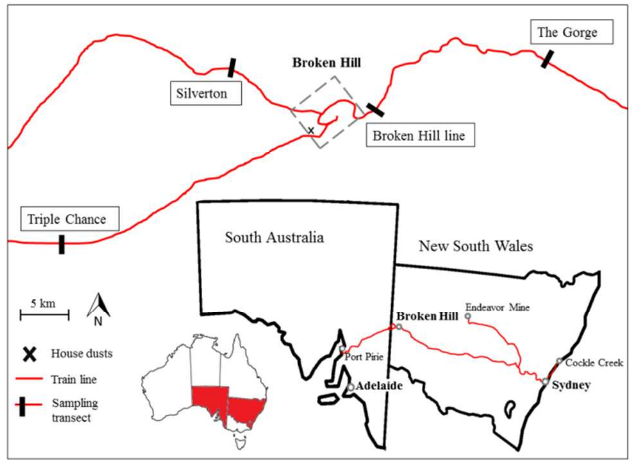

Figure 1: Map of train lines into and out of Broken Hill along with the location of sampling transects.

Laboratory analysis

All 104 soil samples were analysed for lead and zinc concentrations and select samples (n=35) for lead isotopic composition. Samples collected in 2009 (n=36) and 2012 (n=32) were analysed at National Measurement Institute (NMI), North Ryde, NSW and samples collected in 2011 (n=36) were analysed by ChemCentre, Western Australia for lead and zinc concentrations and lead isotopic compositions. Samples were digested in a mix of HCl and HNO_3 and measured for lead and zinc concentrations on a Varian Vista Pro ICP-OES and for lead isotopic compositions on a PerkinElmer Elan DRC II and Agilent 7500ce ICP-MS. Blanks returned less than 0.01 mg/kg for both Pb and Zn.

Duplicate analysis returned relative standard deviations (RSD) <6 % for Pb and <7 % for Zn. Recovery rates at NMI were measured using reference material AGAL-10 and sample matrix spiking. Recovery rates were 91 % for Pb and 100 % for Zn for the certified reference material and 90 % for Pb and 106 % for Zn for the matrix spikes. Lead isotopic compositions were determined for select samples (n=16) and mass fractionation was corrected using NIST SRM981 (common lead standard). RSDs reported by NMI for $^{206}Pb/^{204}Pb$, $^{206}Pb/^{207}Pb$ and $^{208}Pb/^{207}Pb$ were 0.12%, 0.07% and 0.08 % respectively. Recovery rates at ChemCentre were measured using NRCC reference material PACS-2 with recovery rates for Pb 102 % and 106 % for Zn. Lead isotopic compositions were determined for selected samples (n=16) and mass fractionation was corrected using NIST SRM982 (equal-atom lead isotopic standard). RSDs reported by ChemCentre for $^{206}Pb/^{204}Pb$, $^{207}Pb/^{206}Pb$ and $^{208}Pb/^{206}Pb$ were 0.23 %, 0.23 % and 0.24 % respectively.

Two ceiling dust samples (House#1 and House#2) were sampled and approximately 2 g of each material was subjected to heavy media separation using a sodium polytungstate solution at a density of approximately 3 g/cm^3 to remove organic components and minerals such as silica with a low density leaving the heavy metal components. The high density component of the ceiling dust samples were mounted into epoxy resin (Araldite) blocks. A section through the mounted material was polished on a diamond lap to a fine finish and examined with a JEOL-JSM-6480LV high-performance scanning electron microscope (SEM) using a resolution of 3.0 nm in combination with an energy dispersive X-ray spectrometer (EDS). In back scatter electron imaging mode, materials of high atomic density (such as lead minerals) appear brightest on the screen and are therefore more easily identified[16].

Results and Discussion

Concentrations of lead and zinc along the soil sample transects covered a large range; Pb: 7-695 mg/kg; Zn: 19-2230 mg/kg. Lead, zinc along with copper concentrations and lead isotopic composition data for soils sampled along the train line transects can be found in Supplementary Table S1.

Lead and zinc total soil concentrations

Soil samples collected from the original narrow gauge Silverton train line (in operation 1888-1970) show elevated concentrations of lead and zinc as both a function of distance from the train line and with depth through the soil profiles (Figure 2). Lead and zinc concentrations decrease in surface soils (0-2 cm) at the sample points moving outwards in both the northerly and southerly directions from the train line. The deepest sub-surface soils (40-50 cm) remain consistent in both lead and zinc concentration across the transect and within the range considered to be natural soil concentration levels[17] (Pb: <30 mg/kg[17, 18]; Zn: <100 mg/kg[17]). Dispersal patterns of lead and zinc are highly correlated (Pearson correlation r=0.998, p<0.01) indicating that losses of ore from the train wagons was the only likely source.

The change in concentration through the soil profile expressed as a Surface Enrichment Ratio (SER)[15], shows a consistent pattern at each location along the transect. The SER decreases with distance from both sides of the train line reflecting a decreasing contribution from the external point source. It is also evident from the soil profiles (Figure 2) that the most dramatic change in lead and zinc concentrations occur between the top two sampled soil layers: 0-2 cm and 2-10 cm. This demonstrates that elevated lead and zinc concentrations are found only in the surface soils must be sourced from recent atmospheric deposition and are not likely to have existed before the operation of the train line.

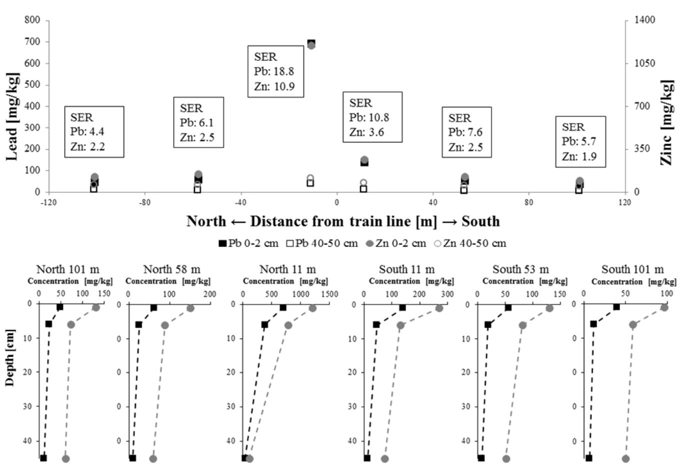

Figure 2: Soil lead and zinc soil concentration profiles either side of Silverton train line.

While the lead concentrations measured in this study are not as high as those found in the Port Pirie Lead Survey[5], the pattern of dispersal from the train lines is consistent. Surface soils analysed by in 1986 Body in the first transect along the Silverton train line returned lead concentrations of 139 and 1404 mg/kg, 10 m north and south of the train line[5], which are consistent with the findings of this study.

Lead and zinc concentrations in the soil samples from the new standard gauge line (1970-present) collected at Triple Chance (Figure 3) display a similar spatial pattern of lead and zinc concentrations to those along the Silverton train line (Figure 2). Soil samples were collected at depth intervals along the Triple Chance transect, with the soil profiles (Figure 3) revealing that surface enrichment generally only occurs in the top (0-2 cm) layer of soils. Below the surface there is a sharp decrease in concentration in the immediate sub-surface layers (2-10 cm), which continues down the soil profile. While the lead and zinc concentrations are also correlated at this train line (Pearson correlation r=0.998, p<0.01), lower lead concentrations and higher zinc concentrations are seen by comparison to those from the Silverton train line. The likely cause of these changes in concentration is the increase in zinc ore production and its transport along the new line in recent years. By 1966, 13.2 million tonnes of lead ore and 9.4 million tonnes zinc ore was produced in Broken Hill[1] while in 1995-1999 only 896,550 tonnes of lead ore compared to 1,857,406 tonnes zinc ore was produced[19].

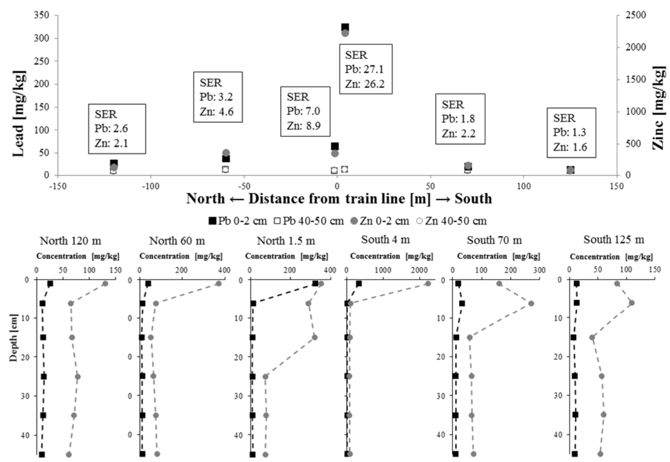

Figure 3: Soil lead and zinc concentration profiles across Triple Chance transect.

In contrast to Silverton and Triple Chance transects, the lead and zinc concentrations across the Broken Hill transect, which is close to the city and mining operations, do not decrease to the same extent with distance from the rail line. The SERs along this transect are the highest of this study and also do not correlate to distance from train line. The lead-zinc soil concentration profiles are likely to have been influenced to a greater degree the nearby mining Broken Hill operations than by ore emissions from trains (Supplementary Figure S1).

In order to ascertain the impact of ore wagons on the Broken Hill to Sydney train line without the confounding effect of the Broken Hill mining operations, the train line was resampled, further along the train line at The Gorge in 2012 (Figure 1), some 19 km from the city (27 km by train). Peak lead and zinc concentrations occur closest to the train line and decline with distance (Supplementary Figure S2), which is characteristic of the effect of ore concentrate losses from the trains. However, it cannot be determined from total lead and zinc concentrations alone whether the losses from the ore wagons are solely from those going east out of Broken Hill. Lead concentrates were also transported west to Broken Hill on route to Port Pirie from 1983[20] from the Endeavor mine, which is 405 km east of Broken Hill (~1220 km by train). While the SERs demonstrate clearly the magnitude of surface soil enrichment when compared to background metal concentrations found at depth, they are all lower than those found at the Broken Hill City location, indicating limited deposition from ore wagons. The lower soil concentrations are consistent with the small tonnage of ore coming westwards from the Endeavor mine combined with that the small tonnage of Broken Hill ore going eastwards to the NSW coast for processing, by comparison with that going to the Port Pirie smelter. The effect of mining operation emissions on The Gorge transect can be dismissed by virtue of the spatial pattern of increasing surface lead and zinc concentrations closer to the rail line, indicating the only logical point source for the depositions are emissions from ore wagons.

Across all transects, the highest concentrations of lead and zinc were found consistently in the surface soils (0-2 cm) closest to the train lines. The lowest concentration of lead and zinc were found both at distance and depth with the majority in the deepest sub-surface samples (40-50 cm) at the most distal points of the samples transects (Supplementary Figure S2). The lower soil metal concentrations at depth and distance from the rail lines are considered to be characteristic of background lead and zinc concentrations[17]. The consistent correlation of lead and zinc concentrations in the soil samples further supports the argument of a single contributing source[15]. For the lead and zinc in the soil to be naturally occurring, other metals would also be expected to display a similar spatial pattern, which they do not. Copper soil concentrations for example, remain more or less constant regardless of the distance from the train lines or depth of sampling (Supplementary Table S1). This consistent pattern of lead and zinc dispersion across the transects is clear evidence that the origin of the elevated metal levels result from emission and dust deposition of ore and concentrate products from the uncovered ore rail wagons. The same soil lead concentration pattern of elevated surface concentration and rapid subsurface concentration decline has been observed around city highways as a result of leaded petrol usage[21].

Lead isotopic compositions

Although lead and zinc concentrations in the soil samples show clear spatial patterns of ore wagon dust emissions and subsequent depositions, total soil concentration values do not allow source apportionment or enable percentage contributions from the Broken Hill ore to be established. To address this question, lead isotopic compositions of the sampled soils collected along the four transects were measured. A sample of Broken Hill ore was also measured for its lead isotopic composition with values (Supplementary Table S1) shown to be match published literature values[22-24]. The lead isotope compositions of the Triple Chance transect are shown in Figure 4, with all other transects in the Supplementary Information (Silverton transect: Supplementary Figure S3; Broken Hill transect: Supplementary Figure S4; The Gorge transect: Supplementary Figure S5).

The lead isotopic compositions of surface soils at all train transects plot directly on a mixing line between local natural soil lead and Broken Hill ore (e.g. the Triple Chance transect, Figure 4). The same, consistent pattern is observed in all train transects not influenced by mining operations, with surface soil samples (0-2 cm) becoming increasingly similar with respect to the background lead isotopic compositions (40-50 cm; at >100 m from the train line) with increasing distance from train line.

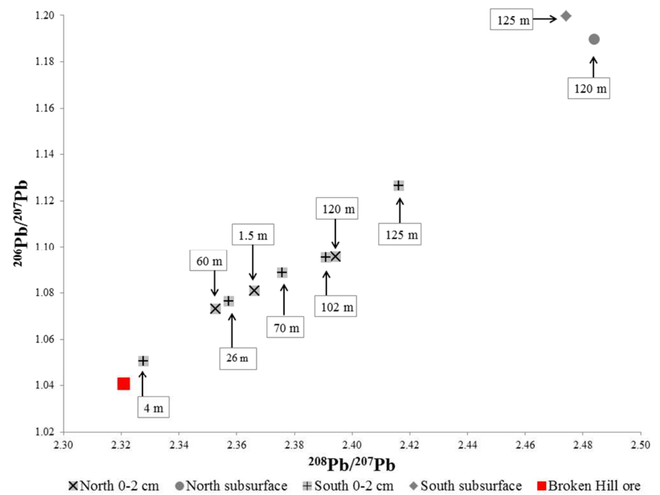

Figure 4: Soil lead isotopic compositions at either side of the rail line at Triple Chance, benchmarked against Broken Hill ore.

A two end member mixing model[25, 26] was applied to the surface soil samples to quantify the contribution of Broken Hill lead derived from the loss of ore from uncovered wagons. Surface soil samples (0-2 cm) at Triple Chance closest to the train line (≤70 m) contained 73-93 % Broken Hill ore, while samples at distance (120 m north; 125 m south) only contained 46-63 % Broken Hill ore. The surface soils along the Silverton transect have higher proportions of Broken Hill ore with 94-97 % closest to the train line. Surface soils furthest from the train line (101 m either side) consist of 77-78 % lead from Broken Hill ore. Of all of transects, lead in surface soils at The Gorge transect were closer to those representing background values (40-50 cm, 100 m). Mixing model calculations apportioned 42-73 % of the lead in surface soil samples ≤51 m from the train line and 35-46 % of lead in soils 100 m from the train line to the Broken Hill ore body. Nevertheless, the effect of the loss of ore on surface soils is demonstrated clearly by comparison to sub-surface soils (40-50 cm) at the same location, which have a markedly different isotopic composition (Figure 4; Supplementary Figures S1-S3).

The Gorge site was sampled in order to assess the Broken Hill to Sydney train line without the confounding impact of mining in Broken Hill as the proximity of the site to the central and north mine in the city of Broken Hill (<2 km). It is highly likely that the surface lead contaminants at the Broken Hill transect are not sourced entirely from train transport of the ore, but also from wind dispersion of ore from current and historic mining practices within Broken Hill city. The fact that the Broken Hill ore contribution at The Gorge is markedly lower than that measured at the other three locations is likely to be due to two main factors. Firstly, there has been significantly less ore transported along this line compared to that transported via the Silverton or the Triple Chance line. Secondly, the ore from the Endeavor mine that was transported to Broken Hill for almost 15 years has a lead isotopic composition not dissimilar to that of the background soils of the Broken Hill region (Endeavor ore $^{208}Pb/^{207}Pb$: 2.448; $^{206}Pb/^{207}Pb$: 1.152)[27] as identified at The Gorge site. From the commencement of mining at Endeavor in 1983, lead concentrates were railed to Port Pirie via Broken Hill, with 896,026 tonnes being transported by 2009[20]. Endeavor mine to Broken Hill ore wagons were however, not covered until 2000[20]. Although lead ore from the Endeavor mine also passed through Broken Hill, it was a fraction of that produced and transported from the Broken Hill mines. For example, 145,576 tonnes of (wet) lead ore concentrates were transported from Endeavor mine to Port Pirie between 1991 and 1994, while by significant contrast, a total of 1,990,157 tonnes of (wet) lead ore concentrates were transported from Broken Hill to Port Pirie in the same time period[28].

At the NSW/SA border town of Cockburn, 50 km from Broken Hill, which is located at the junction of the old narrow gauge (Silverton) and the new standard gauge (Triple Chance) train lines (Figure 1), ceiling dust was collected from a house immediately adjacent to rail corridor. Elevated total concentrations of lead (1,400 mg/kg) and zinc (3,300 mg/kg) and lead isotopic compositions were measured in the ceiling dust, which had the following values: $^{208}Pb/^{207}Pb$: 2.323; $^{206}Pb/^{207}Pb$: 1.045; $^{206}Pb/^{204}Pb$: 16.12. Given that these values are analogous to those of the Broken Hill ore (Supplementary Table S1), there is little doubt that the ceiling dust contaminants were sourced from the uncovered ore concentrates being transported along the train lines.

Overall, the results show conclusively that dust emissions from uncovered ore wagons transporting ore and concentrates to and from Broken Hill left a significant legacy of contamination in surface soils and dusts along the corridor.

SEM Examination

As an additional confirmation of the source of the lead being deposited along the Triple Chance rail line to Port Pirie, two samples of internal ceiling dusts from Broken Hill houses immediately adjacent to the rail corridor were examined using SEM and EDS. The houses from which these samples were obtained were to the north and within ~200 metres of the train line in Broken Hill city. The concentrated ore was crushed to fine particle size, which was predominantly less than 200 µm, making wind transport possible. The shunting of wagons broke the surface ore crusts that had dried in the heat of Broken Hill, rending them available for wind erosion[29]. The prevailing winds in the area would have contributed to the aeolian transport of concentrate dusts emitted from uncovered rail wagons towards these houses. Subsequent photographic evidence of significant dust losses from the wagons confirms the proposition that significant and visible losses from the wagon occurred (Supplementary Figure S6).

Galena (PbS), the principal lead mineral mined and concentrated from Broken Hill ore, is a relatively soft mineral (Mohs hardness 2.5) of high density (7.57 g/cm^3). The longer the material is exposed to the atmosphere, the more likely that the surface will not only react chemically but the external structure will be worn and rounded by collisions with other harder materials. The high density of galena ensures that large crystalline particles (~20 µm and greater) are unlikely to be windblown for significant distances without contacting the ground, thus requiring episodic re-entrainment for large travel distances[30], which would increase the potential for loss of crystallinity and angularity. Therefore, the existence of crystallinity, angularity and lack of surface alteration is indicative of an emission source in the immediate vicinity of deposition, followed by protection (e.g. a roof cavity) from the elements such as wind or rain.

EDS spectral output allows accurate elemental identification of individual mineral grains, and in the case of this study, visual observation of grain morphology and elemental determination were adequate to identify the mineral species. Even though heavy media concentration had been carried out on the ceiling dust samples, SEM examination showed that the dusts were extremely heterogeneous and contained many mineral species including galena, marmatite, pyrite and related alteration products.

The SEM examination concentrated on the larger lead bearing particles in the polished samples and typical examples of lead bearing particle types were selected for more detailed examination. From the polished section of the material from House #1, 23 particles were identified visually as potentially lead bearing examples were subjected to 48 separate EDS analyses. From the House #2 sample, 20 particles were visually selected and 28 EDS analyses were undertaken.

Not all particles selected visually proved to contain lead as there were also other high atomic density materials present, including silver, rare earths and residual sodium polytungstate (from the mineral separation process). The samples shown in Figure 5 contain lead bearing particles with significant similarity to lead concentrates, that is, particles showing crystallinity and lack of surface alteration. However, the sample also contained lead particles that were rounded, with significant surface alteration indicating weathering.

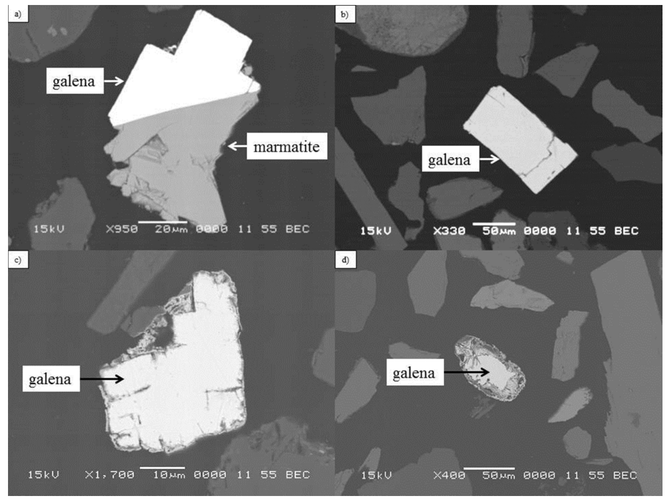

Figure 5: SEM micrographs of dust samples (scales shown on images) a) House #1 ~40-80µm composite crystalline galena and marmatite particle. b) House #2 ~150x 75µm galena particle. c) Galena particle from House #2. d) Galena particle from House #2.

Both the crystallinity, maintenance of angularity and lack of surface alteration in a large proportion of the lead containing particles is indicative of material that was transported short distances before deposition in the roof space of the houses. Examples of crystalline galena of this description are shown in Figures 5a and 5b, which both show cubic crystallinity, maintenance of angularity and lack of surface alteration. These features are typical of fresh, recently mined minerals and not those of old weathered long-term deposits being recycled in the environment.

A smaller proportion of the lead containing particles from the ceiling dusts were rounded, with surfaces that had been chemically altered. The EDS analyses showed that the surface layer frequently included high levels of O, Cl, P, Fe as well as Pb and S. The development of this type of morphology would require particles to have been exposed to the environment for extended periods of time, during which they may have been subject to more frequent and longer transport distances than the particles shown in Figures 5a and 5b. However, the large size of many of the particles implies that the source of origin is more likely to be close to their final deposition site in the roof spaces of the sampled houses[30]. Examples of these types of particles are shown in Figure 5c and 5d, which show rounding and surface alteration. In Figure 5d, the galena particle contains Cl and O in the surface layer encapsulating the galena.

Overall, the SEM examination of the two ceiling dust samples supports strongly the proposition that lead concentrates (and zinc by default) were emitted from uncovered rail wagons and that these emissions were deposited adjacent to the train line and in domestic residences within the city of Broken Hill.

As a result of the uncovered transport of ore concentrates, high levels of lead was shown to be found in house dusts and water tanks along the train lines in 1986[5] when ore wagons were uncovered. SEM examination of dust from ceilings along with recent reports of lead levels 10 times the drinking water guidelines (0.01 mg/L) in water tanks[31] in towns along the Triple Chance train line reveal a lingering legacy and health risks from mining activities outside of Broken Hill. This legacy of lead and zinc ore concentrates deposition along transport routes is not limited to Broken Hill as similar issues have been shown in Canada and Alaska, where uncovered transport of lead and zinc ore concentrates have resulted in elevated levels of toxic metals in adjoining communities[10].

The environmental and health consequences are not limited to lead and zinc transportation and could be found at any location where materials that pose a health risk, whether known or unknown, have been or are currently being transported in a way that causes losses to the environment to occur (Supplementary Figure S6). This was found to be the case in Russia, where the highest incidence of thyroid cancer occurred along transport routes, both train and road, from Chernobyl[32]. Losses of dust from transportation are difficult to quantify, however, experiments of fugitive dusts from uncovered coal trains calculates that 0.0048 % of the load is lost during uncovered transportation[33, 34]. Although not directly transferable to this study, this data provides an initial estimate to establishing overall lead and zinc losses from uncovered ore transportation in Broken Hill.

Conclusions

Soil lead and zinc concentrations from transects across train lines into and out of Broken Hill reveal the environmental impact from transporting ore concentrates in uncovered wagons. The soil lead isotopic composition from all transects show a clear difference between the surface soil samples and the sub-surface soil samples. Generally, surface soils that are closer to the train line have lead isotopic compositions that are similar to the lead isotopic composition of the ore mined from Broken Hill and transported by rail. SEM examination of deposited ceiling dusts from houses near the train line adds further credence to the chemical and isotopic evidence as to the source of lead. The morphology of particles found in these dusts is indicative of a source close to the point of deposition.

While the ore wagons are now covered by EPA mandate and losses of ore to the environment is no longer occurring as a result of train transport, this is not a resolved issue. This study shows that the contamination along the train lines is still present. Given the aridity of region and the prevalence of dust storms, transport of ore-contaminated dust particulates into nearby houses and water supplies is a current concern. Rehabilitation along the train lines is necessary to prevent further movement of contaminated soils.

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Supplementary Information

Supplementary Table S1: Soil copper, zinc and lead concentrations and lead isotopic compositions.

Supplementary Figure S1: Soil lead and zinc concentration profiles across Broken Hill train line transect at Broken Hill.

Supplementary Figure S2: Soil lead and zinc concentration profiles across Broken Hill train line transect at The Gorge.

Supplementary Figure S3: Soil lead isotopic compositions at either side of the rail line at Silverton, benchmarked against Broken Hill ore.

Supplementary Figure S4: Soil lead isotopic compositions at either side of the rail line at Broken Hill, benchmarked against Broken Hill ore.

Supplementary Figure S5: Soil lead isotopic compositions at either side of the rail line at The Gorge, benchmarked against Broken Hill ore.

Supplementary Figure S6: Photograph of a train moving ballast material ~ 50 mm in size, travelling to the east of Broken Hill. Note the plumes of dust coming from the ballast on the uncovered wagons. (Photo taken by M.P. Taylor 2009)

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