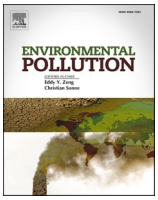
Environmental Pollution 335 (2023) 122257

Contents lists available at ScienceDirect

Environmental Pollution

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



Tracing nickel smelter emissions using European honey bees☆

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ARTICLE INFO

Keywords:
Dust
Nickel smelting
Honey
Honey bee
Nickel
Trace elements
Soil

ABSTRACT

This study investigated trace element contamination in honey bees inhabiting urban areas around the South Pacific's largest and longest operating nickel smelter in Nouméa, New Caledonia. There remains a paucity of research on the environmental impact of nickel smelting, and to date, there has been no assessment of its effects on the popular practice of beekeeping, or whether honey bees are a suitable tracer for nickel smelting emissions. Honey bees and honey were sampled from 15 hives across Nouméa to ascertain linkages between nickel smelter emissions, environmental contamination, and trace element uptake by bees. Comparison of washed and unwashed bees revealed no significant difference in trace element concentrations, indicating trace elements bioaccumulate within the internal tissues of bees over time. Accordingly, trace element concentrations were higher in dead bees than those that were sampled live, with smelter related elements chromium, cobalt and nickel being significantly different at $p < 0.05$. Except for boron, trace element concentrations were consistently higher in bees than in honey, suggesting that the transfer of trace elements from bees during honey production is negligible. Elevated concentrations of potentially toxic trace elements including cobalt, chromium and nickel in bees declined with distance from smelting operations (Spearman's Rho, $p < 0.05$), indicating the relationship between environmental contamination and the uptake of trace elements by bees. The findings of this study emphasise potential environmental and human health risks associated with trace element contamination from nickel smelting operations and affirm the use of honey bees as a biomonitor of potentially harmful nickel smelting emissions.

1. Introduction

Assessing the source and risk of harm from potentially toxic trace elements remains an ongoing challenge. Acceptable levels of exposure are constantly being lowered in response to mounting evidence that there is no lower safe threshold of exposure for many chemicals (Lanphear, 2017). Concerns are typically more acute where there is an intersection between contamination, urban environments and food production (Leake et al., 2009). In this study, we examine the trace element relationships between smelter emissions and the popular practice of beekeeping in the French territory and city of Nouméa, New Caledonia, which is home to the South Pacific's largest, and longest operating, nickel (Ni) smelter.

Globally, trace element contamination has been greatly reduced since the latter part of the 20th century due to more effective pollution emission control and stringent environmental guidelines (Ericson et al., 2021). Nevertheless, environmental pollution remains a significant and ongoing health risk, being responsible for approximately 1 in 6 deaths, and totalling 9 million global deaths per year (Fuller et al., 2022). Localised contamination of urban and natural environments remains a challenge due to economically necessary industrial activities that produce significant environmental contamination, even under approved licence conditions (cf. Taylor et al. (2014)). In this regard, New Caledonia is no exception, especially given that it is host to ~10% of the

☆ This paper has been recommended for acceptance by Philip N. Smith.

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https://doi.org/10.1016/j.envpol.2023.122257
Received 22 May 2023; Received in revised form 8 July 2023; Accepted 24 July 2023
Available online 26 July 2023

world's nickel reserves (Grandcolas et al., 2008; Losfeld et al., 2015) and is responsible for ~8.2% of the world's production of mined Ni (Plaza-Toledo, 2018).

New Caledonia's laterite soils are formed from igneous ultrabasic rocks that are naturally rich in the trace elements cobalt (Co), chromium (Cr), manganese (Mn) and Ni (Becquer et al., 2006). As a result, it is not surprising that a study of adults and children across the New Caledonian archipelago by St-Jean et al. (2018) showed that urine concentrations in areas with the naturally occurring laterite soil exceeded international human reference values for elements Cr and Ni, and to a lesser degree, Co and Mn. Children were most affected by Ni and Cr exposures, with 13% exceeding the reference value for Ni and 90% for Cr. Soil was considered a significant source of exposure, especially for children, who have greater hand to mouth exposure, lower body weight, are lower to the ground and have an overall greater body burden (Agency for Toxic Substances and Disease Registry, 2022).

Chronic lifetime exposure to potentially toxic trace elements is particularly important in the context of New Caledonia and French Polynesia because they have some of highest globally reported thyroid cancer incidence and mortality rates (Ferlay et al., 2012; International Agency for Research on Cancer, 2023; Tervonen et al., 2017). While the specific causal elements are not well known, environmental (Baumann et al., 2011; Luce et al., 2000) and occupational sources (Menvielle et al., 2003) have been identified as potential human health risk factors in New Caledonia. Lifetime chronic exposure to trace elements in the environment, naturally or anthropogenically enriched, is a hazard that warrants concern for human health outcomes.

The country's capital city, Nouméa (population ~100,000), is home to the major Ni smelting Doniambo facility that has been operating since 1910 (Mudd, 2010; Mudd & Jowitt, 2014). The complex is located ~2 km from the Nouméa city centre and is within ~500 m of residential homes. Air quality studies by the New Caledonian Air Quality Monitoring Association (Scal Air, 2018) have demonstrated that the primary source of dust contamination (in particular Ni, cadmium (Cd), zinc (Zn)) originates from the Doniambo complex and that Ni values frequently exceed acceptable environmental standards for deposited dust (Scal Air, 2018). Further, a recent assessment of urban soils, indoor and outdoor dusts showed that not only was the smelter site a primary source of Ni and Cr, but that potential exposures presented a human health risk to Nouméa residents (Fry et al., 2021). These findings are of concern because Ni and its compounds are classified as human carcinogens and the inhalation of Ni is associated with lung and nasal cancers (National Toxicology Program, 2021). In addition, Ni refinery dust also includes Ni_3S_2 (nickel subsulfide) that is classified as a human and environmental carcinogen (Agency for Toxic Substances and Disease Registry, 2005; Australian Government, 2022a, 2022b).

Despite there being 76 smelters world-wide (Nickel Institute, 2023), of which three are in New Caledonia (Doniambo, Goro Nickel, and Koniambo), there is a paucity of research examining the environmental impacts of Ni smelting. Nickel smelting and recycling of Ni-rich products will increase markedly in the coming decades. It is a critical metal in new energy technologies and is estimated to increase five-fold to 48 Mt by 2050 (Bruce et al., 2021). Moreover, Ni can be recycled indefinitely without loss of quality, contributing to a genuine circular economy (Nickel Institute, 2023).

While several studies have demonstrated the efficacy of using European honey bees (*Apis mellifera*) to assess spatial and temporal changes in organic and inorganic trace contaminants (Cunningham et al., 2022), no other studies have specifically addressed the impact of Ni smelting. Here, we examine the efficacy of the commensal European honey bee as a sentinel environmental biomarker (Taylor, 2019) of human exposure to Ni smelter related emissions alongside natural and urban anthropogenic trace element sources. Honey bees are an efficient biomonitoring species for airborne transmitted contaminants due to their interaction with different environmental matrices during foraging, their small range, and short lifespan (Taylor, 2019). Due to the popularity of urban beekeeping (Egerer & Kowarik, 2020) hives in locations of relevance to human and environmental health can be examined, and citizen scientist beekeepers can take part in the sampling. Previous studies have shown that trace element concentrations in European honey bees are reflective of local urban use sources (Goretti et al., 2020; Smith et al., 2019; Zhou et al., 2018b) and have identified that bee trace elements correspond to local mining activities (Satta et al., 2012; Zhou et al., 2018b).

In this first New Caledonian honey bee biomarker study of anthropogenic Ni smelter-related contaminants, we address the following research questions:

(1) How do trace element concentrations in honey bees and honey relate to natural, urban and smelter sources of contamination?
(2) Where do trace elements accumulate within honey bees and how does that inform their use for contaminant biomonitoring?
(3) Do trace element concentrations in honey bees reflect potential environmental health risks and are there any identifiable harms related to the consumption of bee products?

2. Methods and approach

Identification of locations and volunteer beekeepers to participate in sampling was facilitated by the Centre d'Apiculture, Technopole de Nouvelle-Calédonie (ADECAL), who have a database of registered European honey beekeepers across New Caledonia. In addition, ADECAL manages sentinel hives that are used for the early detection of exotic diseases and honey bee[1] pests. Three sentinel hives were included in the study since they were purposely installed close to the Noumea smelter facility's harbour. Beekeepers and their hives were selected for participation to provide spatial coverage across the urban area of Nouméa and characterise the range and spread of trace elements with respect to the Doniambo smelter facility and city centre, which are proximally located (Fig. 1). At each hive, dead bees, live bees and honey were sampled as described below.

2.1. Honey bee and honey sampling

Bee and honey samples were collected from 15 hives (one hive per apiary) in Nouméa over a 1-month period in July 2019 while the smelter was operational. In addition, a single dead queen bee that was in front of its hive was collected and analysed as per the method for the other bee samples. In total, the study was comprised of 37 pooled bee samples from 15 study hives plus a single queen bee. Each pooled sample contained an average of ~30 bees, hereafter referred to as 'samples' for simplicity, totalling 1143 individual bees (live bees n = 538; dead bees n = 352; washed bees n = 252 (see below for explanation on washed bees); queen bee n = 1). In addition, single honey samples were collected from Nouméa study hives (n = 15) and 5 apiaries from across greater New Caledonia (Fig. 1). The number of bees used for the analysis of live, dead, washed and unwashed comparisons are detailed in Supplementary Table S1. The five honey samples from across New Caledonia and its islands were provided by ADECAL for comparative purposes to characterise concentrations more broadly (Fig. 1).

Samples of live bees and corresponding honey were collected from each hive once during the study period. To do this, a frame was lifted from the hive and approximately 30 live bees were collected directly into a sterile 50 mL centrifuge test tube. Live pooled bee samples from n = 15 hives were stored at −20 °C to enable rapid immobilisation and euthanasia (Human et al., 2013) before freeze drying using a Christ Alpha 1–2 LDPLUS instrument for 72-h and homogenising using a sterile stainless steel spatula. At the same time, honey samples (n = 15) were

[1] Unless stated, the term "honey bees" and "bees" are used synonymously; i.e. they both refer to European honey bees, *Apis mellifera*, the species analysed in this study.

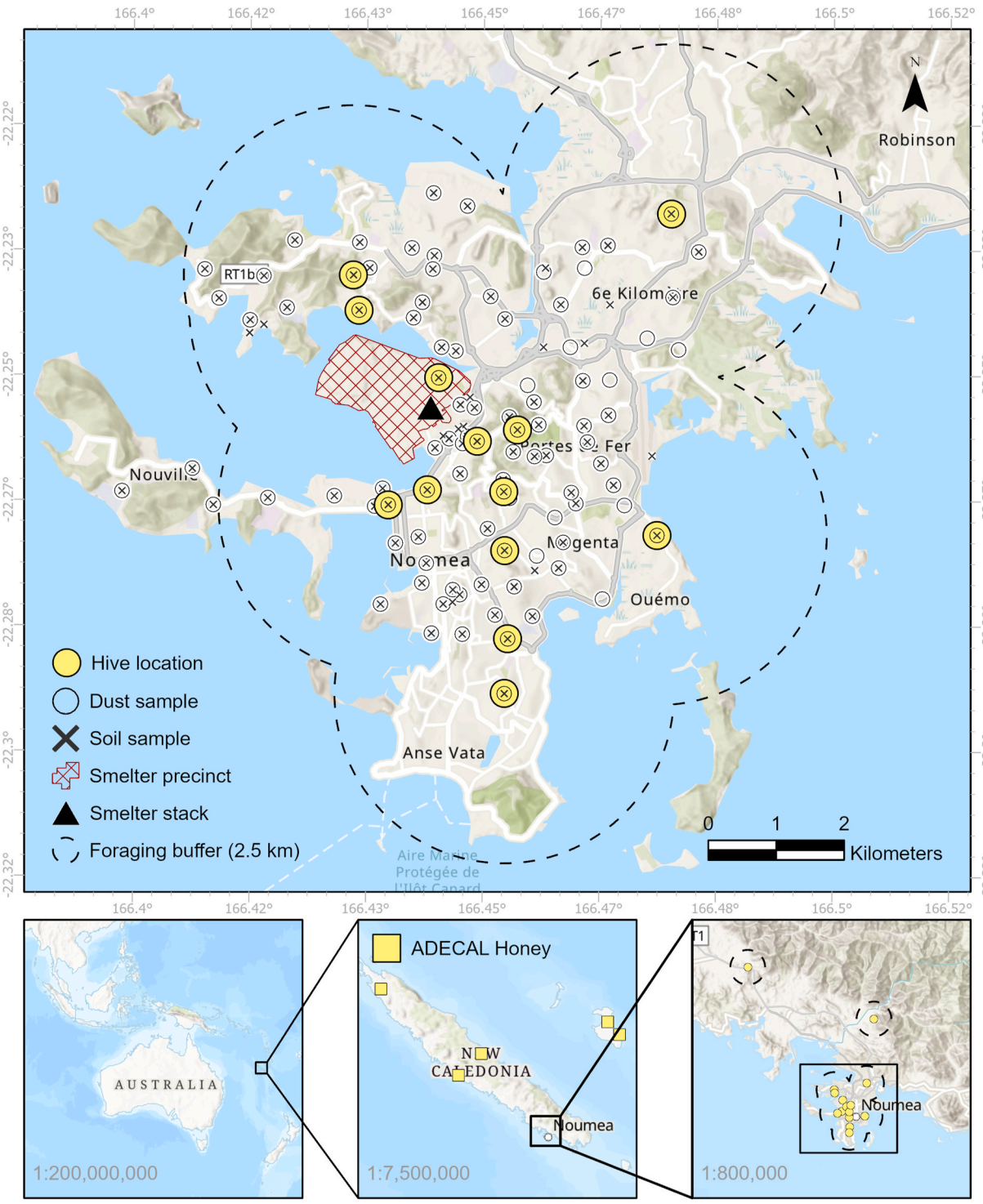

Fig. 1. Location of study hives (n = 15) and honey sample sites in Nouméa (n = 15) and across New Caledonia (n = 5). The ADECAL (Centre d'Apiculture, Technopole de Nouvelle-Calédonie) honey samples (n = 5; bottom centre panel) were collected to characterise trace elements in honey from across the country. Also shown is a 2.5 km bee foraging buffer around each of the sampled hives in Nouméa. Marked soil and dust sampling locations are detailed in Fry et al. (2021).

collected from the frame by scraping a sterile 50 mL centrifuge tube along the honey cells, allowing raw honey to drip directly into the sampling container. Honey samples were transported on ice and stored at 4 °C prior to transport to Australia for trace element analysis. In addition, dead bee samples (n = 14) were collected weekly from each hive on a pre-cleaned plastic surface that was installed at the base of the hive. Dead bees were pooled for each hive, enabling matched live and dead bee samples for 14 of the 15 hives, with one hive not sampled for dead bees due to it being included in the study towards the end of field work period. Dead bees were processed in the same manner as the live bees (Supplementary Fig. S2). The process for preparing honey bees for export to Australia, including the legal requirement for sample storage and transport is provided in Supplementary Text S3.

2.2. Dead and live honey bee analyses

This research inquiry included assessment of both live and dead bees to better understand whether trace elements accumulated in bees over their lifetime. To understand if contaminants were accumulating on the surface of the bee or in the tissue and organs of the bee, where sufficient live bees were sampled from a hive (n = 8 hives), samples were split and one half was subject to a wash treatment prior to freeze drying, homogenising and analysis. The wash treatment consisted of the following protocol: (a) bees were soaked for 2 min in 30 mL of MilliQ water in a 50 mL centrifuge test tube; (b) the sealed tubes were rotated gently on their vertical axis 6 times by hand and the MilliQ supernatant decanted. (c) Steps (a) and (b) were repeated once. (d) An additional 30 mL of MilliQ water was added, and without further soaking, samples were rotated 6 times with the supernatant decanted on completion.

2.3. Trace element analysis

Honey bee and honey samples were analysed for 21 acid digestible trace elements at the National Measurement Institute (NMI): aluminium (Al), barium (Ba), boron (B), calcium (Ca), chromium (Cr), cobalt (Co), copper (Cu), iron (Fe), lead (Pb), magnesium (Mg), manganese (Mn), nickel (Ni), phosphorus (P), potassium (K), rubidium (Rb), sodium (Na), strontium (Sr), sulfur (S), tin (Sn), titanium (Ti) and zinc (Zn). Trace elements were analysed using either an Agilent Varian 730-ES or an Agilent 7900 quadrupole inductively coupled plasma mass spectrometer (Q-ICP-MS) equipped with an ISIS sample introduction system and an ORS helium collision cell, depending on the elements analysed.

The honey bee and honey samples were processed using established methods (National Measurement Institute, 2022). In summary, 1 g sample from each pooled sample of homogenised bees was digested in a 3 mL nitric acid and 1 mL hydrochloric acid solution and heated on a hot block at 95–100 °C for 2 h. Following digestion, samples were made up to a volume of 40 mL with high purity water, then diluted as required prior to analysis. The NMI's analytical quality assurance of honey bee and honey analyses are reported in full in Supplementary Tables S4a–b. Limits of reporting (LoR) varied between elements, ranging from 0.01 to 1.0 mg/kg, with blanks less than the LoR for all elements (Supplementary Tables S4a–b). Sample duplicate analyses for the bees (n = 2) and honey (n = 1) returned a mean relative percent difference (RPD) of 10% (0–20.9%) and 3% (0–23.2%), respectively. A single laboratory control sample (n = 1) was analysed and returned a mean recovery of 97% of the reference value (range: 87–106%). Matrix spike analyses for the honey bees (n = 2) and honey (n = 1) returned a mean recovery of 102% (range: 92–120%) and 101% (range: 81–123%), respectively.

Honey bee and honey data were compared to corresponding soil and dust wipes collected at each hive along with samples from the surrounding environment as reported by Fry et al. (2021). This dataset consisted of 134 dust wipes (n = 26, April 2019; n = 108, July 2019), urban roadside soil (n = 91), garden soil (n = 15), smelter slag (n = 4), background rock (n = 12) and soil (n = 12) collected in July 2019. These data were used to evaluate trace element gradients in honey bees across Nouméa with respect to other, more traditional matrices used for smelter contamination-related assessments.

2.4. Scanning electron microscopy of honey bees

To further examine whether contaminants were present on or in the bees, the exterior surface of dead bee specimens were visualised using scanning electron microscopy (SEM). Individual honey bees were collected in 50 mL centrifuge tubes and stored at 4 °C prior to analysis. Specimens (n = 4) were carbon coated (~10 nm; Carbon Coater ACE 600, Leica Microsystems) and observed using a JEOL scanning electron microscope (SEM) under low vacuum mode (JSM IT-300) using a secondary electron detector with an acceleration voltage of 20 kV. The integrated energy dispersive X-ray (EDX) analyser was used to identify the elemental composition of particles identified on the surface of bees.

2.5. Lead isotopic composition analyses

To better understand the linkage and pathways between environmental trace element sources from anthropogenic and natural inputs, Pb isotopic compositions (PbIC; $^{204}Pb/^{207}Pb$, $^{206}Pb/^{207}Pb$, $^{208}Pb/^{207}Pb$) in honey bees were measured with the NMI's Agilent 7900 Q-ICP-MS using established methods (National Measurement Institute, 2021; Zhou, Taylor, Davies, et al., 2018). Seven pooled bee samples totalling 259 individual bees were assessed for their PbIC. Five of the pooled samples consisted of dead bees (n = 182 bees); one of the pooled samples was washed live bees (n = 36 bees) and the other was unwashed live bees (n = 41 bees; details provided in Supplementary Table S1).

Bee sample digest solutions were diluted as required to bring the Pb concentration between 1 and 40 ppb and analysed with samples bracketed by concentration matched NIST SRM 981 (certified Pb isotopic standard). Raw counts were corrected for blanks and potential Hg (mercury) isobaric interference, and the NIST SRM981 was used to bracket concentration matched samples and correct for mass discrimination and instrument drift. National Measurement Institute method validation data analytical uncertainties (expressed for NIST SRM 981) are as follows: $^{204}Pb/^{207}Pb = 0.0645 \pm 0.0005$, $^{206}Pb/^{207}Pb = 1.093 \pm 0.005$, $^{208}Pb/^{207}Pb = 2.370 \pm 0.01$. The standard reference material NIST 2709a was analysed with every analytical run as an independent control and all results ($^{204}Pb/^{207}Pb = 0.0638$ (±0.0006), $^{206}Pb/^{207}Pb = 1.218$ (±0.006) and $^{208}Pb/^{207}Pb = 2.486$ (±0.010) were within published reference value ranges (Aung et al., 2004; Reimann et al., 2011; Souto-Oliveira et al., 2019; Takaoka et al., 2006; Unruh et al., 2000). Lead concentrations were too low to determine the PbIC in honey samples, hence only data for bee samples were obtained. The study data were compared to published values for Nouméa indoor and outdoor dust, smelter slag, soils and background samples (Fry et al., 2021).

2.6. Data analysis

Sample data were analysed using a combination of Microsoft® Excel for Mac v. 16.70, ArcGIS Pro 10.0.2 and Python 3.9.13. Graphs and figures were compiled using ArcGIS Pro 10.0.2, Python 3.9.13 and Affinity Designer 1.10.4.1198. Spearman's rank correlation coefficient and the Mann-Whitney *U* test were used to identify statistically significant associations and differences between the biochemical and geochemical data.

3. Results and discussion

Analysis of trace element concentrations in honey bees and honey from the 15 hives is presented below. Study data are supplemented with honey samples from across New Caledonia (Fig. 1) along with Nouméa soil and dust deposition data analysed previously by the authors (Fry et al., 2021) to evaluate the nexus between ambient values and those measured in co-located honey bees and honey. Although the study sample matrices were analysed for 21 trace elements, the primary focus presented here is Ni, given this is the element of concern in relation to the Doniambo smelter facility. Summary data for all trace elements in honey and bees are provided in Supplementary Table S5.

3.1. Trace elements in Noumea dust and soil

In terms of ambient dust deposition, Fry et al. (2021) showed that near smelter-related values of Cr, Fe, Mn, Ni, S declined markedly beyond 3 km, though only the decline of Ni was statistically significant (Spearman's Rho, $p < 0.001$; $r_s = -0.43$). This was most probably due to the effects of particle deposition and accumulation in the environment from contemporary smelter emissions coupled with >100 years of operation. Fry et al. (2021) also showed that mean dust wipe element

concentrations in Nouméa were elevated relative to background locations, particularly for Pb (2.8 ×), Ni (2.6 ×) and Cr (2.0 ×).

With respect to roadside soils, Fe, Mn and Ni concentrations declined significantly (Spearmans Rho, Fry et al. (2021)) with distance from the smelter, with Ni showing the strongest relationship ($p < 0.001$; $r_s = -0.37$), versus Mn ($p = 0.017$; $r_s = -0.25$) and Fe ($p = -0.037$; $r_s = -0.22$). Common anthropogenic elements (e.g., Pb, Zn) and those associated with the smelter operations (Fry et al. (2021) - Cr, Fe, Mn, Ni, S) were more elevated in roadside and garden soils compared to background values (Supplementary Fig. S6).

Overall, Ni was the only element to have a statistically significant decline in both dust loading values and roadside soils with respect to the distance to the Doniambo smelter (Supplementary Table S9 in Fry et al. (2021)), making this a critical element for biomarker assessment in corresponding honey bee and honey samples.

3.2. Trace elements in bees and honey

In terms of honey bees and honey data, aside from boron (B), all elements were enriched in the bees versus the honey (Fig. 2; Supplementary Table S5). In particular the bee/honey ratio (live and dead bees combined) showed that Ni in bees was 31 × more enriched than in honey. These data show that while honey bees absorb trace elements from the environment, most elements are not transferred in the same quantity to the honey they produce (Smith & Weis, 2020; Smith et al., 2019; Zhou, Taylor, & Davies, 2018; Zhou et al., 2018a,b). Indeed, for the majority of the Nouméa honey samples, Al, Co, Cr, Pb, Sn and Ti concentrations were below limits of reporting (LoR, Supplementary Table S5). Further, the international foods standard CODEX CXS 12–1981 Standard for Honey (Food and Agriculture Organization of the United Nations and World Health Organization, 2019) does not set maximum residue limits or heavy metal (trace element) concentrations, rather it states: "*Honey shall be free from heavy metals in amounts which may represent a hazard to human health*" (Food and Agriculture Organization of the United Nations and World Health Organization, 2019). Concentrations of smelter-related elements of Cr and Ni are more than an order of magnitude less in honey than what is present in the bees (Fig. 2), suggesting that the exposure risk to potentially harmful trace elements via honey is minimal.

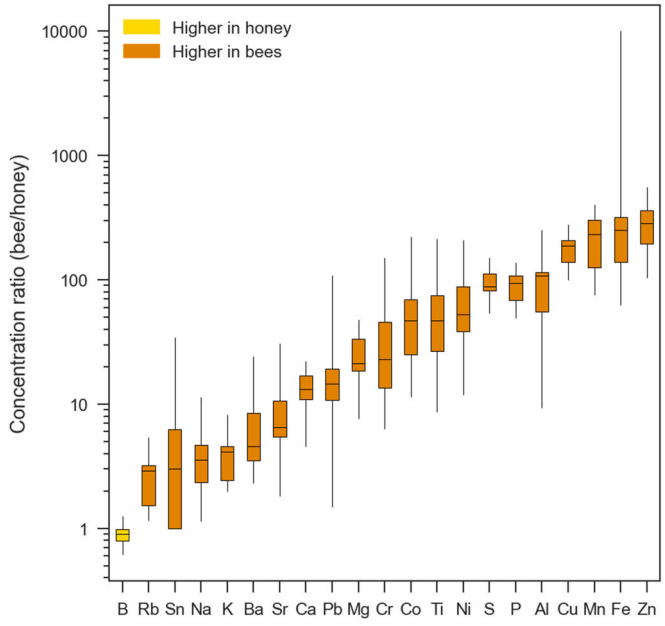

Fig. 2. Trace element concentration enrichment in honey bees (live and dead bee samples compared to honey samples from corresponding hives. Boxplots show the minimum, 25th percentile, median, 75th percentile and maximum values.

Honey samples from across the main island of New Caledonia (n = 5) were also collected and analysed to compare to those produced in Nouméa. Mann-Whitney *U* tests comparing trace element concentrations revealed that the two sets of honeys were largely similar with only B being significantly higher in Nouméa honey, and K being significantly higher in honey from across the island. The primary element of interest in this study, Ni, returned a p-value of 0.05, having slightly higher concentrations in the Nouméa honey: 0.17 mg/kg versus 0.12 mg/kg in the honey samples collected from across the main island of New Caledonia. Despite the small sample size, the data indicate that the trace element distribution in honey produced in Nouméa was not materially different to that produced across the wider islands of New Caledonia.

3.3. Spatial distributions in honey and honey bee data

Trace element concentrations for live and dead bees from the 15 study hives (n = 29) and corresponding honey samples (n = 15) were assessed with respect to distance from the Doniambo smelter using a Spearman Rho correlation to ascertain their potential as a biomarker of emission sources (Supplementary Table S7). To allow comparison of honey bee and honey data to other environmental matrices, soil and dust values published by Fry et al. (2021) within a 2.5 km radius of each hive were aggregated. While a 2.5 km radius is representative of the mean honey bee foraging radius (Visscher & Seeley, 1982), some of the area in Nouméa is sea so was not included in the foraging area estimate. Sample data includes one mean garden soil value (determined from n = 5 soil samples per garden; Fry et al. (2021)) and one dust sample collected at the hive plus any other samples of those media collected within that radius. While there is some overlap of the mean foraging areas (Fig. 1), Visscher and Seeley (1982) showed many visits were much shorter than 2.5 km from the hive, depending on the quality of the different food sources available. Observations in Nouméa during sampling indicated a plentiful supply of seasonal flowers, facilitated by the tropical climate.

Previous work by Free et al. (1983) identified that analysis of pollen collected at different hives could be a proxy method for assessment of possible soil heavy metal contamination across large areas. While many studies of bees have examined them for Pb contamination, or have targeted urban pollution sources (e.g. Smith and Weis (2020); Zarić et al. (2017); Zhou, Taylor, Davies, et al. (2018); Zhou et al. (2018a,b)) none have examined the impact of Ni smelting emissions on environmental quality using honey bees as a biomonitor.

As shown in Fig. 3, it is evident that the smelter-related elements Co, Cr, Ni in combined live and dead bee samples display significant distant decay relationships ($p < 0.05$) with respect to the smelter (Supplementary Table S7), similar to that identified from a study of locally-sourced lichens (Pasquet et al., 2016). This also reflects the findings of Fry et al. (2021), who observed similar relationships in soil and dust, but for which Ni was the only element that retained statistical significance across matrices. The clear trends depicted here using a suite of 21 trace elements suggests that honey bees are a more sensitive and informative biomarker than traditional matrices like soil and dust.

Trace elements Al, Rb and Ti are predominantly geogenic in origin and were found at higher concentrations in honey bees closer to the smelter facility. This may reflect their increased deposition and availability in the ambient environment due to ore processing and smelter emissions (Supplementary Table S7). Higher concentrations of Sn closer to the smelter is also likely reflective of Sn concentrations present in the New Caledonian Ni-laterite deposits (Ulrich et al., 2019) that are processed at the Doniambo facility.

A significant decrease in environmental concentrations of Pb in honey bees with distance from the urbanised area of Nouméa is consistent with environmental Pb emissions and depositions (Kristensen, 2015; Kristensen et al., 2017) from automotive use of Pb additives in French gasoline (Fry et al., 2021; Monna et al., 1997). Further, declines in Pb concentrations in honey bees (Smith & Weis, 2020; Smith

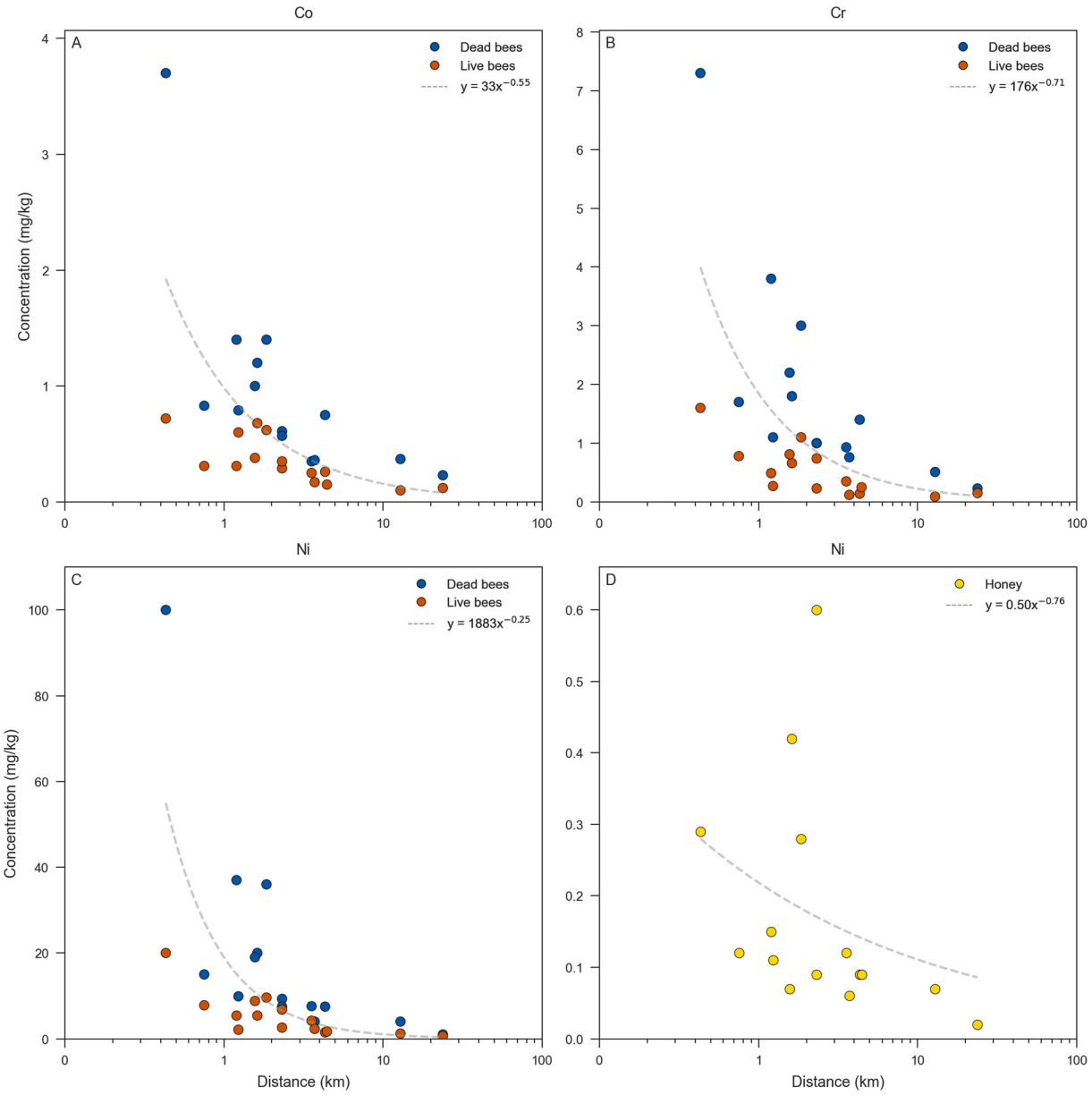

Fig. 3. Relationship between smelter related trace element concentrations (mg/kg) of cobalt (Co), chromium (Cr), and nickel (Ni) in honey bees (A–C) relative to distance from the Doniambo smelter stack (km). Trace element concentrations from dead and live bees are distinguished for observation, but the power regression model is calculated using all data points. Also shown are corresponding hive honey Ni concentrations (D). With the exception of two honey samples for Co and one for Cr, concentration values for these smelter elements were below the LoR (Supplementary Table S5).

et al., 2019; Zhou et al., 2018b) and native bees (Zhou et al., 2018a) away from other major urban areas has been identified elsewhere, indicating their utility as a biomarker of contamination as a result of the former use of leaded gasoline and lead-based paints.

Other studies of trace elements in honey bees have also revealed decreased concentrations of common anthropogenic elements (As, Pb, V (vanadium), Zn with distance from urbanised and industrial activities (e.g. (Smith & Weis, 2020; Zarić et al., 2017; Zhou et al., 2018a,b)). The smaller size and more recent industrialisation of Nouméa is reflected in the lower average concentrations of Pb in honey bees (mean 0.08 mg/kg), which were similar to locations adjacent to national parks (<0.056 mg/kg) in Australia's largest city, Sydney (Zhou et al., 2018b). In that city's CBD and inner city areas, honey bee Pb concentrations were several fold higher (0.15–0.44 mg/kg; Zhou et al. (2018b)) than corresponding samples in Nouméa. Nouméa smelter elements Co, Cr and Ni are seldom reported in honey bee studies, but the effect of emissions is evident by comparison to data from two cities in Serbia (Zarić et al., 2017). Here, average honey bee Co, Cr and Ni were 0.11 mg/kg, 0.18 mg/kg and 0.48 mg/kg, respectively, versus those in Nouméa bees: 0.35 mg/kg, 0.52 mg/kg and 5.33 mg/kg, for Co, Cr and Ni, respectively (Supplementary Table S5).

In contrast to honey bees, honey samples (n = 15) had low trace element concentrations, with the majority of the smelter-related elements (Co, Cr) having concentrations < LoR (<0.01 mg/kg and <0.05 mg/kg, respectively, Supplementary Table S5). In terms of distance from the smelter, only honey Ni concentrations were statistically significant ($p = 0.014$, $r_s = -0.619$), indicating, as per dust and soil, that Ni is the key element released from smelter activities.

Overall, the spatial distribution, and remobilisation of Ni along with other potentially toxic smelter-related elements into bees indicates that the greatest risk of exposure are for honey bee communities within ~2 km of Doniambo facility. This finding is consistent with other global

smelter-impacted locations (Ettler, 2016) and a recent assessment of health risk modelling using soil and dust samples from across the Nouméa urban area (Fry et al., 2021).

3.4. Lead isotopic signatures - markers of contaminant pathways in honey bees

Lead isotopic compositions (PbIC) of honey bees were measured to evaluate potential sources and pathways of exposure from environmental matrices. Fry et al. (2021) measured PbIC in dust wipes, slag, subsurface soil, topsoil and vacuum dust to determine sources of Pb with respect to smelter emissions, French gasoline (Monna et al., 1997), the Koum-Borindi Massif and the Saint Louis Massif Ni ore deposits (Cluzel et al., 2005). The distinctive low radiogenic PbIC Broken Hill ore signature was included in Fig. 4 given its use as an automotive gasoline additive in neighbouring Australia (Kristensen & Taylor, 2016), and also in French gasoline (Véron et al., 1999).

The data show that the PbIC in both live and dead bees corresponded closely to deposited ambient dust, which was also similar to the signatures measured in homes. The PbIC signatures in honey bees (Supplementary Table S8) did not directly match French gasoline, background (subsurface) soils or smelter sources and slags, but instead reflected a composite of these sources (Fig. 4), as was also identified in Nouméa indoor and outdoor dust (Fry et al., 2021). From this data and previous works (Fry et al., 2021) it is evident that deposited dust is being remobilised within the environment and interacting with biota including honey bees (Zhou et al., 2018b). This confirms both persistence and mobility of smelter-derived contamination, as well as the utility of applying honey bees as a biomonitor of environmental pollution (Smith & Weis, 2020; Smith et al., 2019; Taylor, 2019).

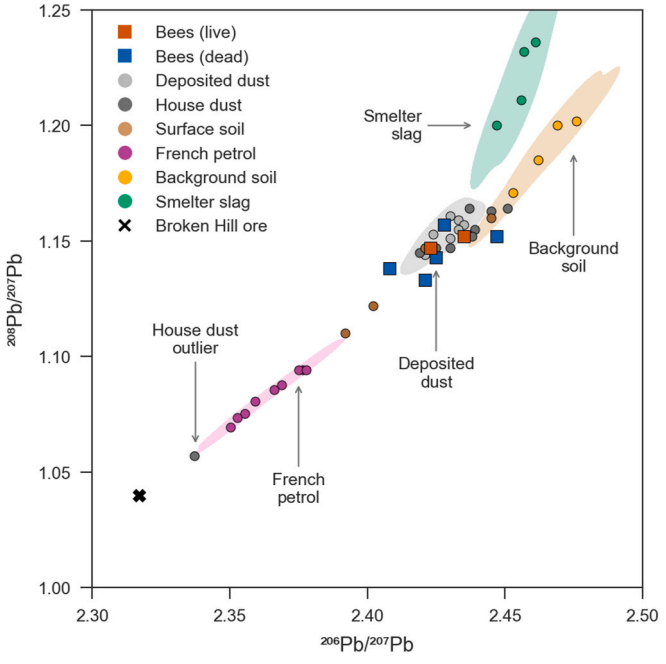

Fig. 4. Lead isotopic compositions (PbIC) of environmental and biological samples from Noumea, New Caledonia. Data includes PbIC of live bees (n = 2) and dead bees (n = 5) from this study along with other matrices from Fig. 4 in Fry et al. (2021). Due to the comparatively low Pb concentrations in honey, honey PbICs was unable to be measured. Envelopes indicate groupings of PbIC values and are calculated using a kernel density estimate. Broken Hill ore is included as a proxy for lead paint and petrol emissions (Kristensen & Taylor, 2016) along with French petrol values (Monna et al., 1997).

3.5. Honey bees as biomarkers – on the bee, or in the bee?

To assess if contaminants were stored on or in the bees, a subset of live pooled bees from 8 hives were washed prior to analysis and compared to unwashed pooled bees from the same hive (Supplementary Table S2). Other researchers (c.f. Negri et al. (2015)) have contended that attachment of particulate contaminants on the body of honey bees is a critical pathway for the accumulation and measurement of atmospheric contaminants on honey bees. In this study however, there were no statistically significant differences in washed versus unwashed bees for concentrations of the 21 trace elements analysed (Fig. 5a). Thus, as noted by others (Dżugan et al., 2018; Zhou et al., 2018b) contaminants are more likely stored in the bee rather than on the bee.

To supplement the assessment of contaminants in live and dead bees along with the washed and unwashed live bee analyses, a small subset of dead (unwashed) bees from a hive adjacent to the port ~1.3 km south-south-east from Doniambo were examined using SEM-EDX. The purpose was to ascertain if inorganic particles, particularly those with a chemical composition corresponding to the elements associated with smelter operations or the urban environment were identifiable. Surprisingly, and in contrast to other research that examined the exterior of honey bees for fine particulate matter (Negri et al., 2015), we were unable to identify inorganic particles on the exterior of the Nouméa bees. Where particles could be identified (Supplementary Fig. S9), they were assessed using the integrated EDX analyser and shown to be characteristic of organic particles comprising of carbon and oxygen. The absence of fine particles on these honey bees may be an artefact of normal honey bee grooming behaviours (Cini et al., 2020), which could be an important pathway for contaminant ingestion in honey bees.

3.6. Honey bees as biomarkers – dead or live samples?

While it is clear that contaminants are present in the bee, the role of exposure over time, and the subsequent accumulation of trace elements is not well established. If exposure to potentially toxic trace elements is cumulative over the honey bee's lifespan and the honey bees do not eliminate the trace elements they ingest, then there is increased potential for neurological and behavioural deficits (Monchanin et al., 2021b,c; Søvik et al., 2015). This, in turn, is likely to affect their productivity, having implications for honey bee survivorship in a climate where pollinator population decline is already of alarming concern (Burden et al., 2019; Monchanin et al., 2021a; Søvik et al., 2015).

To assess the accumulation of contaminants in honey bees over their lifetime, dead bees were collected at the front of each hive on a pre-wiped plastic sheet (Supplementary Fig. S3) and analysed for trace element concentrations. This data was compared to live bee samples, which were collected directly from within the same hive. Fig. 5b shows a summary of live versus dead bees (n = 14 hives) across the full suite of trace elements analysed. The majority of trace elements were statistically higher in dead bee samples. Median concentrations of Ba, Ca, Cu, K, Na, P, Rb, Sr were not significantly different between live and dead bees, though were sometimes higher in live bees. An exception was found for B, which was significantly higher in live bees at the $p < 0.01$ level. In particular, smelter-related elements Co, Cr, Fe were significantly different at the $p < 0.01$ level and Ni at $p < 0.05$. Other common anthropogenic contaminants including Cu, Mn, Pb and Zn also returned greater concentrations in dead bees than live bees ($p < 0.01$).

Thus, as indicated in previous studies examining dead honey bees (Leita et al., 1996; Zhou et al., 2018b) it is evident that they accumulate contaminants as they age, becoming better indicators of their foraging environment over time. Given the magnitude of trace element concentration difference between honey, live bees and dead bees (Table 1; e.g., mean Ni concentrations in honey = 0.17 mg/kg; live bees = 5.33 mg/kg; dead bees = 19.8 mg/kg), it is clear that honey bees are more appropriate environmental biomarkers than honey. Furthermore, sampling and analysis of dead bees returned the same spatial patterns but with a

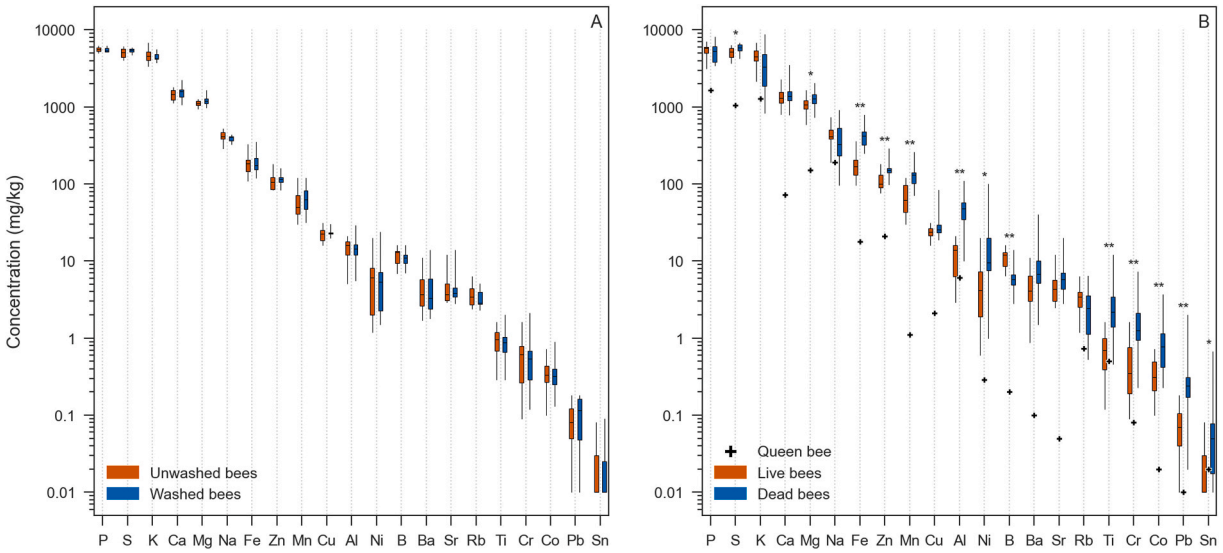

Fig. 5. Comparison of element concentrations in hive-matched samples of washed and unwashed honey bees (A) and live and dead bees (B). Boxplots show minimum, 25th percentile, median, 75th percentile and maximum. Asterisks above boxes indicate significant differences between live/dead bees and washed/unwashed bees at the <0.05 (*) and <0.01 (**) significance levels (no significant difference was detected between unwashed/washed bee concentrations for the elements assessed). Elements are arranged in descending order according to median concentrations of combined live and dead bees. Trace element concentrations for a queen bee collected ~2.3 km from the Doniambo smelter site are also shown for comparative purposes.

Table 1
Summary trace element concentrations of the different samples analysed. Additional sample concentration information is provided in Fig. 5 and Supplement Table S5. The number of individual bees sampled at each at the 15 hives is detailed in Supplementary Table S1.

Element	Dead bees (n = 14 pooled been hive samples; mean, mg/kg)	95% confidence interval	Live bees (n = 15; pooled bee hive samples mean, mg/kg)	95% confidence interval	Unwashed live bees (n = 8, pooled bee hive samples; mean, mg/kg)	95% confidence interval	Washed live bees (n = 8, pooled bee hive samples mean, mg/kg)	95% confidence interval	Queen bee (n = 1; mg/kg)	Honey (n = 15; mean, mg/kg)	95% confidence interval
Al	46.4	13.0	11.9	2.96	14.4	4.11	14.6	5.00	6.10	0.44	0.19
B	6.61	1.65	11.2	1.60	11.8	2.35	10.8	1.95	0.20	10.3	1.39
Ba	10.0	5.34	4.70	1.36	4.64	2.17	4.93	2.80	0.10	1.48	0.70
Ca	1570	374	1370	187	1450	174	1550	261	72.0	121	24.5
Co	0.97	0.46	0.35	0.10	0.37	0.14	0.38	0.18	0.02	0.01	0.00
Cr	1.91	0.96	0.52	0.22	0.63	0.33	0.67	0.43	0.08	0.03	0.01
Cu	30.0	8.37	24.0	2.28	22.4	3.51	23.5	2.10	2.10	0.16	0.03
Fe	429	81.0	186	38.5	189	47.4	201	55.9	18.0	1.56	0.54
K	3680	1200	4620	576	4720	718	4500	405	1270	1102	87.0
Mg	1330	190	1080	120	1110	73.7	1220	146	150	55.5	12.9
Mn	134	26.6	70.2	16.1	61.5	21.7	67.5	21	1.10	0.51	0.13
Na	399	123	447	67.7	419	49.8	394	26	190	136	41.7
Ni	19.8	13.4	5.33	2.53	6.73	4.23	6.96	5.07	0.29	0.17	0.08
P	5240	785	5480	478	5570	305	5540	268	1650	63.6	8.79
Pb	0.34	0.26	0.08	0.03	0.09	0.04	0.11	0.05	0.01	0.01	n/a
Rb	2.60	0.95	3.49	0.67	3.8	0.98	3.35	0.68	0.73	1.16	0.13
S	5860	378	5120	423	5080	523	5420	256	1060	60.8	7.47
Sn	0.09	0.09	0.02	0.01	0.02	0.02	0.03	0.02	0.01	0.01	n/a
Sr	6.91	2.28	4.79	1.20	4.88	2.11	5.09	2.55	0.05	0.89	0.28
Ti	3.15	1.61	0.75	0.22	0.94	0.33	0.93	0.37	0.25	0.03	0.01
Zn	159	22.1	114	15.6	112	22.5	116	16.0	21.0	0.55	0.13

greater degree of sensitivity to the pollution source than live honey bees.

It is common behaviour for 'undertaker' honey bees to deposit dead companions at the front of the hive (Visscher, 1983). Even though most invertebrate studies do not require ethical approval, any concerns raised about the ethics of conducting insect-based studies can be negated by sampling bees from the hive that have died naturally during a study period. While the cause of death and exact age of the dead bees is not known with certainty, the assumption here is that the sampled dead bees had completed their lifecycle as a forager and, as supported by the data collected in this study (Fig. 5b), reflect the surrounding environment accordingly.

3.7. Queen honey bee analysis

During sampling, a single dead queen bee was collected as part of the dead bees sampling process at a hive ~2.3 km south-east of the Doniambo smelter facility. All trace element concentrations in the dead queen bee were considerably less than those in dead forager bees and live forager bees (Fig. 5b; Supplementary Table S5). For example, with respect to Ni, dead and lives bees were 68.4 and 18.4 times greater than

those in the queen bee. While there was only a single queen bee sample available, these findings align with expectations of the queen bee life cycle. The queen bee takes a single flight to mate at the start of her life and spends the remainder of her life in the hive (unless the hive forms a swarm), and is fed by worker bees. Thus, queen honey bees are predominantly exposed to trace elements via two processes: the ambient environment, for which they have little interaction with, and from worker bees that feed them. The low trace element concentrations in the queen bee sample suggests that contaminant remobilisation within the hive and between bees is limited, highlighting an area for further research.

4. Limitations

This study sampled honey bees over a 1-month period in mid-2019, providing a snapshot of trace element contaminants in both bees and honey. Even though this study's data is limited to a 1-month period, others (Smith & Weis, 2020; Smith et al., 2019) have shown that honey bees and honey collected from the same hive display limited variation in their trace element contaminant concentrations over different seasons. Nevertheless, temporal assessment of honey bees and honey should be considered in ongoing studies using sentinel hive networks monitoring for environmental pollution.

The small number of hives and honey sampled for this study present some limitations for statistical power and confidence in data interpretation. The data collection at each of the hives was optimised by collecting multiple bees (n = ~30, Supplementary Table S1) from each location. Given that bees forage over a ~2.5 km radius from their hive (Visscher & Seeley, 1982), their bioaccumulation of trace elements represents an average across their working area (Free et al., 1983). Further, by homogenising and analysing pooled bee samples, trace element concentrations represent an average for each hive.

The number of pooled bee samples used for Pb isotopic composition was relatively small (n = 7; Supplementary Table S1, S8). However, the hives assessed for PbIC comprised 40% (6/15) of those assessed in the study, the pooled samples of which consisted of 259 individual bees (22.7% of total bees examined). In addition, the samples were collected from hives distributed across the urban area of Nouméa providing representative coverage over the study site.

Sampling for this study collected honey bees directly from their honey frames inside the hive likely comprising a mixture of younger nurse bees and foragers. Other studies have collected only foragers, which would be older honey bees, as they were exiting the hive to avoid those carrying pollen and nectar (Smith & Weis, 2020). While this study did not discriminate for age in the live bee collection, sampling of dead bees permitted a comparison of trace element concentrations in bees at the end of their life stage with values from the live bees within the hive.

Our opportunistic sampling of the single queen bee provided an interesting and useful comparator to both dead and live bees. Although single queen limits the certainty of conclusions that can be drawn, the stark difference in trace element concentrations warrants further investigation. For example, sampling of queens or even drones may be particularly useful for understanding the transfer of emerging contaminants of concern, such as antimicrobial resistance genes (Cenci-Goga et al., 2020; Fry et al., 2023; Piva et al., 2020), into the hive and through the brood cycle. Indeed, others (Zarić et al., 2022) have shown the benefits of sampling individual honey bees versus a pooled sample of honey bees as per this study. While sampling individual bees can present some logistical and analytical challenges, variation at the hive level is able to be assessed, providing additional insight into the interaction between honey bees and their environment.

Despite the limitations, the study findings are clear. Both honey and more particularly honey bees reflect trace element spatial patterns independently identified in soil and dust associated with smelter emissions and urban sources (Fry et al., 2021). Moreover, it is clear that sampling a smaller number of hive locations can characterise a wider area due to their foraging range of ~2.5 km compared to more standard environmental sampling of air, dust or soil, which is more representative of the immediate surrounds.

5. Conclusions

This study demonstrates that honey bees are suitable biomarkers for Ni smelter-related environmental contamination. The popular practice of bee keeping in urban areas provides ample opportunity for their use as sentinel pollution markers, which can identify sources and spatial variations of potential harmful exposures.

With respect to the research questions posed by this study the results show:

(1) Smelter trace elements Co, Cr, Ni were elevated in honey bee samples and declined with distance from Ni processing operations.
(2) Trace elements concentrations in washed and unwashed bees were not significantly different, but were higher in dead bees versus those sampled live, revealing that bees bioaccumulate pollutants inside their bodies as they age.
(3) Low trace element concentrations in honey samples indicates that honey consumption poses a minimal risk of harm for human health.

The findings of this study emphasise the risks posed to a Ni smelting community and affirm that honey bees are efficient biomarkers of trace element emissions. The study data reveal the prevalence, concentration and mobility of Ni and other toxic smelter elements close to the facility. These data indicate potential risk of harms to environmental health that warrant further action by industry and their regulators to mitigate any additional adverse impacts.

Credit statement

M.P. Taylor: Conceptualisation, Methodology, Validation, Formal analysis, Investigation, Resources, Data curation, Writing – original draft; Writing – review & editing, Visualisation, Supervision, Project administration, Funding acquisition; M.M. Gillings: Methodology, Formal analysis, Investigation, Data curation, Writing – review & editing, Visualisation; K.L. Fry: Methodology, Formal analysis, Investigation, Data curation, Writing – review & editing; C.F. Barlow: Conceptualisation, Methodology, Investigation, Writing – review & editing, Visualisation, Supervision; P. Gunkel-Grillion: Resources, Writing – review & editing; Romain Gueyte: Investigation, Resources, Writing – review & editing; Margot Camoin: Investigation, Resources, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgements

Macquarie University is acknowledged for supporting this project via provision of funding support for an Outside Studies Program to MP Taylor. This work has benefited from support and assistance from staff and facilities at the University of New Caledonia (Université de la Nouvelle-Calédonie): The SEM-TEM facility (P2M), Université de la Nouvelle-Calédonie, managed by Dr Aurélie Monnin; office and

laboratory access for sample preparation at the Institute of Pure and Applied Sciences (ISEA), managed by Ms Monika Lemestre. The ISEA is acknowledged for hosting MP Taylor as a visiting researcher for this project. We are grateful to the beekeepers of Nouméa who participated in this study by providing access to their hives. MP Taylor acknowledges the enduring support received from the staff at the National Measurement Institute's Inorganics Chemistry Section, Analytical Services Branch (North Ryde, Sydney) in regard to the analysis of samples and related advice. In particular, Mr Richard Coghlan and Dr Andrew Evans are thanked for their cogent guidance and assistance with the acquisition, analysis and transfer of the ethanol to New Caledonia along with specific sample method information. Finally, even though MPT completed the field work while he was on sabbatical, it would not have been possible with the help of "Uncle" Tim Stone who helped care for MPT's sons Henry and James Taylor when they were not assisting with the sampling.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2023.122257.

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