Environmental Pollution 242 (2018) 54–62

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

Tracing natural and industrial contamination and lead isotopic compositions in an Australian native bee species☆

Xiaoteng Zhou [a, *], Mark Patrick Taylor [a, b, **], Peter J. Davies [a]

[a] Department of Environmental Sciences, Faculty of Science and Engineering, Macquarie University, North Ryde, Sydney, NSW 2109, Australia
[b] Energy and Environmental Contaminants Research Centre, Faculty of Science and Engineering, Macquarie University, North Ryde, Sydney, NSW 2109, Australia

ARTICLE INFO

Article history:
Received 9 February 2018
Received in revised form
19 June 2018
Accepted 21 June 2018
Available online 22 June 2018

Keywords:
Bees
Bio-indicator
Honey
Leaded petrol
Pb isotopes
Tetragonula carbonaria

ABSTRACT

This study investigates trace element concentrations (arsenic (As), manganese (Mn), lead (Pb) and zinc (Zn)) and Pb isotopic compositions in an Australian native bee species, *Tetragonula carbonaria*, and its products of honey and wax. Co-located soil and dust samples were simultaneously analysed with the objective of determining if the bees or their products had potential application as a proxy for monitoring environmental contamination. The most significant relationships were found between Pb concentrations in honey ($r = 0.814$, $p = 0.014$) and wax ($r = 0.883$, $p = 0.004$) and those in co-located dust samples. In addition, Zn concentrations in honey and soil were significantly associated ($r = 0.709$, $p = 0.049$). Lead isotopic compositions of native bee products collected from background sites adjacent to Sydney national parks ($^{206}Pb/^{207}Pb = 1.144$, $^{208}Pb/^{207}Pb = 2.437$) corresponded to local geogenic rock and soil values ($^{206}Pb/^{207}Pb = 1.123–1.176$, $^{208}Pb/^{207}Pb = 2.413–2.500$). By contrast, inner Sydney metropolitan samples, including native bees and wax ($^{206}Pb/^{207}Pb = 1.072–1.121$, $^{208}Pb/^{207}Pb = 2.348–2.409$), co-located soil and dust ($^{206}Pb/^{207}Pb = 1.090–1.122$, $^{208}Pb/^{207}Pb = 2.368–2.403$), corresponded most closely to aerosols collected during the period of leaded petrol use ($^{206}Pb/^{207}Pb = 1.067–1.148$, $^{208}Pb/^{207}Pb = 2.341–2.410$). A large range of Pb isotopic compositions in beehive samples suggests that other legacy sources, such as Pb-based paints and industrials, may have also contributed to Pb contamination in beehive samples. Native bee data were compared to corresponding samples from the more common European honey bee (*Apis mellifera*). Although Pb isotopic compositions were similar in both species, significant differences in trace element concentrations were evident across the trace element suite, the bees and their products. The statistical association between *T. carbonaria* and co-located environmental contaminant concentrations were stronger than those in European honey bees, which may be attributable to its smaller foraging distance (0.3–0.7 km versus 5–9 km, respectively). This implies that *T. carbonaria* may be more suitable for assessing small spatial scale variations of trace element concentrations than European honey bees.

1. Introduction

Urban development and industrial activities have caused extensive trace element contamination of surface soil, dust and water across terrestrial environments in major cities around the world (Su et al., 2014). A large number of studies have reported elevated levels of trace elements in urban environments, giving rise to concern about potential harmful effects on human health from exposure to air, water, dust or soil contamination (e.g. Li et al., 2001; Turer et al., 2001; Islam et al., 2015; Rodriguez Martin et al., 2015; Trujillo-Gonzalez et al., 2016).

The rapid urbanisation of Sydney following European occupation in 1788 has resulted in trace element contamination of soils and dusts, including arsenic (As), lead (Pb), manganese (Mn) and zinc (Zn) (e.g. Gulson et al., 2006; Gulson et al., 2008; Birch et al., 2011; Laidlaw and Taylor, 2011; Gulson et al., 2014; Rouillon et al.,

☆ This paper has been recommended for acceptance by Prof. W. Wen-Xiong.

* Corresponding author.
** Corresponding author. Department of Environmental Sciences, Faculty of Science and Engineering, Macquarie University, North Ryde, Sydney, NSW 2109, Australia.
E-mail addresses: xiaoteng.zhou@students.mq.edu.au (X. Zhou), mark.taylor@mq.edu.au (M.P. Taylor).

https://doi.org/10.1016/j.envpol.2018.06.063
0269-7491/

2017; Zhou et al., 2018), all of which have known potential to impair human neurological systems, behavioural and cognitive abilities (Siegel, 2002). Sources of enriched trace elements are associated with emissions from vehicular traffic (Davis and Birch, 2010; Huber et al., 2016), urban and industrial wastes (Zhang et al., 2013), and mining and smelting of industrial activities (Kristensen and Taylor, 2016; Dong and Taylor, 2017). In addition, the former use of Pb-based paints and leaded petrol remain persistent in the environment (Gulson et al., 1995; Kristensen, 2015; Kristensen et al., 2017), and have contaminated urban soils (Birch et al., 2011; Rouillon et al., 2017) and dusts (Davis and Gulson, 2005; Laidlaw and Taylor, 2011; Dong et al., 2015). Across Australian cities, former leaded petrol depositions have also been measured in lichens (Wu et al., 2016b), fungi (Wu et al., 2016a), wildfire ash deposits (Kristensen et al., 2014; Wu et al., 2017) and seagrasses (Serrano et al., 2016).

The common European honey bee (*Apis mellifera*) has been assessed previously for its potential as bio-indicators of trace element contamination in urban (Perugini et al., 2011; Zarić et al., 2016), industrial (Bromenshenk et al., 1991; Matin et al., 2016) and mining areas (Lopes et al., 2010; Satta et al., 2012; Alvarez-Ayuso and Abad-Valle, 2017), as well as for apportioning Pb sources (Zhou et al., 2018). However, to the best knowledge of the authors, there is no equivalent research examining the utility of local native bees for the same purposes. The keeping of *T. carbonaria* has been promoted extensively in recent years in Australia (Halcroft et al., 2015; Heard and Dollin, 2015) as part of government and permaculture programs to connect people to nature. In the tropics, *T. carbonaria* is a commonly used bee species to assist with pollination services (Heard, 2000).

Compared to European honey bees (*A. mellifera*, introduced to Australia in 1822), the Australian native bee (*T. carbonaria*) has a number of important biological and behavioural differences. *T. carbonaria* has a smaller body (4 mm vs. 15 mm) (Smith et al., 2017), smaller foraging distance (0.3–0.7 km vs. 5–9 km) (Beekman and Ratnieks, 2000; Smith et al., 2017), shorter foraging time per day (7 h vs. 10 hrs) (Heard, 1999), produces less honey per year (1 kg vs. 50 kg) (Heard, 2016a), and has a longer lifespan (100 days vs. 40 days) (Heard, 2016b).

Given that previous studies have shown that European honey bees and their products can be used as proxy indicators to trace environmental contamination sources, it was considered that native bees could also be used as a bio-indicator. We hypothesise that their smaller foraging area may confer an advantage over European honey bees in that it may reduce the effect of contaminant heterogeneity that is known to exist in soils and dusts, even over small areas in urban environments (Schwarz et al., 2012; Rouillon et al., 2017). Various studies of European honey bees and their products have shown that they cannot distinguish subtle differences in trace element contamination across heterogeneous urban environments (Jones, 1987; Fakhimzadeh and Lodenius, 2000; Conti and Botre, 2001; Saunier et al., 2013; Alvarez-Ayuso and Abad-Valle, 2017). We also hypothesise that lower honey production in native bees may result in higher trace element concentrations in honey and wax providing a more robust measure of local environmental contamination, where present. Our previous work investigating European honey bee hive products across Sydney suburbs revealed that they were suitable bio-indicators for tracing As, Pb, Mn and Zn contamination (Zhou et al., 2018). This study, builds on these findings by investigating the same trace elements in the Australian native bee, *T. carbonaria*, and its honey and wax in order to: (a) to investigate its utility as a proxy marker for urban contaminants and its sources; and (b) assess its efficacy compared to European honey bees, which are more commonly used as a contaminant bio-indicator.

2. Materials and methods

2.1. Study area

Eighteen Australian native bee hives were sampled across Sydney, New South Wales (NSW) (Fig. 1). Eleven of those hives were located in Roseville, with other suburbs having only one native bee hive available for sampling. For comparative purposes, nine European honey bee hives (i.e. one European honey bee hive per native bee hive per site) close to the sampled native bee hives were also included in the study analysis (Fig. 1). Sample and analytical details for the European honey bee data have been published in Zhou et al. (2018). The locations of all the sampled bee hives range from sites in outer Sydney, coastal Sydney and inner Sydney, close to the central business district (CBD) (Fig. 1). All locations have been exposed to varying degrees of metal contamination, as a consequence of industrial activity (Birch et al., 2011; Rouillon et al., 2017), the use of Pb-based paints and emissions from vehicles previously using leaded petrol (Kristensen, 2015; Rouillon et al., 2017).

2.2. Sample collection

Australian native bees are very temperature dependent (Norgate et al., 2010) with their most intensive foraging period being over the warmer Australian summer months from November to early April. During this peak foraging time, pollen and nectar are collected and enough honey produced to sustain the native bees over the cooler winter months.

Over the sampling period (2015–2016), 30 live native bees were collected at the entrance of each beehive when they were observed entering or exiting the hive. Apiarists (beekeepers), normally split their native bee hives for propagating purposes in the spring (November) of each season. At this time, raw, unprocessed samples of native bee honey ($n = 20$) and wax ($n = 21$) were collected twice—once in November 2015 and then in November 2016. The native bee data were compared with co-located soil, dust and European honey bees samples, collected between November 2015 to April 2016 (Zhou et al., 2018).

Dust deposition samples gauges, were constructed and deployed in accordance with the Australian Standard 3580.10.1–2003 (Standards Australia, Dong and Taylor, 2017; Zhou et al., 2018). These were used to collect atmospheric dust samples monthly from November 2015 to April 2016, incorporating the most intensive foraging period for both the native and European honey bees. Surface (0–2 cm) soil samples ($n = 37$) were collected around each beehive in November 2015, and were dried at 40 °C for 48 h and sieved to < 2 mm for trace element analysis.

2.3. Sample and data analysis

Samples of native bees, honey, wax, soil and dust were analysed at the National Measurement Institute (NMI), using National Association of Testing Authorities accredited in-house reference methods for food, soil and dust. Raw, unprocessed native bees and honey samples were used for analysis. Wax samples were rinsed with Milli–Q™ (Milli–Q) water until all honey residues were removed and then oven dried at 60 °C. One decigram of native bees was digested with 1 ml HNO_3 (15.6 M) and 1 g honey and wax were digested with 3 ml HNO_3 before heating at 100 °C for 2 h.

Soil samples were digested by adding HCl (12.1 M) and HNO_3 (15.6 M) (3 ml and 3 ml, respectivelyl) followed by heating at 100 °C for 1 h. After cooling, 10 ml of Milli–Q water was added and the sample reheated to 100 °C for another hour. Dust deposition samples were collected on glass microfibre filters (Whatman 934-AH™, 47 mm diameter), and after weighing, digested using HCl (12.1 M)

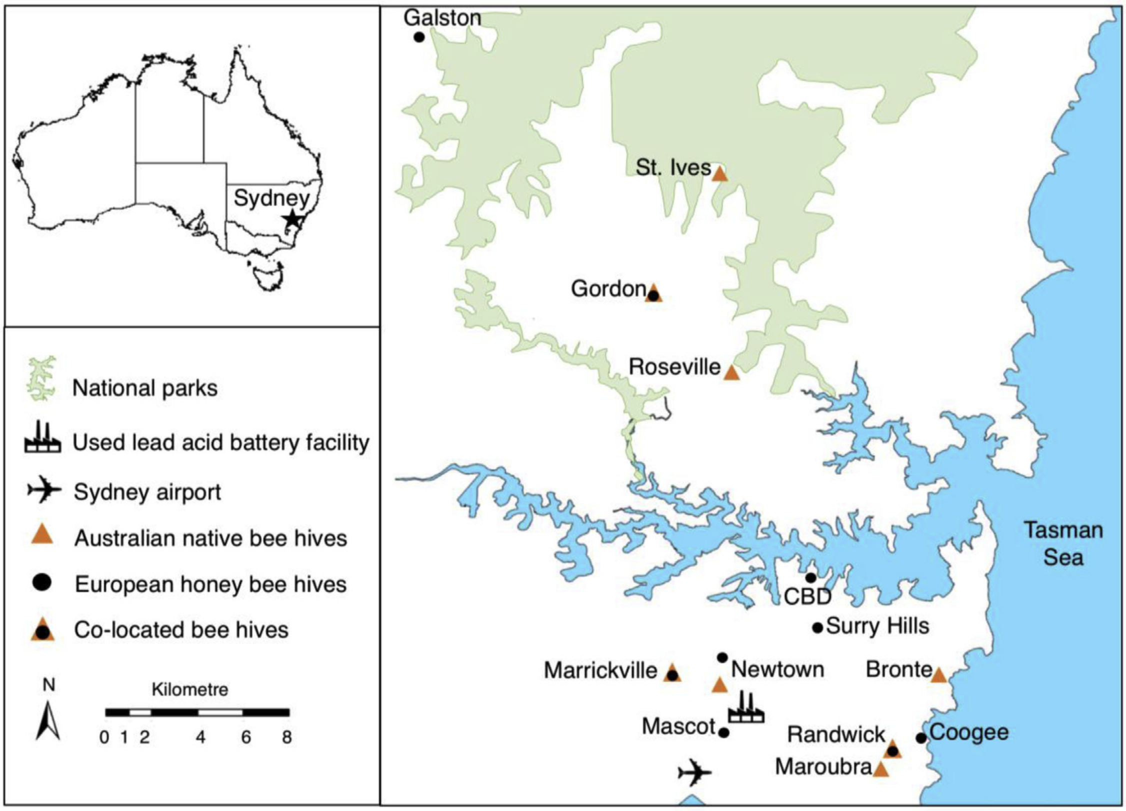

Fig. 1. Beehive locations of the Australian native bee hives (*T. carbonaria*) and European honey bee (*A. mellifera*) hives in Sydney, New South Wales (NSW). Locations of all the beehives range from sites in outer Sydney (Galston, St Ives, Gordon, Roseville), coastal Sydney (Bronte, Coogee, Randwick, Maroubra) and inner Sydney (Sydney Central Business District (CBD), Surry Hill, Newtown, Marrickville and Mascot).

and HNO_3 (15.6 M) (2 ml and 6 ml, respectively) for 2 h. Digested native bees were topped to 10 ml with Milli–Q water with the other samples (honey, wax, soil and dust) topped up to 40 ml. Samples were diluted (*2 for native bees, honey and wax; *5 for dust; *20 for soil) prior to analysis for As, Mn, Pb and Zn concentrations on an Inductively Coupled Plasma Mass Spectrometer (Agilent 7900 equipped with an Integrated Sample Introduction System). Each sample batch (n = 20) contained a laboratory reagent blank and duplicate, blank spike, blank matrix, duplicate sample and matrix spikes.

Procedural blanks were below NMI's Limit of Reporting (LOR) of 10 μg/kg for As, Mn, Pb, and Zn in honey bees, honey and wax. Dust sample blanks were < LOR of 0.1 mg/kg and 0.05 mg/kg for As and Zn, Pb and Mn, respectively. Procedural blanks for the soil samples were < LOR of 0.05 mg/kg for As, and 0.01 mg/kg for Pb, Mn and Zn. The procedural blanks (i.e. the filter material used in the dust analysis process and reagent solutions) were used to correct trace elements concentrations and Pb isotopic compositions.

The NMI's internal reference materials AGAL-10 (Hawkesbury River Sediment, n = 4) and AGAL-12 (bio-soil, n = 4) were processed with soil and dust samples. Elemental relative standard deviations (RSDs) for AGAL-10 and AGAL-12 were < 5.5% and < 4.8%, and mean recovery rates were 101% and 108%, respectively. Sample RSDs for As, Pb, Mn and Zn were < 1.8% for soil, < 4% for dust, < 6% for native bees and <4% for native bee honey and wax. Recovery rates of the AGAL-10 and AGAL-12 reference materials for As, Pb, Mn and Zn were 96–99% for soil, 98–100% for dust, and matrix spike recovery rates were 81–98% for native bees, 97–100% for honey, and 92–99% for wax.

Australian native bee samples (bees n = 5) and wax (n = 19) were subjected to Pb isotopic composition analysis ($^{204}Pb/^{207}Pb$, $^{206}Pb/^{207}Pb$, $^{208}Pb/^{207}Pb$), after optimising sample volumes based on their Pb concentrations (where > 1 μg/kg of Pb after digestion). The National Institute of Standards and Technology (NIST) SRM981 (natural Pb isotope composition standard) was used to correct for mass discrimination. Analytical uncertainties for Pb isotopic compositions (expressed for the NIST SRM981) were $^{204}Pb/^{206}Pb = 0.0590 \pm 0.0005$, $^{204}Pb/^{207}Pb = 0.065 \pm 0.0005$, $^{206}Pb/^{207}Pb = 1.093 \pm 0.005$, $^{208}Pb/^{207}Pb = 2.370 \pm 0.01$, respectively. The mean RSDs for NIST981 $^{204}Pb/^{207}Pb$, $^{206}Pb/^{207}Pb$, $^{208}Pb/^{207}Pb$ were 0.55%, 0.23%, 0.22%, respectively. The mean RSDs for sample analysis $^{204}Pb/^{207}Pb$, $^{206}Pb/^{207}Pb$, $^{208}Pb/^{207}Pb$ were 0.29%, 0.56% and 0.31%.

Data were analysed using GraphPad Prism 7.0 statistics software for arithmetic mean, standard deviation, minimum and maximum values and tests of significance (Mann–Whitney *U* test). Minitab was used for generating linear regressions and tests of significance (Pearson correlations). Concentrations of As, Mn, Pb and Zn in Australian native bees, honey and wax with < LOR (10 μg/kg) were treated as having a concentration of 5 μg/kg for statistical analysis purposes. Trace element concentrations in various beehive products and environmental samples of soil and dust were non-normally distributed and were $\log_{10}$ transformed for correlation analysis. Pearson correlation coefficients and two-tailed tests of significance were applied to assess the relationships between bee and hive products (native bees, honey, wax) and the environment (soil, dust) across the sampling sites. The data pertaining to European honey bees and their products is from our previous work (Zhou et al., 2018). These data were compared to the results of this study using the Mann–Whitney *U* test to compare concentrations of As, Mn, Pb and Zn in beehive samples between Australian native bees and European honey bees.

3. Results

3.1. Trace element concentrations in native bee samples and corresponding dust and soil

Arsenic and Pb concentrations in native bees, honey, wax, soils and dusts were greatest from the inner city Sydney metropolitan areas of Newtown and Marrickville and lowest at sites distant from Sydney CDB (St. Ives, Gordon and Roseville, Fig. 1). The concentration of As in all native bee honey samples was lower than the LOR of 10 μg/kg. Minimum As concentrations in native bees (21 μg/kg) were measured in Roseville samples, with the lowest concentrations of As in wax (12 μg/kg) and dust (2 mg/kg) being measured in samples from the northern Sydney areas of St. Ives and Gordon, respectively. Arsenic concentrations in wax and dust samples were typically more elevated in the Sydney city metropolitan city, with the notable exception of Roseville which recorded 102 mg/kg of As in dust (Supplementary Table S1).

Native bee honeys contained low levels of Pb with many being < 10 μg/kg, with the maximum concentration being measured in samples from the inner city suburb of Marrickville (34 μg/kg). This area is characterised with elevated soil lead levels (811 mg/kg, this study (Supplementary Table S1); and 689 mg/kg, Rouillon et al. (2017)). The lowest concentrations of Pb in native bees (24 μg/kg), wax (100 μg/kg) and dust (45 mg/kg) were found at Gordon, which were 85 times, 34 times, 16 times lower, respectively, than corresponding samples from the inner Sydney area of Newtown (Supplementary Table S1). The concentrations of Mn and Zn in native bee, honey or wax samples were not spatially consistent across the Sydney sites unlike corresponding concentrations of As and Pb. However, maximum values were recorded for Mn (46,400 μg/kg) and Zn (70,600 μg/kg) in native bees and honey (Mn 2700 μg/kg) from the inner Sydney areas of Newtown or Marrickville (Supplementary Table S1).

Lead concentrations in honey ($r = 0.814$, $p = 0.014$) and wax ($r = 0.883$, $p = 0.004$) were significantly related to its corresponding concentrations in dust, and Pb in honey was associated with its co-located soil ($r = 0.658$, $p = 0.076$) (Fig. 2a–c). The Zn concentrations in honey also significantly correlated to those in soil ($r = 0.709$, $p = 0.049$) (Fig. 2d). By comparison, no statistically significant relationships were evident in the data set for As or Mn concentrations in beehive products with respect to corresponding soil or dust samples ($r = -0.390$–0.501, $p = 0.206$–0.984) (Table S2).

3.2. Comparison of trace element concentrations in different bees and their products

Arsenic, Pb, Mn and Zn concentrations of Australian native bees, honey and wax products are presented in Supplementary Table S1. The concentrations measured in Australian native bee hives are compared below to data from co-located European honey bee hives, published in Zhou et al. (2018).

The data show that Australian native bees had significantly higher concentrations of As (median 72 μg/kg, range 21–140 μg/kg) than European honey bees (median 31 μg/kg, range 11–95 μg/kg) ($p < 0.001$) from all sites collected across Sydney (Fig. 3, Supplementary Table S3). By contrast, Mn and Zn were significantly lower in Australian native bees when compared to European honey

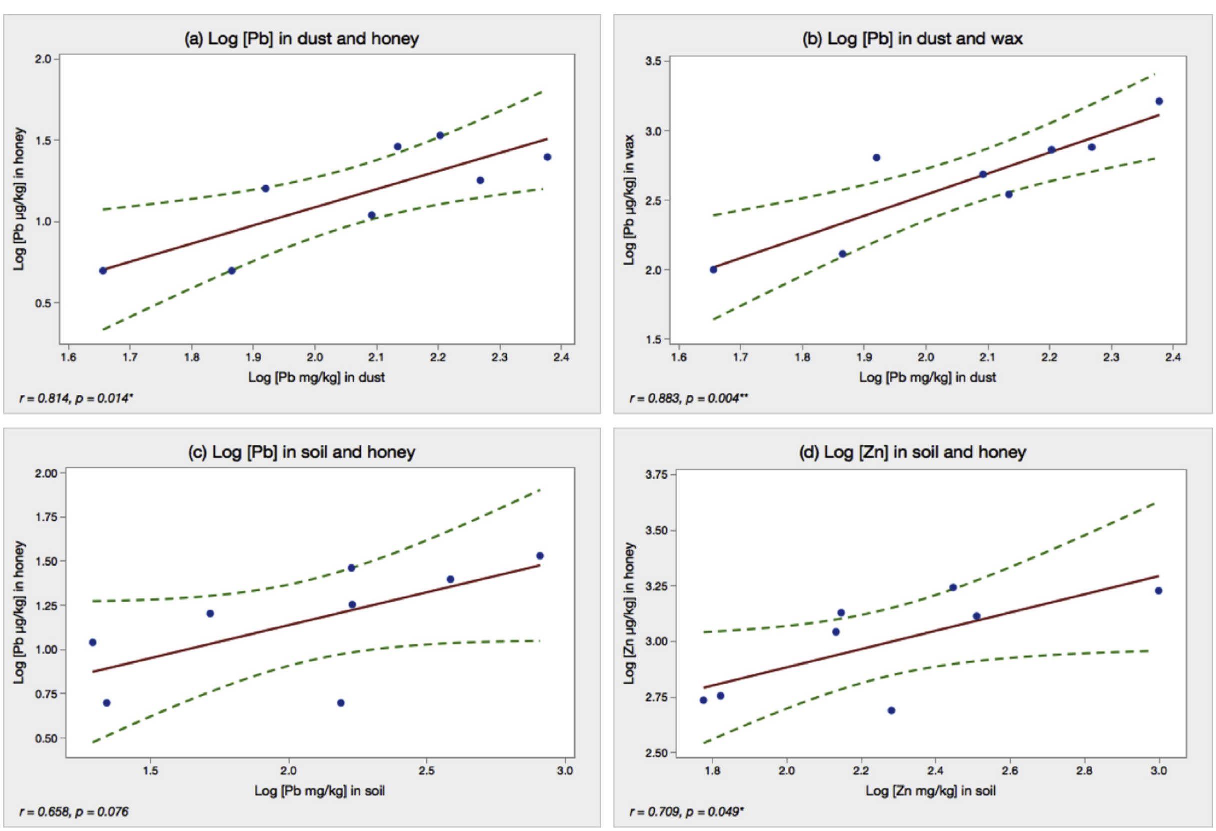

Fig. 2. Linear relationships with Pearson correlation coefficients (r) and *p* values between various Australian native bee products and soil or dust. Pearson correlation significance (*p*) at the following levels: * = 0.05 and ** = 0.01.

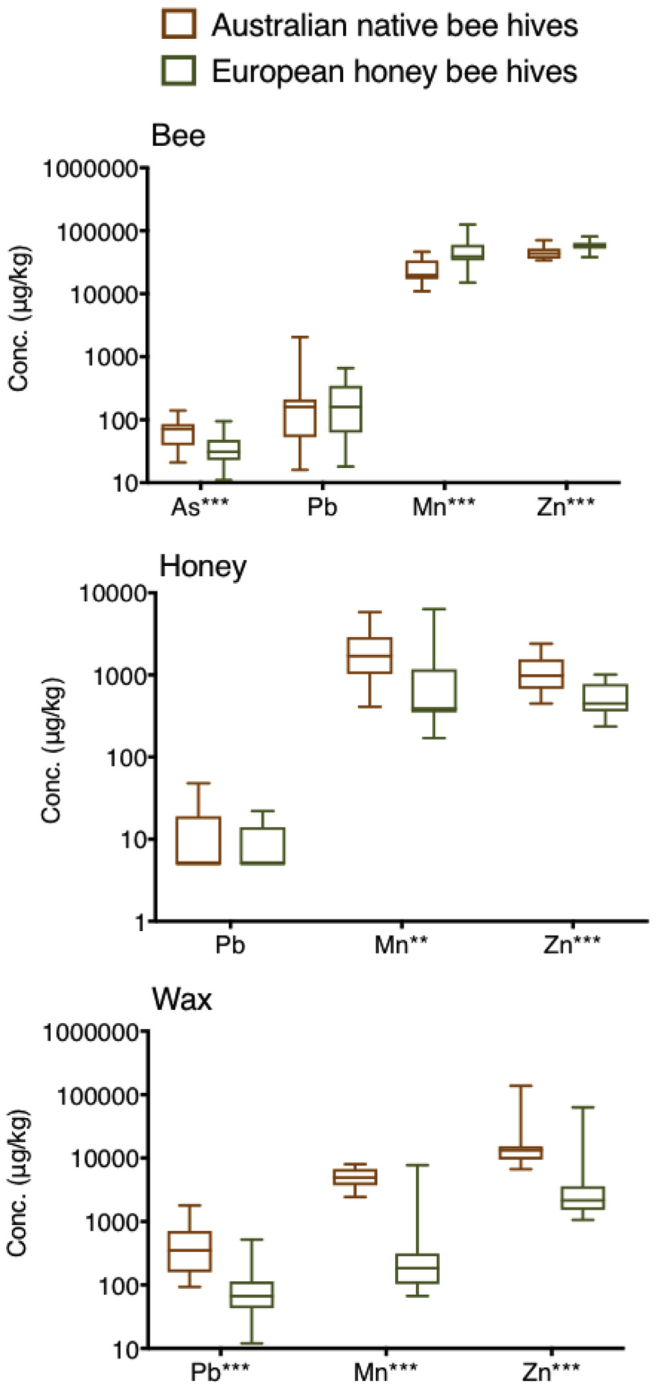

Fig. 3. Box-whisker plots for As, Pb, Mn and Zn concentrations (µg/kg) in native bees (n = 16), their honey (n = 20) and wax (n = 21) compared to European honey bees (n = 37), their honey (n = 15) and wax (n = 14). The *p* value significant at the following levels: *0.05, **0.01 and ***0.001.

bees (Mn $p < 0.001$, Zn $p < 0.001$). Trace element concentrations in the products of Australian native bees (honey and wax) were greater than those from European honey bees (Fig. 3): native bee honey contains more Mn (1700 (range: 410–5800) µg/kg vs. 390 (range: 170–6300) µg/kg, $p = 0.001$) and Zn (980 (range: 450–2400) µg/kg vs. 450 (range: 235–1010) µg/kg, $p < 0.001$) than that in honey produced by European honey bees (Supplementary Table S3). By contrast, Pb concentrations in native and European honey bees had identical median values of 160 µg/kg, with ranges of 16–2050 and 18–660 µg/kg, respectively (Fig. 3, Supplementary Table S3). Wax from native bee hives had significantly greater concentrations of Pb ($p < 0.001$), Mn ($p < 0.001$) and Zn ($p < 0.001$) than that from European honey bee hives.

3.3. Lead isotopic compositions in bees and their products

The mean values of Pb isotopic compositions ($^{206}Pb/^{207}Pb$, $^{208}Pb/^{207}Pb$) of Australian native bees and their products are plotted alongside co-located European honey bee data (Fig. 4). The Broken Hill ore body (Gulson, 1984; Kristensen and Taylor, 2016) is included as an ultimate end member, given that it was the primary source of Pb used in leaded petrol in Australia (Kristensen, 2015; Kristensen and Taylor, 2016). Lead isotopic compositions of Sydney basin sub-surface soils and rocks, along with aerosols (1979–2004) are from Wu et al. (2016b). Additional potential contaminant sources of environmental Pb are included in Fig. 4. These Pb isotopic composition data include Australian coal (Díaz-Somoano et al., 2009), industrial contaminants from fly ash and smelter fumes (Chiaradia et al., 1997) and Pb paints (Laidlaw et al., 2014; Rouillon, 2017).

Native bee samples (honey and wax) collected to the north of Sydney CBD close to national parks at St Ives, Gordon and Roseville had Pb isotopic compositions that could be differentiated from Sydney metropolitan samples (Table 1) and plotted either within or close to the envelope for natural Sydney Pb (Fig. 4). The mean Pb isotopic composition of native bee wax from Gordon ($^{206}Pb/^{207}Pb$ 1.120, $^{208}Pb/^{207}Pb$ 2.409) and Roseville ($^{206}Pb/^{207}Pb$ 1.115, $^{208}Pb/^{207}Pb$ 2.396) (Table 1) either overlap or are close to the Sydney surface soil and dust envelope determined in this study. Interestingly, the values also match the Pb isotopic compositions for surface soils ($^{206}Pb/^{207}Pb$ 1.098–1.121, $^{208}Pb/^{207}Pb$ 2.388–2.432) as measured by Wu et al. (2016b), which collectively implies that the natural composition of soils even at distance to the CBD have been adulterated by atmospheric depositions from leaded petrol (cf. Gulson et al., 1981).

The mean Pb isotopic compositions of Australian native bees and their wax ($^{206}Pb/^{207}Pb$ 1.072–1.121, $^{208}Pb/^{207}Pb$ 2.348–2.405) from Sydney metropolitan areas correspond closely to the more restricted range associated with aerosol values (1979–2004) compared to other possible sources of Pb contamination. Lead contamination from petrol consumption was dispersed widely across urban environments resulting in significant soil and dust contamination (e.g. Davis and Birch, 2010; Laidlaw et al., 2017; Rouillon et al., 2017). The atmospheric lead concentrations were collected in Sydney mainly during the period dominated by Pb-petrol use, which ended in 2002 (Gulson et al., 1985; Kristensen, 2015; Kristensen and Taylor, 2016; Wu et al., 2016b). The native bee data analysed here parallels that measured in European honey bees and their wax from hives across Sydney ($^{206}Pb/^{207}Pb$ 1.082–1.106, $^{208}Pb/^{207}Pb$ 2.352–2.388) (Zhou et al., 2018). Although the native bee data matches the envelope characterising the dominant historic source of atmospheric Pb contamination from leaded petrol (Fig. 4), it is notable that there is a variation of Pb isotopic compositions in native bees, wax and dust from each site (Table 1). This indicates that there are a range of Pb sources (e.g. paints (Laidlaw et al., 2014; Rouillon, 2017), industry emissions (Chiaradia et al., 1997) and coal (Díaz-Somoano et al., 2009)) present across the Sydney metropolitan area in addition to the main source of deposition from historic leaded petrol.

4. Discussion and conclusions

The median Pb concentration in *T. carbonaria* was the same as that in European honey bees (160 µg/kg) (Supplementary Table S3).

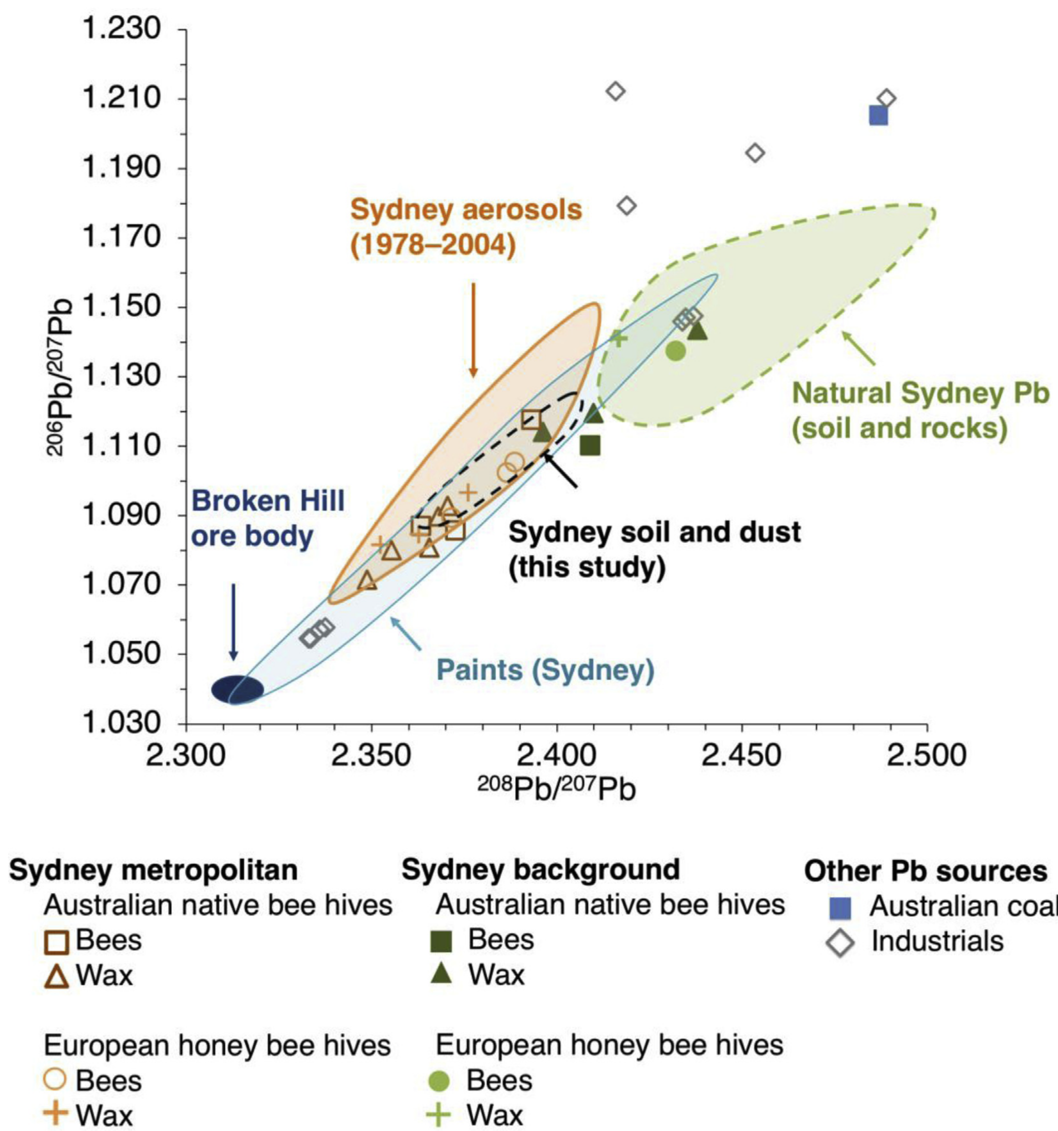

Fig. 4. Mean lead isotope compositions ($^{208}Pb/^{207}Pb$, $^{206}Pb/^{207}Pb$) for Australian native bee and European honey bee products across the Sydney region. Data comprises of Australian native bees (n = 5) and wax (n = 19) (Table 1); European honey bees (n = 15) and their wax (n = 3) (Zhou et al., 2018). The shaded areas represent characteristic signatures of: green – Sydney background (Wu et al., 2016b); orange – Sydney aerosols (Wu et al., 2016b); light blue – Sydney Pb-based paints (Laidlaw et al., 2014; Rouillon, 2017); dark blue – Broken Hill ore body (Gulson, 1984; Kristensen and Taylor, 2016). Coal and industrial data were obtained from Díaz-Somoano et al. (2009) and Chiaradia et al. (1997), respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

However, *T. carbonaria* showed a greater concentration range over equivalent sampling areas (16–2050 μg/kg vs. 18–660 μg/kg) (Fig. 3, Supplementary Table S3). This large variation in Pb concentrations in *T. carbonaria* may relate to its longer lifespan of *T. carbonaria* compared to that of European honey bees (100 days vs. 40 days) (Heard, 2016b), increasing exposure time and potential for uptake of environmental contaminants. The length of the native bees life cycle may also explain higher concentrations of As in *T. carbonaria* than those in European honey bees ($p < 0.001$) (Supplementary Table S3). However, essential elements of Mn ($p < 0.001$) and Zn ($p < 0.001$) had lower concentrations in *T. carbonaria* than European honey bees, which may be attributed to metabolic processes to support specific nutrient requirements (Ben-Shahar et al., 2004; Zhang et al., 2015) or its body size.

Although As concentrations were measurable in Australian native bees (median 72 μg/kg, range 21–140 μg/kg), concentrations of As in honey were below the laboratory reporting limit of 10 μg/kg (Supplementary Table S1). This finding is consistent with our recent investigation of European honey bees and their honey in Sydney (Zhou et al., 2018) as well as with other studies (Alvarez-Ayuso and Abad-Valle, 2017). Lower trace element concentrations in honey versus bees is thought to be due to metabolic 'filtering' and storage within the body of the bee (Leita et al., 1996; Fakhimzadeh and Lodenius, 2000; Porrini et al., 2002; Bogdanov et al., 2003; Bogdanov 2006; Ruschioni et al., 2013). Trace element contaminants are also excreted in bees faecal mass (Zhelyazkova et al., 2010).

Some trace element concentrations in honey (Mn $p = 0.01$, Zn $p < 0.001$) and wax (Pb $p < 0.001$, Mn $p < 0.001$ and Zn $p < 0.001$) from native bee products were significantly higher than those from European honey bees. Although not statistically significant ($p = 0.744$), median Pb concentrations in native bee honey (23 μg/kg) were greater than those in European honey bees (15 μg/kg) as were Zn concentrations in wax (13,200 μg/kg vs. 2150 μg/kg,

Table 1
Lead isotopic compositions ($^{206}Pb/^{204}Pb$, $^{206}Pb/^{207}Pb$, $^{208}Pb/^{207}Pb$) in native bees, wax, soil and dust across the Sydney metropolitan region, Australia. The range of isotopic composition measurements for different environmental media are provided, where available, in parentheses.

Sites	Sample	$^{206}Pb/^{204}Pb$	$^{206}Pb/^{207}Pb$	$^{208}Pb/^{207}Pb$
St. Ives	Native bees (n = 1)	17.435	1.111	2.408
	Wax (n = 1)	18.022	1.144	2.437
	Soil (n = 3)	17.127 ± 0.459	1.100 ± 0.019	2.378 ± 0.017
		(16.671–17.589)	(1.081–1.118)	(2.362–2.395)
	Dust (n = 4)	17.382 ± 0.357	1.118 ± 0.024	2.4398 ± 0.031
		(16.922–17.673)	(1.091–1.147)	(2.363–2.434)
Gordon	Wax (n = 1)	17.061	1.120	2.409
	Soil (n = 3)[a]	16.864 ± 0.585	1.095 ± 0.026	2.371 ± 0.029
		(16.525–17.540)	(1.077–1.125)	(2.350–2.404)
	Dust (n = 3) [a]	17.419 ± 0.160	1.122 ± 0.019	2.403 ± 0.019
		(17.314–17.602)	(1.111–1.143)	(2.389–2.424)
Roseville	Wax (n = 10)	17.282 ± 0.385	1.115 ± 0.017	2.396 ± 0.024
		(16.752–17.862)	(1.096–1.139)	(2.368–2.444)
	Soil (n = 7)	16.784 ± 0.405	1.087 ± 0.023	2.365 ± 0.023
		(16.280–17.366)	(1.064–1.119)	(2.340–2.401)
	Dust (n = 8)	16.980 ± 0.459	1.090 ± 0.026	2.368 ± 0.025
		(16.339–17.592)	(1.045–1.120)	(2.322–2.391)
Maroubra	Wax (n = 1)	16.549	1.081	2.355
	Soil (n = 2)	16.861	1.090	2.372
		(16.495–17.227)	(1.070–1.111)	(2.349–2.395)
	Dust (n = 5)	17.207 ± 0.105	1.106 ± 0.004	2.385 ± 0.012
		(17.072–17.315)	(1.099–1.110)	(2.363–2.393)
Bronte	Native bees (n = 2)	16.780	1.088	2.363
		(16.639–16.920)	(1.080–1.095)	(2.346–2.380)
	Wax (n = 1)	16.613	1.081	2.365
	Soil (n = 3)	17.359 ± 0.274	1.121 ± 0.007	2.405 ± 0.004
		(17.050–17.570)	(1.113–1.126)	(2.401–2.409)
	Dust (n = 5)	17.150 ± 0.243	1.104 ± 0.010	2.382 ± 0.011
		(16.863–17.505)	(1.090–1.114)	(2.371–2.392)
Randwick	Native bees (n = 1)	16.926	1.086	2.372
	Wax (n = 2)	16.728	1.090	2.367
		(16.715–16.741)	(1.089–1.091)	(2.364–2.371)
	Soil (n = 8) [a]	17.140 ± 0.190	1.102 ± 0.012	2.382 ± 0.015
		(16.887–17.524)	(1.090–1.129)	(2.366–2.412)
	Dust (n = 5) [a]	17.133 ± 0.199	1.109 ± 0.012	2.389 ± 0.010
		(16.851–17.358)	(1.093–1.120)	(2.372–2.398)
Newtown	Native bees (n = 1)	17.315	1.118	2.393
	Wax (n = 2)	16.539	1.072	2.348
		(16.344–16.734)	(1.063–1.081)	(2.340–2.357)
	Soil (n = 8) [a]	17.379 ± 0.373	1.116 ± 0.020	2.398 ± 0.022
		(16.872–17.901)	(1.089–1.142)	(2.369–2.427)
	Dust (n = 5) [a]	17.021 ± 0.218	1.099 ± 0.009	2.381 ± 0.018
		(16.692–17.222)	(1.1092–1.115)	(2.365–2.411)
Marrickville	Wax (n = 1)	16.923	1.093	2.370
	Soil (n = 7) [a]	17.107 ± 0.190	1.103 ± 0.010	2.385 ± 0.014
		(16.867–17.364)	(1.087–1.112)	(2.363–2.400)
	Dust (n = 4) [a]	17.248 ± 0.135	1.115 ± 0.004	2.401 ± 0.004
		(17.055–17.348)	(1.110–1.120)	(2.397–2.404)

Analytical uncertainties for Pb isotopic compositions were as follows: $^{204}Pb/^{206}Pb = 0.0590 \pm 0.0005$, $^{206}Pb/^{207}Pb = 1.093 \pm 0.005$, $^{208}Pb/^{207}Pb = 2.370 \pm 0.01$, respectively.
[a] Lead isotopic composition data from Zhou et al. (2018).

$p = 0.114$). Elevated trace element concentrations in native bee products is likely influenced by the significantly smaller amount of honey produced each year when compared to European honey bees (1 kg vs. 50 kg of honey) (Benecke, 2007; Heard, 2016c) and the smaller volume of wax produced per beehive box (8 L vs. 40 L) per year (Heard, 2016a).

Statistical analysis showed that Pb in honey ($p = 0.014$), wax ($p = 0.004$) and Zn in honey ($p = 0.049$) from native bees were strongly correlated to corresponding dust Pb or soil Pb/Zn concentrations, respectively (Fig. 2). This is not surprising because Pb depositions from petrol emissions blanketed Sydney during its use between 1932 and 2002 and remain persistent in the environment, including in European honey bees (Zhou et al., 2018). There are very limited current sources in the city of Sydney, with the largest single emitter being a used lead acid battery recycling facility (ULAB, Fig. 1) that released 400 kg of Pb to the atmosphere in 2015–2016 (NPI, 2017). By comparison, Sydney European honey bee data (Zhou et al. (2018) were more weakly associated to co-located soil (i.e. $p = 0.404$ vs 0.049 for honey Zn) and dust (i.e. $p = 0.340$ vs 0.014 for honey Pb) values than native bee samples (Fig. 2). The fact that Pb and Zn in Australian native bee samples are more strongly associated with surrounding dust and soil concentrations is possibly due to the markedly different life cycle and foraging behaviour of Australian native bees. Australian native bees live for ~100 days (cf. European honey bees' lifespan of ~40 days) and their foraging distance is much smaller at 0.3–0.7 km vs. 9 km in European honey bees. The strong association between Australian native bees (and their products) to corresponding environmental dust Pb and soil Zn levels indicate that they are more sensitive bio-indicators than European honey bees.

European honey bees and Australian native bees and their wax from sites in inner Sydney corresponded closely to the Pb isotopic compositions in historic aerosols (1978–2004) sampled when the use of leaded petrol began to decline (Kristensen, 2015) (Fig. 4).

Lead isotopic compositions of Australian native bees show that historic leaded petrol depositions remain a persistent contaminant in biological samples, as confirmed in a number of other local (Wu et al., 2016a, 2016b) and international studies (e.g. Farmer et al., 2002; Flegal et al., 2010; Odigie and Flegal, 2011; Hansson et al., 2017). Aside of leaded petrol depositions, other Pb sources including Pb-based paint (Laidlaw et al., 2014; Rouillon et al., 2017), traffic emissions (Birch and Scollen, 2003), coal combustion (Díaz-Somoano et al., 2009; Cohen et al., 2012) and industrial sources (Cohen, 1999; Cohen et al., 2002; Davis and Gulson, 2005; Gulson et al., 2007; Marx et al., 2010; Wu et al., 2016b) may also be present in the environment. Indeed, the variation of Pb isotopic compositions at individual sample sites suggests that a range of Pb sources persist in the Sydney environment (Table 1).

Overall, this study indicates that Australian native bees have clear potential to be used as a bio-indicator species to evaluate environmental trace element concentrations and Pb sources at small scales (up to 0.7 km) over different land use types (cf. Holt and Miller, 2011). For example, data from native bee hives could be used to supplement standard soil sampling investigations as part of rezoning and development approval processes, which often result in the reuse of former industrial sites as residential land or public open space. The data suggest that Australian native bees are at least equivalent to those of European honey bees as a proxy environmental marker and for certain media (e.g. dust and wax) may be even more sensitive. The increased interest in and keeping of all bee types in Australia and elsewhere presents an opportunity to supplement traditional aerosol and dust deposition sampling that typically relies on a limited number of monitors to characterise air quality. Given the persistence of legacy contaminants in the environment (e.g. Pb) and the emergence of new compounds of concern including those listed on the Stockholm Convention (United Nations, 2015), bees and their products provide an obvious choice for measuring the prevalence, distribution and recycling of toxic pollutants in food and ecological systems.

Acknowledgements

Special thanks to Peter Clark (Ku-ring-gai Council), Phil Wick, Gavin Smith, Derek Spielman, Michel Kettler, and Ariane Wicks, for providing access to their Australian native bee hives in Sydney. We thanks Chenyin Dong (Macquarie University) for sampling assistance, and Dr Marek Rouillon (Macquarie University) for comments on a previous draft of this manuscript. We acknowledge the assistance of Dr Shiva Prasad, Dr Andrew Evans, Dr Michael Wu, Dr Ping Di and Lisa Liu of the National Measurement Institute, North Ryde, Sydney, for their considerable help with sample processing and analytical interpretation. Professor Brian Gulson is thanked for his earlier comments relating to Pb isotopes in European honey bees and their products. The Ku-ring-gai Council is thanked for small grant support via its community grant Program (2015 Environmental Small Grants Scheme), field assistance and transport support. Xiaoteng Zhou is funded by an International Macquarie University Research Excellence Scholarship (iMQRES No. 2014157).

Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.envpol.2018.06.063.

References

Alvarez-Ayuso, E., Abad-Valle, P., 2017. Trace element levels in an area impacted by old mining operations and their relationship with beehive products. Sci. Total Environ. 599, 671–678.
Beekman, M., Ratnieks, F.L.W., 2000. Long-range foraging by the honey-bee, *Apis mellifera L.* Funct. Ecol. 14, 490–496.
Ben-Shahar, Y., Dudek, N.L., Robinson, G.E., 2004. Phenotypic deconstruction reveals involvement of manganese transporter malvolio in honey bee division of labor. J. Exp. Biol. 207 (19), 3281–3288.
Benecke, F.S., 2007. Commercial Beekeeping in Australia. Australian Government - Rural Industries Research and Development Corporation, Canberra.
Birch, G.F., Scollen, A., 2003. Heavy metals in road dust, gully pots and parkland soils in a highly urbanised sub-catchment of Port Jackson, Australia. Aust. J. Soil Res. 41, 1329–1342.
Birch, G.F., Vanderhayden, M., Olmos, M., 2011. The nature and distribution of metals in soils of the Sydney estuary catchment, Australia. Water, Air, Soil Pollut. 216 (1–4), 581–604.
Bogdanov, S., 2006. Contaminants of bee products. Apidologie 37 (1), 1–18.
Bogdanov, S., Imdorf, A., Charriere, J., Fluri, P., Kilchenmann, V., 2003. The Contaminants of the Bee Colony. Swiss Bee Research Centre, Bern.
Bromenshenk, J., Gudatis, J., Carlson, S., Thomas, J., Simmons, M., 1991. Population dynamics of honey bee nucleus colonies exposed to industrial pollutants. Apidologie 22 (4), 359–369.
Chiaradia, M., Chenhall, B.E., Depers, A.M., Gulson, B.L., Jones, B.G., 1997. Identification of historical lead sources in roof dusts and recent lake sediments from an industrialized area: indications from lead isotopes. Sci. Total Environ. 205 (2–3), 107–128.
Cohen, D.D., 1999. Seasonal and regional variations in ambient fine particle concentrations and sources in New South Wales, Australia: a seven year study. In: International Conference on Urban Climate, pp. 8–12. Sydney.
Cohen, D.D., Crawford, J., Stelcer, E., Atanacio, A.J., 2012. Application of positive matrix factorization, multi-linear engine and back trajectory techniques to the quantification of coal-fired power station pollution in metropolitan Sydney. Atmos. Environ. 61, 204–211.
Cohen, D.D., Stelcer, E., Garton, D., 2002. Ion beam methods to determine trace heavy metals concentrations and sources in urban airsheds. Nucl. Instrum. Meth. Phys. Res. B 190, 466–470.
Conti, M.E., Botre, F., 2001. Honeybees and their products as potential bioindicators of heavy metals contamination. Environ. Monit. Assess. 69 (3), 267–282.
Davis, B.S., Birch, G.F., 2010. Spatial distribution of bulk atmospheric deposition of heavy metals in metropolitan Sydney, Australia. Water, Air, Soil Pollut. 214 (1–4), 147–162.
Davis, J.J., Gulson, B.L., 2005. Ceiling (attic) dust: a "museum" of contamination and potential hazard. Environ. Res. 99 (2), 177–194.
Díaz-Somoano, M., Kylander, M.E., López-Antón, M.A., Suárez-Ruiz, I., Martínez-Tarazona, M.R., Ferrat, M., Kober, B., Weiss, D.J., 2009. Stable lead isotope compositions in selected coals from around the world and implications for present day aerosol source tracing. Environ. Sci. Technol. 43 (4), 1078–1085.
Dong, C., Taylor, M.P., 2017. Applying geochemical signatures of atmospheric dust to distinguish current mine emissions from legacy sources. Atmos. Environ. 161, 82–89.
Dong, C., Taylor, M.P., Kristensen, L.J., Zahran, S., 2015. Environmental contamination in an Australian mining community and potential influences on early childhood health and behavioural outcomes. Environ. Pollut. 207, 345–356.
Fakhimzadeh, K., Lodenius, M., 2000. Honey, pollen and bees as indicator of heavy metal pollution. Acta Univ. Carol. Environ. 14, 13–20.
Farmer, J.G., Eades, L.J., Atkins, H., Chamberlain, D.F., 2002. Historical trends in the lead isotopic composition of archival *Sphagnum* mosses from scotland (1838–2000). Environ. Sci. Technol. 36 (2), 152–157.
Flegal, A.R., Gallon, C., Hibdon, S., Kuspa, Z.E., Laporte, L.F., 2010. Declining—but persistent—atmospheric contamination in central California from the resuspension of historic leaded gasoline emissions as recorded in the lace lichen (*Ramalina menziesii* taylor) from 1892 to 2006. Environ. Sci. Technol. 44 (14), 5613–5618.
Gulson, B., Korsch, M., Dickson, B., Cohen, D., Mizon, K., Davis, J.M., 2007. Comparison of lead isotopes with source apportionment models, including SOM, for air particulates. Sci. Total Environ. 381 (1–3), 169–179.
Gulson, B., Mizon, K., Taylor, A., Korsch, M., Davis, J.M., Louie, H., Wu, M., Gomez, L., Antin, L., 2014. Pathways of Pb and Mn observed in a 5-year longitudinal investigation in young children and environmental measures from an urban setting. Environ. Pollut. 191, 38–49.
Gulson, B., Mizon, K., Taylor, A., Korsch, M., Stauber, J., Davis, J.M., Louie, H., Wu, M., Antin, L., 2008. Longitudinal monitoring of selected elements in blood of healthy young children. J. Trace Elem. Med. Biol. 22 (3), 206–214.
Gulson, B., Mizon, K., Taylor, A., Korsch, M., Stauber, J., Davis, J.M., Louie, H., Wu, M., Swan, H., 2006. Changes in manganese and lead in the environment and young children associated with the introduction of methylcyclopentadienyl manganese tricarbonyl in gasoline—preliminary results. Environ. Res. 100 (1), 100–114.
Gulson, B.L., 1984. Uranium-lead and lead-lead investigations of minerals from the Broken Hill lodes and mine sequence rocks. Econ. Geol. 79 (3), 476–490.
Gulson, B.L., Davis, J.J., Bawden-Smith, J., 1995. Paint as a source of recontamination of houses in urban environments and its role in maintaining elevated blood leads in children. Sci. Total Environ. 164, 221–235.
Gulson, B.L., Porritt, P.M., Mizon, K.J., Barnes, R.G., 1985. Lead isotope signatures of stratiform and strata-bound mineralization in the broken Hill block, New South Wales, Australia. Econ. Geol. 80 (2), 488–496.
Gulson, B.L., Tiller, K.G., Mizon, K.J., Merry, R.H., 1981. Use of lead isotopes in soils to identify the source of lead contamination near Adelaide, South Australia. Environ. Sci. Technol. 15 (6), 691–696.

Halcroft, M.T., Spooner-Hart, R., Haigh, A.M., Heard, T.A., Dollin, A., 2015. The Australian stingless bee industry: a follow-up survey, one decade on. J. Apic. Res. 52 (2), 1–7.
Hansson, S.V., Claustres, A., Probst, A., De Vleeschouwer, F., Baron, S., Galop, D., Mazier, F., Le Roux, G., 2017. Atmospheric and terrigenous metal accumulation over 3000 years in a French mountain catchment: local vs distal influences. Anthropocene 19, 45–54.
Heard, T.A., 1999. The role of stingless bees in crop pollination. Annu. Rev. Entomol. 44, 183–206.
Heard, T.A., 2000. What are stingless bees, and why and how to use them as crop pollinators? a review. JARQ 34 (3), 183–190.
Heard, T.A., 2016a. Honey from stingless bee hives. In: Craig, C. (Ed.), The Australian Native Bee Book. Sugarbag Bees, Brisbane, pp. 158–177.
Heard, T.A., 2016b. Understanding the highly social bees. In: Craig, C. (Ed.), The Australian Native Bee Book. Sugarbag Bees, Brisbane, pp. 24–37.
Heard, T.A., 2016c. Constructing hive boxes for stingless bees. In: Graig, G. (Ed.), The Australian Native Bee Book. Sugarbag Bees, Brisbane, pp. 119–131.
Heard, T.A., Dollin, A.E., 2015. Stingless bee keeping in Australia: snapshot of an infant industry. Bee World 81 (3), 116–125.
Holt, E.A., Miller, S.W., 2011. Bioindicators: Using Organisms to Measure Environmental Impacts. Nature Education Knowledge. Retrieved June 10, 2017, from https://www.nature.com/scitable/knowledge/library/bioindicators-using-organisms-to-measure-environmental-impacts-16821310.
Huber, M., Welker, A., Helmreich, B., 2016. Critical review of heavy metal pollution of traffic area runoff: occurrence, influencing factors, and partitioning. Sci. Total Environ. 541, 895–919.
Islam, M.S., Ahmed, M.K., Raknuzzaman, M., Habibullah -Al- Mamun, M., Islam, M.K., 2015. Heavy metal pollution in surface water and sediment: a preliminary assessment of an urban river in a developing country. Ecol. Indicat. 48, 282–291.
Jones, K.C., 1987. Honey as an indicator of heavy metal contamination. Water, Air, Soil Pollut. 33 (1–2), 179–189.
Kristensen, L.J., 2015. Quantification of atmospheric lead emissions from 70 years of leaded petrol consumption in Australia. Atmos. Environ. 111, 195–201.
Kristensen, L.J., Taylor, M.P., 2016. Unravelling a ‘miner's myth’ that environmental contamination in mining towns is naturally occurring. Environ. Geochem. Health 38 (4), 1015–1027.
Kristensen, L.J., Taylor, M.P., Flegal, A.R., 2017. An odyssey of environmental pollution: the rise, fall and remobilisation of industrial lead in Australia. Appl. Geochem. 83, 3–13.
Kristensen, L.J., Taylor, M.P., Odigie, K.O., Hibdon, S.A., Flegal, A.R., 2014. Lead isotopic compositions of ash sourced from Australian bushfires. Environ. Pollut. 190, 159–165.
Laidlaw, M.A.S., Mohmmad, S.M., Gulson, B.L., Taylor, M.P., Kristensen, L.J., Birch, G., 2017. Estimates of potential childhood lead exposure from contaminated soil using the US EPA IEUBK Model in Sydney, Australia. Environ. Res. 156, 781–790.
Laidlaw, M.A.S., Taylor, M.P., 2011. Potential for childhood lead poisoning in the inner cities of Australia due to exposure to lead in soil dust. Environ. Pollut. 159 (1), 1–9.
Laidlaw, M.A.S., Zahran, S., Pingitore, N., Clague, J., Devlin, G., Taylor, M.P., 2014. Identification of lead sources in residential environments: Sydney Australia. Environ. Pollut. 184, 238–246.
Leita, L., Muhlbachova, G., Cesco, S., Barbattini, R., Mondini, C., 1996. Investigation of the use of honey bees and honey bee products to assess heavy metals contamination. Environ. Monit. Assess. 43 (1), 1–9.
Li, X., Poon, C.-S., Liu, P.S., 2001. Heavy metal contamination of urban soils and street dusts in Hong Kong. Appl. Geochem. 16, 1361–1368.
Lopes, I., Sedlmayr, A., Moreira-Santos, M., Moreno-Garrido, I., Blasco, J., Ribeiro, R., 2010. European bee-eater (*Merops apiaster*) populations under arsenic and metal stress: evaluation of exposure at a mining site. Environ. Monit. Assess. 161 (1), 237–245.
Marx, S.K., Kamber, B.S., McGowan, H.A., Zawadzki, A., 2010. Atmospheric pollutants in alpine peat bogs record a detailed chronology of industrial and agricultural development on the Australian continent. Environ. Pollut. 158 (5), 1615–1628.
Matin, G., Kargar, N., Buyukisik, H.B., 2016. Bio-monitoring of cadmium, lead, arsenic and mercury in industrial districts of Izmir, Turkey by using honey bees, propolis and pine tree leaves. Ecol. Eng. 90, 331–335.
National Pollutant Inventory 2015-2016, 2017. Australian Government. Department of Environment and Energy, Canberra.
Norgate, M., Boyd-Gerny, S., Simonov, V., Rosa, M.G., Heard, T.A., Dyer, A.G., 2010. Ambient temperature influences Australian native stingless bee (Trigona carbonaria) preference for warm nectar. PLoS One 5 (8), e12000.
Odigie, K.O., Flegal, A.R., 2011. Pyrogenic remobilization of historic industrial lead depositions. Environ. Sci. Technol. 45 (15), 6290–6295.
Perugini, M., Manera, M., Grotta, L., Abete, M.C., Tarasco, R., Amorena, M., 2011. Heavy metal (Hg, Cr, Cd, and Pb) contamination in urban areas and wildlife reserves: honeybees as bioindicators. Biol. Trace Elem. Res. 140 (2), 170–176.
Porrini, C., Ghini, S., Girotti, S., Sabatini, A.G., Gattavecchia, E., Celli, G., 2002. Use of honey bees as bioindicators of environmental pollution in Italy. In: Devillers, J., Pham-Delegue, M.-H. (Eds.), Honey Bees: Estimating the Environmental Impact of Chemicals. Taylor & Francis, New York, pp. 186–248.
Rodriguez Martin, J.A., De Arana, C., Ramos-Miras, J.J., Gil, C., Boluda, R., 2015. Impact of 70 years urban growth associated with heavy metal pollution. Environ. Pollut. 196, 156–163.
Rouillon, M., 2017. Enhancing the Application of Field Portable X-ray Fluorescence Technology for the Measurement of Metal Contaminated Soils. Doctor of Philosophy. Macquarie University.
Rouillon, M., Harvey, P.J., Kristensen, L.J., George, S.G., Taylor, M.P., 2017. VegeSafe: a community science program measuring soil-metal contamination, evaluating risk and providing advice for safe gardening. Environ. Pollut. 222, 557–566.
Ruschioni, S., Riolo, P., Minuz, R.L., Stefano, M., Cannella, M., Porrini, C., Isidoro, N., 2013. Biomonitoring with honeybees of heavy metals and pesticides in nature reserves of the Marche Region (Italy). Biol. Trace Elem. Res. 154 (2), 226–233.
Satta, A., Verdinelli, M., Ruiu, L., Buffa, F., Salis, S., Sassu, A., Floris, I., 2012. Combination of beehive matrices analysis and ant biodiversity to study heavy metal pollution impact in a post-mining area (Sardinia, Italy). Environ. Sci. Pollut. Res. 19 (9), 3977–3788.
Saunier, J.B., Losfeld, G., Freydier, R., Grison, C., 2013. Trace elements biomonitoring in a historical mining district (les Malines, France). Chemosphere 93 (9), 2016–2023.
Schwarz, K., Pickett, S.T., Lathrop, R.G., Weathers, K.C., Pouyat, R.V., Cadenasso, M.L., 2012. The effects of the urban built environment on the spatial distribution of lead in residential soils. Environ. Pollut. 163, 32–39.
Serrano, O., Davis, G., Lavery, P.S., Duarte, C.M., Martinez-Cortizas, A., Mateo, M.A., Masqué, P., Arias-Ortiz, A., Rozaimi, M., Kendrick, G.A., 2016. Reconstruction of centennial-scale fluxes of chemical elements in the Australian coastal environment using seagrass archives. Sci. Total Environ. 541, 883–894.
Siegel, F.R., 2002. Environmental Geochemistry of Potentially Toxic Metals. Springer, New York, U.S.A.
Smith, J.P., Heard, T.A., Beekman, M., Gloag, R., 2017. Flight range of the Australian stingless bee *Tetragonula carbonaria* (Hymenoptera: Apidae). Aust. Entomol. 56 (1), 50–53.
Standards Australia. Australian/New Zealand 3580.10.1:2003. Determination of particulate matter-Deposited matter-Gravimetric method. Retrieved November 26, 2016, from http://infostore.saiglobal.com/store/details.aspx?ProductID.373364.
Su, C., Jiang, L., Zhang, W., 2014. A review on heavy metal contamination in the soil worldwide: situation, impact and remediation techniques. Environ. Skep. Crit. 3 (2), 24–38.
Trujillo-Gonzalez, J.M., Torres-Mora, M.A., Keesstra, S., Brevik, E.C., Jimenez-Ballesta, R., 2016. Heavy metal accumulation related to population density in road dust samples taken from urban sites under different land uses. Sci. Total Environ. 553, 636–642.
Turer, D., Maynard, J.B., Sansalone, J.J., 2001. Heavy metal contamination in soils of urban highways: comparison between runoff and soil concentrations at Cincinnati, Ohio. Water, Air, Soil Pollut. 132, 293–314.
United Nations, 2015. Stockholm Convention on Persistent Organic Pollutants. Stockholm.
Wu, L., Taylor, M.P., Handley, H.K., 2017. Remobilisation of industrial lead depositions in ash during Australian wildfires. Sci. Total Environ. 599–600, 1233–1240.
Wu, L., Taylor, M.P., Handley, H.K., Gulson, B.L., 2016a. Insights into past atmospheric lead emissions using lead concentrations and isotopic compositions in historic lichens and fungi (1852–2008) from central and southern Victoria, Australia. Atmos. Environ. 139, 46–55.
Wu, L., Taylor, M.P., Handley, H.K., Wu, M., 2016b. Australian atmospheric lead deposition reconstructed using lead concentrations and isotopic compositions of archival lichen and fungi. Environ. Pollut. 208, 678–687.
Zarić, N., Ilijević, K., Stanisavljević, L., Gržetić, I., 2016. Metal concentrations around thermal power plants, rural and urban areas using honeybees (Apis mellifera L.) as bioindicators. Int. J. Environ. Sci. Technol. 13 (2), 413–422.
Zhang, G., Zhang, W., Cui, X., Xu, B., 2015. Zinc nutrition increases the antioxidant defenses of honey bees. Entomol. Exp. Appl. 156 (3), 201–210.
Zhang, H., Luo, Y., Makino, T., Wu, L., Nanzyo, M., 2013. The heavy metal partition in size-fractions of the fine particles in agricultural soils contaminated by waste water and smelter dust. J. Hazard Mater. 248–249, 303–312.
Zhelyazkova, I., Atanasova, S., Barakova, G., Mihaylova, G., 2010. Content of heavy metals and metalloids in bees and bee products from areas with different degree of anthropogenic impact. Agric. Sci. Technol. 3 (1), 136–142.
Zhou, X., Taylor, M.P., Davies, P.J., Prasad, S., 2018. Identifying sources of environmental contamination in European honey bees (*Apis mellifera*) using trace elements and lead isotopic compositions. Environ. Sci. Technol. 52 (3), 991–1001.