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Regional and global perspectives of honey as a record of lead in the environment

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ABSTRACT

Honey from *Apis mellifera* is a useful and inexpensive biomonitor for mapping metal distributions in urban centers. The sampling resolution of a biomonitoring survey (e.g., city versus global scale) determines which geochemical processes are reflected in the results. This study presents Pb isotopic compositions and metal concentrations in honey from around the world, sampled at varying resolutions: honey from Canada (n = 21), the United States (n = 111), Belgium (n = 25), and New Zealand (n = 10), with additional samples from Afghanistan, Brazil, Cuba, Germany, Liberia, Taiwan, and Turkey. Honey was sampled at high resolution in two uniquely different land-use settings (New York Metro Area and the Hawaiian island of Kauaʻi), at regional-scale resolution in eastern North America (including the Great Lakes region), and Pb isotopic compositions of all samples were compared on a global scale. At high sampling resolution, metal concentrations in honey reveal spatially significant concentration gradients: in New York City, metals associated with human activity and city infrastructure (e.g., Pb, Sb, Ti, V) are more concentrated in honey collected within the city compared to honey from upstate New York, and metal concentrations in honey from Kauaʻi suggest polluting effects of nearby agricultural operations. At lower resolution (regional and global scales), lead isotopic compositions of honey are more useful than metal concentrations in revealing large-scale Pb processes (e.g., the enduring legacy of global leaded gasoline use throughout the twentieth century) and the continental origin of the honey. Lead isotopic compositions of honey collected from N. America (especially from the eastern USA) are more radiogenic (^{206}Pb/^{207}Pb: 1.132-1.253, ^{208}Pb/^{206}Pb: 2.001-2.129) compared to European honey, and honey from New Zealand, which has the least radiogenic isotopic compositions measured in this study (^{206}Pb/^{207}Pb: 1.077-1.160, ^{208}Pb/^{206}Pb: 2.090-2.187). Thus, biomonitoring using honey at different resolutions reflects differing processes and, to some extent, a honey *terroir* defined by the Pb isotopic composition. The data presented here provide important (and current) global context for future studies that utilize Pb isotopes in honey. Moreover, this study exhibits community science in action, as most of the honey was collected by collaborators around the world, working directly with local apiarists and hobby beekeepers.

KEYWORDS

honey, lead isotopes, New York City, Kauaʻi, global lead array

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1. INTRODUCTION

Since the Industrial Revolution, natural emissions of certain metals and metalloids (e.g., Pb, V, Sb, Hg, Ni, Cd, Zn) have been dwarfed by anthropogenic emissions (Han et al., 2002; Nriagu, 1989; Rauch and Pacyna, 2009). Of ongoing concern is lead (Pb), a neurotoxic metal and known teratogen, which is difficult (and costly) to remediate once deposited in the environment (Landrigan et al., 2018; Mielke et al., 2011; O'Connor et al., 2020). Natural sources of Pb in the environment include volcanic emissions, sea spray, and weathering and transport of local rocks, soils, and sediments. Anthropogenic inputs include emissions from mining operations, smelting, fuel combustion (low-grade coal, fuel oil, gasoline), and battery manufacturing and recycling. The most notable modern anthropogenic inputs of Pb to the environment are the use of leaded gasoline throughout most of the twentieth century and leaded paints resulting in the contamination of indoor dust and residential topsoil (affecting gardens and playgrounds) (Filippelli and Laidlaw, 2010; Mielke, 2018; Mielke and Reagan, 1998; O'Connor et al., 2018). The chemical 'legacy' of leaded gasolines and paints govern Pb-focused environmental studies to this day, decades after termination of their widespread use, including soil remediation efforts and public health concerns relating to elevated blood lead levels, especially in children (Filippelli et al., 2015; Zahran et al., 2013).

The results of modern lead use are apparent on local and global scales. From point sources, many of which release coarse particulates (> 10 µm), the extent of environmental enrichment for Pb is local or regional since coarse particulates have shorter atmospheric residence time, minimizing transport and causing an exponential decrease in pollutant concentration in topsoils as a function of distance from the source (Dong et al., 2020; Masri et al., 2015; Reimann et al., 2011). For example, degradation of exterior paints, resuspension of road dust or contaminated soil, or mechanical wear from metal refining processes all tend to cause local Pb pollution (Laidlaw and Filippelli, 2008; Weiss et al., 2006). Conversely, fine particulates (< 2.5 µm) have longer residence times in the atmosphere before settling/deposition, so they are more susceptible to transport than coarse particulates (Wilson et al., 2005). The consequences of leaded gasoline use are evident on local, continental, hemispheric, and global scales because combustion of leaded gasoline produces both coarse particulates that are deposited along roadways, especially in areas of high traffic (i.e., cities), and fine particulates, approximately 35 % of which fall into the ‘ultrafine’ size fraction (< 0.1 µm) (Mielke et al., 2011) that are subject to long-range (i.e., transcontinental) transport. In general, high-resolution geochemical mapping reveals metal distribution at smaller scales, highlighting the impact of local point or diffuse sources, which tend to be more anthropogenic (e.g., Pb from an urban center, a smelter, or a mining operation) (Demetriades et al., 2010; Locutura and Bel-lan, 2011; Mielke, 1994). Lower-resolution studies reveal continental and global-scale processes, most of which are natural (i.e., crustal geochemistry, glaciation boundaries, large-scale climate trends) (Smith et al., 2018), but also reflect some anthropogenic activity, of which the global use of leaded gasoline is a key component.

The extent and history of Pb pollution is recorded in natural environmental archives and the analysis of Pb stable isotopes (^{208}Pb, ^{207}Pb, ^{206}Pb, ^{204}Pb) in these archives aids researchers in differentiating Pb sources (natural, anthropogenic, and legacy anthropogenic) (e.g., Komárek et al., 2008) on various scales. Environmental proxies for Pb with local or regional spatiotemporal constraints include lichens and mosses (Simonetti et al., 2003), fish and shellfish (Li et al., 2020; Shiel et al., 2012), and tree rings (Marcantonio et al., 1998; Novak et al., 2010; Patrick and Farmer, 2006). Sediment, peat, ice, and snow cores provide anywhere from continental to global-scale records of Pb pollution (e.g., Hong et al., 1994; McConnell et al., 2018; Shotyk et al., 1998; Weiss et al., 1999). Agricultural and consumer products (e.g., wine, vinegar, tobacco) have Pb isotopic compositions that reflect their local environment of origin, thus serving as an ‘archive’ of the Pb chemistry where the product originated (Epova et al., 2020; Guo et al., 2015; Kristensen et al., 2016; Medina et al., 2000; Ndung’u et al., 2011).

Honey, and European honeybees (*Apis mellifera*), are effective biomonitors of chemicals in the environment. Honey and bees have been used for monitoring pesticide residues on or near farming operations (e.g., Codling et al., 2016; de Oliveira et al., 2016; Porrini et al., 2003) and for determining metal distributions (including Pb) near point sources of metal pollution (Smith et al., 2019; Zhou et al., 2018b). Since bees forage within a few kilometers of their hive (Eckert, 1933) and passively collect dust and particulates while they forage for pollen and nectar, the bees and their products (honey, bee pollen, wax) develop a chemical composition of spatial significance, that reflects their foraging environment (Negri et al., 2015; Smith and Weis, 2020; Taylor, 2019). City-scale investigations (tens of kilometers) of metal distributions using honey and hive products are numerous (e.g., Bromenshenk et al., 1985; Conti and Botrè, 2001; Giglio et al., 2017; Skorbiłowicz et al., 2018; Van der Steen et al., 2015; Zarić et al., 2016). However, source apportionment investigations using Pb isotopic compositions in honey (and other hive products) are still somewhat rare, and the existing studies are limited to city-scale and point-source investigations (Smith et al., 2019; Smith and Weis, 2020; Zhou et al., 2018b). Published metal concentration data compiled for honey from around the world is typically reported for food science and food quality purposes (e.g. Solayman et al. 2016), rather than for geochemical surveys, or verification of honey authenticity, which are sometimes used in conjunction with carbon stable isotopes or strontium isotopes (Baroni et al., 2015; Batista et al., 2012; Zhou et al., 2018c). Missing from the literature is a global Pb isotopic database of honey. For honey to be a viable biomonitor of Pb in the future, it is essential to provide a current, global context for existing and future city-scale studies.

In this study, we present applications of honey as a biomonitor for Pb in two disparate regions: the very populated and industrialized eastern North America (n = 58, featuring samples from the New York Metropolitan Area and the Great Lakes region), and the very remote Hawaiian Islands (n = 52). We then examine honey as a biomonitor for Pb on a global scale, using a suite of honey samples collected around the world (n = 181). Using these data, we demonstrate how honey fits within the modern global Pb array, as defined by previous regional and city-scale honey studies as well as anthropogenic and natural Pb isotopic compositions measured in consumer products and aerosols. The database for Pb isotopes in honey presented here validates a ‘community science’ approach, as many of the samples were sourced from collaborators and hobby apiarists around the world.

2. MATERIALS AND METHODS

2.1. Field and sampling information

2.1.1. Field methods

Honey samples were collected by community scientists who volunteered honey from their backyard apiaries or were purchased (as commercially available honey) in the country of origin (i.e., while traveling) or in Canada and the United States (USA) as imported products. Community scientists used clean sampling vials, sampling instructions, and field worksheets for consistent metadata collection, after Smith and Weis (2020).

2.1.2. Sampling sites

Eastern North America (including the Great Lakes region) hosts the majority of the continent's population and is rich in large urban centers, which feature concentrated areas of industrial activity (U.S. EPA, 2017; Canada NPRI, 2017). The New York Metro Area includes the five boroughs of New York City (NYC) and extends into parts of New Jersey (NJ), Pennsylvania (PA), and Connecticut (CT) and is home to more than 23 million people (U.S. Census Bureau, 2016). A community science effort, notably led by a high school student, provided honey samples from NY Metro Area (n = 29) and samples from upstate NY, PA, and CT (n = 14), collected during summer 2019.

The Hawaiian Islands offer a stark contrast to eastern North America. With a total population of around 1.4 million, there are few urban and industrial (i.e., manufacturing) centers on the Hawaiian Islands; the main industries are agriculture, tourism, and military activities. The Hawaiian Islands are very remote (a volcanic island chain, located in the central Pacific Ocean), and each island is small (< 10,500 km^2), with a compact watershed and wet, tropical climate. These factors, and year-round crop production, make the Hawaiian Islands a unique setting to investigate honey as a biomonitor. Honey was sampled on the Island of Kauaʻi (31 samples from 22 unique sites) through community sampling efforts between 2013-2016 (Berg et al., 2018). These Kauaʻi samples were collected directly from feral hives, managed hives, or as commercial honey from local producers. For context, additional commercial honey samples were purchased on the Islands of Oʻahu, Maui, and Hawaiʻi (Big Island) in 2017-2018. Other significant community sampling efforts for this study occurred in and around Brussels, Belgium; Madison, Wisconsin (WI, USA); and in New Zealand (from North and South Island).

Mississippi Valley Type (MVT) galena samples (n = 4) from Lafayette County, WI, part of the Upper Mississippi Valley Mining District, were provided by the WI Geological and Natural History Survey. While not used in tetraethyl lead (TEL) production to the same extent as the southeast (SE MO) MVT ores, these samples provide important geochemical context for Upper

Mississippi Valley Pb ores used throughout the Great Lakes Megalopolis Manufacturing Belt during and after the Industrial Revolution.

2.2. Sample preparation and analysis

2.2.1. Honey

Samples were prepared and analyzed in clean laboratories at the Pacific Centre for Isotopic and Geochemical Research (PCIGR) at the University of British Columbia (UBC), except for the honey and galena collected in Wisconsin (described in section 2.2.2). All acids reagents used at PCIGR were purchased as highest-purity grade available or were distilled in-house, and only ultrapure H_2O was used (18.2 MΩ·cm). Honey samples were digested and analyzed using the methods described by Smith et al. (2019): microwave digested in concentrated HNO_3, then evaporated and reconstituted in 2 % HNO_3 + 10 ng/g indium (In, added as an internal standard for metals analysis by inductively-coupled plasma mass spectrometry, ICPMS). Each honey digestion batch also included the standard reference material (SRM) NIST 1568b (Rice Flour, National Institute of Standards and Technology, Gaithersburg, MD, USA).

Metal concentrations (Mg, Al, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Rb, Sr, Zr, Mo, Cd, Sn, Sb, Ba, Pb) were determined by ICPMS (Agilent 7700x, Agilent Technologies, Santa Clara, CA, USA). These elements include major and trace elements that originate from lithogenic and anthropogenic sources (or both), are measurable in honey, and have been useful in past studies for city-scale metal distribution surveys using honey (Smith et al., 2019; Smith and Weis, 2020). Lead isotopic compositions were measured in the same digests by high resolution (HR-) ICPMS (Nu AttoM, Nu Instruments Ltd., Wrexham, UK). In-house multicollector (MC-) ICPMS verification of NIST 1568b was previously presented by Smith and Weis (2020).

2.2.2. Wisconsin honey and galena

The Wisconsin honey (n = 7) and galena (n = 4) samples were prepared and analyzed at the Trace Element Clean Lab (TECL) facility at the Wisconsin State Laboratory of Hygiene, University of Wisconsin-Madison. All reagents were of Optima-grade, purchased from Thermo Fisher Scientific (Waltham, MA, USA). Honey samples were weighed into pre-cleaned 30 mL Teflon beakers (Savillex Corporation, MN, USA) and digested using 10 mL of 16 M HNO_3. Samples were dried, then re-digested in 9.9 mL 16 M HNO_3 + 0.1 mL 30% H_2O_2. One mL of this digest was removed and diluted in 2% HNO_3 for metals analysis using an Element2 HR-ICPMS (Thermo Fisher Scientific). The remaining solution was dried and reconstituted in 5 mL of 1 M HBr. Samples were loaded onto BioRad PolyPrep columns (BioRad Laboratories, Inc., CA, USA) containing 1 mL of pre-cleaned AG1-X8 anion exchange resin (BioRad) preconditioned in 1 M HBr. Matrix elements were removed using washes of 1 M HBr, and the Pb fraction was collected using ultrapure H_2O and 1 M HNO_3. Galena samples were dissolved using 16 M HNO_3 and were not purified prior to analysis. Final samples were diluted in 2 % HNO_3 and spiked with Tl to correct for mass bias. Pb and Tl isotopes were analyzed using a Neptune Plus MC-ICPMS (Thermo Fisher Scientific) in static mode using 6 faraday collectors, with an additional collector used to monitor for Hg interferences. Honey sample solutions were introduced into the plasma using an Aridus 3 desolvating nebulizer (Teledyne CETAC Technologies, NE, USA) and the Jet sampler and X-skimmer cone configuration. Galena sample solutions were introduced into the plasma using the standard cyclonic spray chamber with the standard sampler cone and X-skimmer cone configuration. Lead isotopic standard NIST 981 was analyzed periodically, and final isotope ratios are reported relative to NIST 981 values determined by Galer and Abouchami (1998). NIST 1568b was included for quality control.

2.4. Calculations and statistical analysis

All statistical calculations were made using R, v. 4.0.2 (R Core Team, 2020). Honey data from different global regions (e.g., Fig. 1) were compared using a Kruskal-Wallis rank sum test (Kruskal and Wallis, 1952). If the results were significant, indicated by an adjusted *p*-value ($p_{adj.}$) < 0.05, then a post hoc Dunn’s multiple comparison test (Dunn, 1964) was used to identify significantly different subgroups.

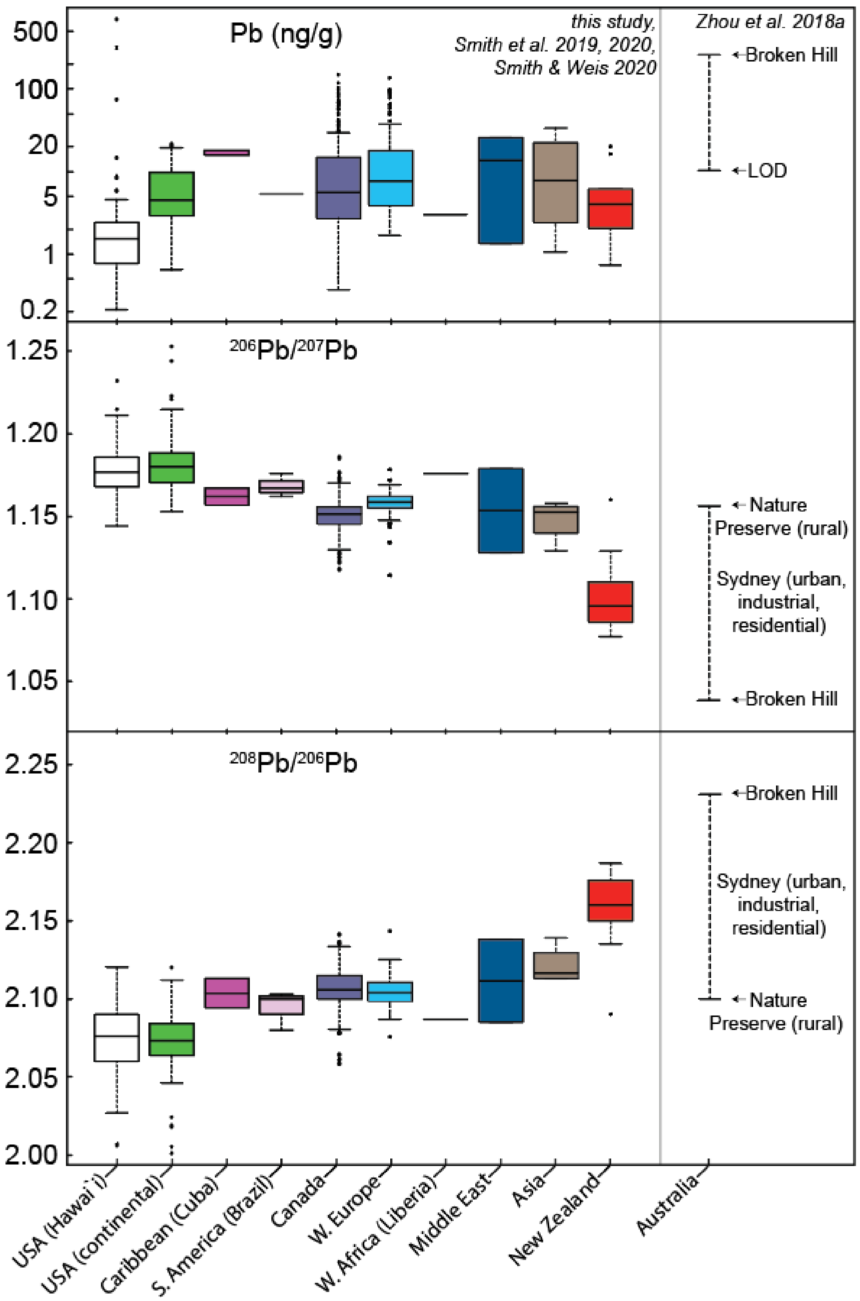

Figure 1. Box plots comparing Pb concentrations and isotopic compositions for honey, by region. The plotted data include this study (Table S1, S2), and other honey data from France (included in the 'W. Europe' region, Smith et al., 2020) and previous honey data reported for Canada (Smith et al., 2019; Smith and Weis, 2020). For comparison, Australian honey data from Zhou et al. (2018b) is also included; collected from Sydney (urban, industrial, and residential area), a nature preserve (rural), and from near the Broken Hill mine (LOD = limit of detection reported by Zhou et al., 2018b). Note that there is a logarithmic scale for the Pb concentration panel (top panel), only. A summary for pairwise statistical comparison, by region (for all regions with ≥ 10 measurements), for metal concentrations and Pb isotopes is included in the Supplementary Material (Table S3).

3. RESULTS

3.1. Blanks and standard reference materials

All metal concentrations, Pb isotope data, and summary for statistical tests are reported in the Supplementary Material (Tables S1-S3). Trace element results for microwave digestion blanks (honey digestion method) are reported in previous studies (Smith et al. 2019, Smith and Weis 2020, with an average of 36 pg total Pb). Refer to Smith and Weis (2020) for in-house HR-ICPMS and MC-ICPMS results summaries for NIST 1568b (dataset doi:10.5683/SP2/Y9MKTN). An aliquot of the same NIST 1568b used at the PCIGR (UBC) was analyzed at the TECL (UW) facility (n = 2). Average MC-ICPMS results between the two facilities are in good agreement, with relative mean differences of 3.84 per mille (‰), 0.19 ‰, and 2.75 ‰ for ^{208}Pb/^{204}Pb, ^{207}Pb/^{204}Pb, and ^{206}Pb/^{204}Pb, respectively. It is worthwhile to note that analytical results of aliquots from a newer bottle of NIST 1568b (purchased in 2020) were not in good agreement with previous results (from either facility), indicating that this SRM does not have a homogenous Pb isotopic composition. This supports previous claims (e.g., Pohl et al., 2017; Smith and Weis, 2020) that a suitable (well characterized and widely available) SRM would be very useful for studies using hive products for metal biomonitoring purposes.

3.2. Metal concentrations

Metal concentrations in all honey analyzed in this study are comparable to ranges reported in honey from all over the world, compiled by Solayman et al. (2016), and do not indicate any concern for consumer health. Our results are also generally comparable to more recent analyses of global honey by Zhou et al. (2018c), with some honey from this study exceeding their reported ranges: Kauaʻi, HI (Al, Ba, Fe), Metro New York (Fe), Brazil (Cu, Mg), Belgium (Rb), and New Zealand (Rb). While the Pb concentration range for Pb in this study is large (0.2 to 709 ng/g), most of the honey samples (> 98 %, 179 out of 181) have Pb contents below 135 ng/g. The suite of Hawaiian honey samples is unique in that it includes honey with the lowest (0.2 ng/g, Hawaiʻi Is.) and highest (709 ng/g, Kauaʻi) concentrations of Pb measured in this study. The two highest Pb concentrations measured in Kauaʻi (709 and 317 ng/g Pb) exceed concentrations previously measured in honey from urban centers: Vancouver, BC; Broken Hill, Australia; Paris, France (Smith et al., 2020; Smith and Weis, 2020; Zhou et al., 2018b), but are lower than Pb concentrations reported in honey in other studies from Europe, North Africa, Malaysia, and the USA (Solayman et al., 2016 and references therein).

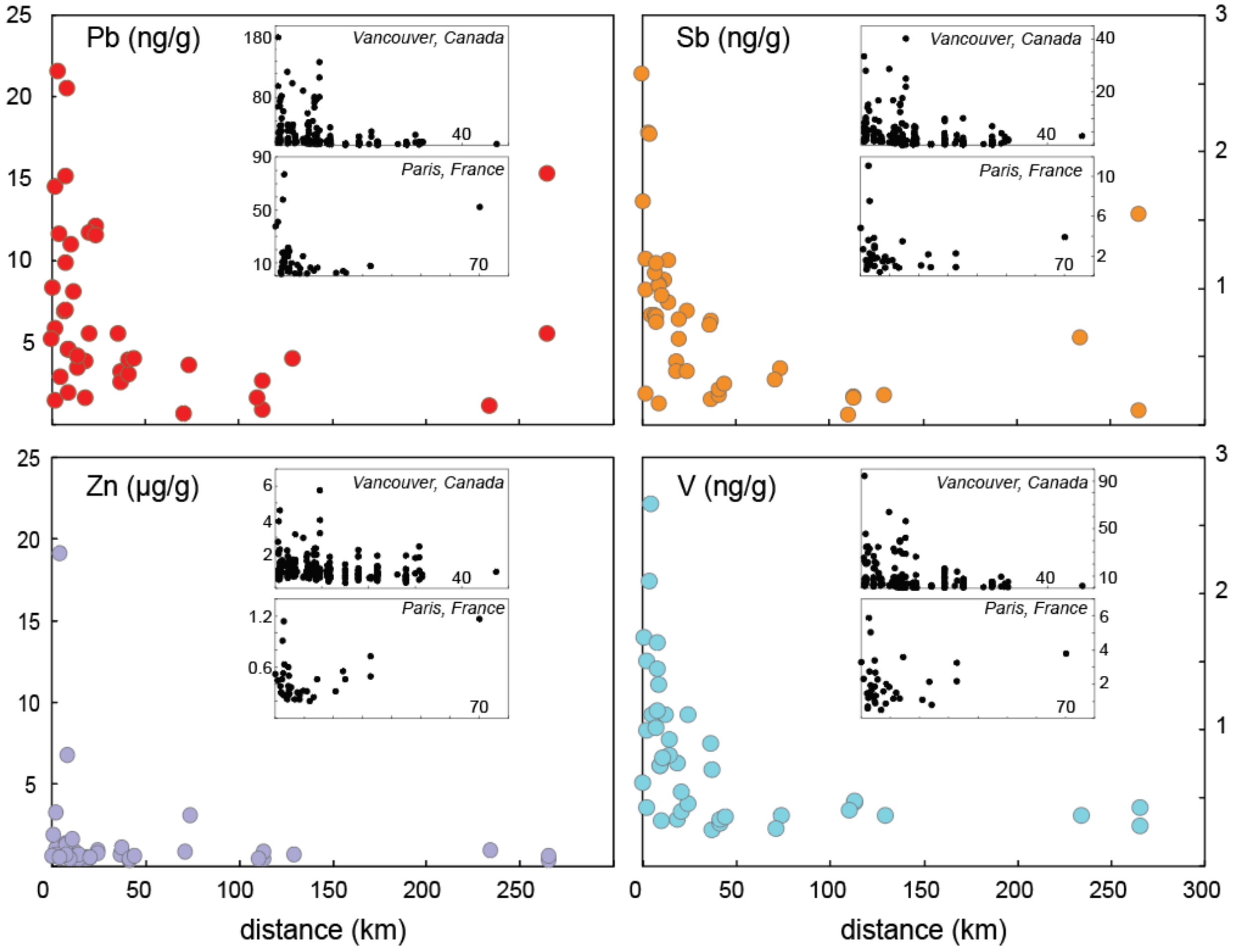

Figure 2. Metal concentrations in honey from the NY Metro Area as a function of radial distance (km) from the center of Bryant Park, located in Midtown Manhattan. That site was selected to demonstrate the effect on certain metal concentrations in honey as a function of distance from the city center. Note that units for zinc (Zn) are µg/g, while others are ng/g. Concentration maps for these metals in NY Metro honey are in the Supplementary Material (Fig. S1). For figure clarity, error bars are not included (errors are reported in Table S1). For comparison, insets show concentration plots for these metals in honey from two other cities: Metro Vancouver, Canada (data compiled from Smith et al., 2019 and Smith and Weis, 2020) and Paris, France (data from Smith et al., 2020). Distances are measured from the Port of Vancouver (after Smith et al. 2019) in Metro Vancouver and from the Notre Dame cathedral in Paris. Insets have the same units as their corresponding main plot for both axes.

For all metal concentrations, there is no systematic, geographic variation on a regional or global scale, (Table S3, Fig. 1), but in areas where sampling density was high (high sampling resolution), the local geospatial variations in metal concentrations are apparent. For example, in New York City, we observe decreasing concentrations of certain metals in honey (Pb, Ti, V, Sb, Cd; i.e., metals often associated with anthropogenic activity) as a function of distance from central Manhattan: the highest concentrations are observed in honey from within the city (Fig. 2, S1). On Kauaʻi, honey on the south side of the island has uniquely elevated levels of certain metals (e.g., Pb, Cd, Ti, V, Zn, Mn) relative to honey from elsewhere on Kauaʻi and from other Hawaiian Islands (Fig. 3, S2).

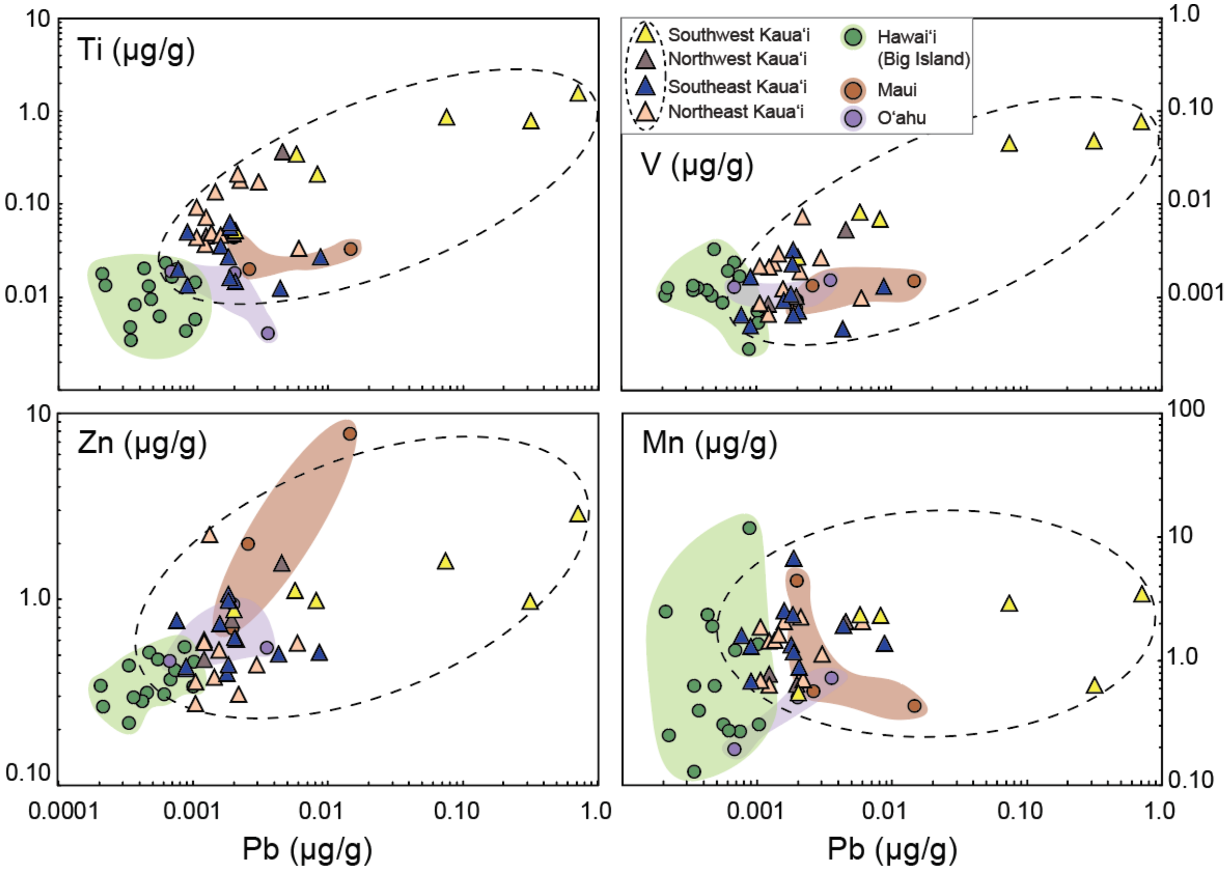

Figure 3. Metal concentration bivariate plots (Ti, V, Zn, Mn vs. Pb) for honey from four Hawaiian Islands: Hawaiʻi, Maui, Oʻahu, and Kauaʻi. All units are µg/g; note the logarithmic scale. For figure clarity, error bars are not included (errors are reported in Table S1).

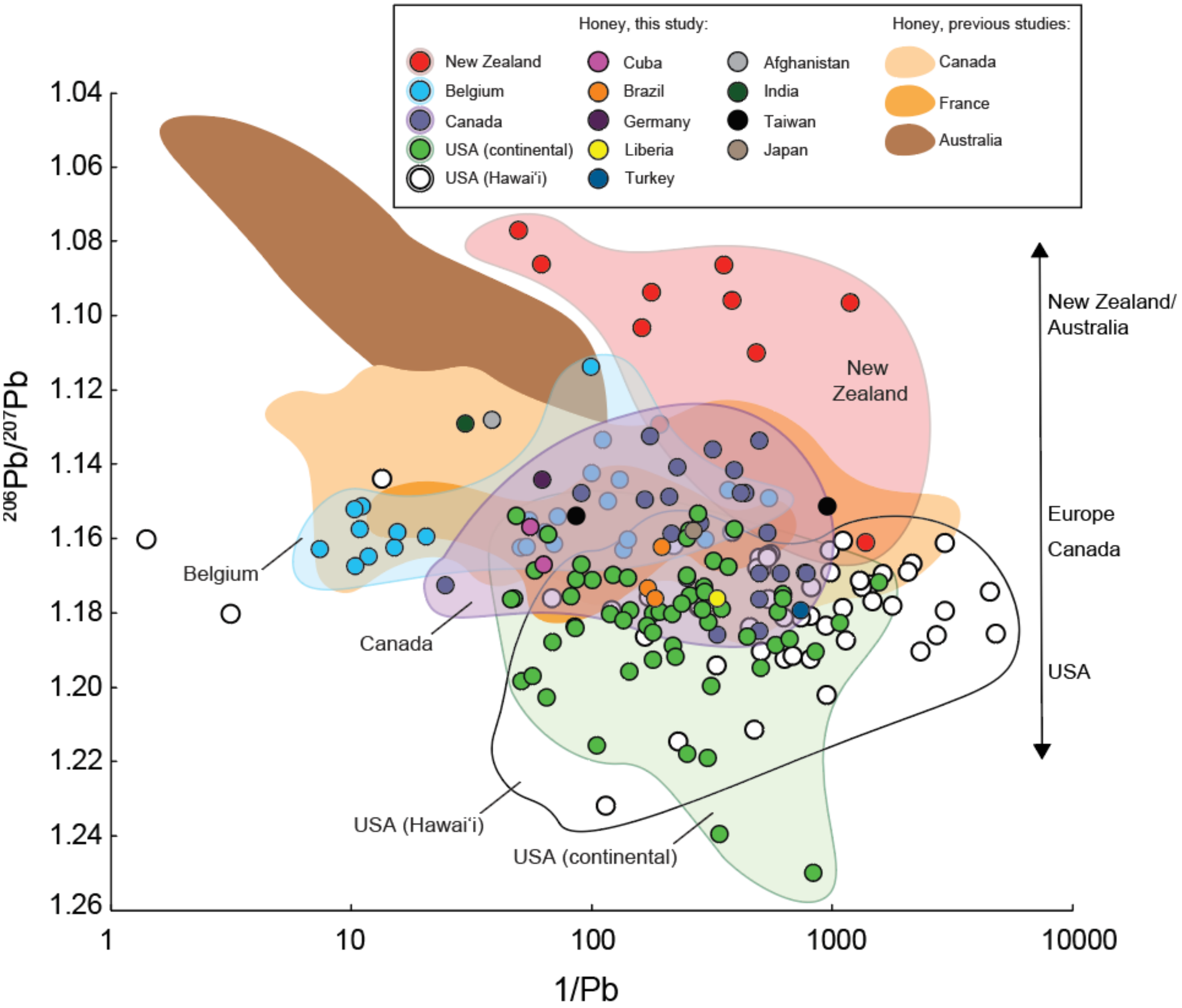

Figure 4. A plot of 1/Pb (g/µg) versus ^{206}Pb/^{207}Pb for all honey analyzed in this study. Honey from previous studies is included for context (as shaded fields): Broken Hill and Sydney, Australia (Zhou et al., 2018b); France (Smith et al., 2020); Vancouver, Canada (Smith et al., 2019; Smith and Weis, 2020). Regions from this study that include ≥ 10 samples are also shaded (Continental USA and Hawaiʻi, Canada, Belgium, New Zealand).

3.3 Pb isotopes

All honey samples analyzed in this study have a Pb isotopic range of 1.077 to 1.253 and 2.001-2.187 for ^{206}Pb/^{207}Pb and ^{208}Pb/^{206}Pb, respectively (Table S1-S2). The Pb isotopic compositions of the honey vary based on their region of origin. In general, honey collected from N. America (especially from the eastern USA) is significantly more radiogenic (higher ^{206}Pb/^{207}Pb, lower ^{208}Pb/^{206}Pb) compared to European honey, and compared to honey from New Zealand, which has the least radiogenic isotopic compositions measured in this study ($p_{adj} < 0.05$ for $^{206}Pb/^{207}Pb$ and $^{208}Pb/^{206}Pb$, Fig. 1, 4). Based on geology and histories of anthropogenic Pb use (e.g., which Pb gasoline additives were used in various parts of the world), the isotopic results were as expected for all geographic regions given that they plot along the global Pb array, which is strongly governed by the twentieth century's leaded gasoline mixing line (Fig. 5). We observe one exception in this study: honey collected from the northeast part of Kauaʻi deviates slightly from the Pb isotopic compositions of other Hawaiian honey (and the global Pb array), Hawaiian geology, and input from Eurasian dust (Fig. 6).

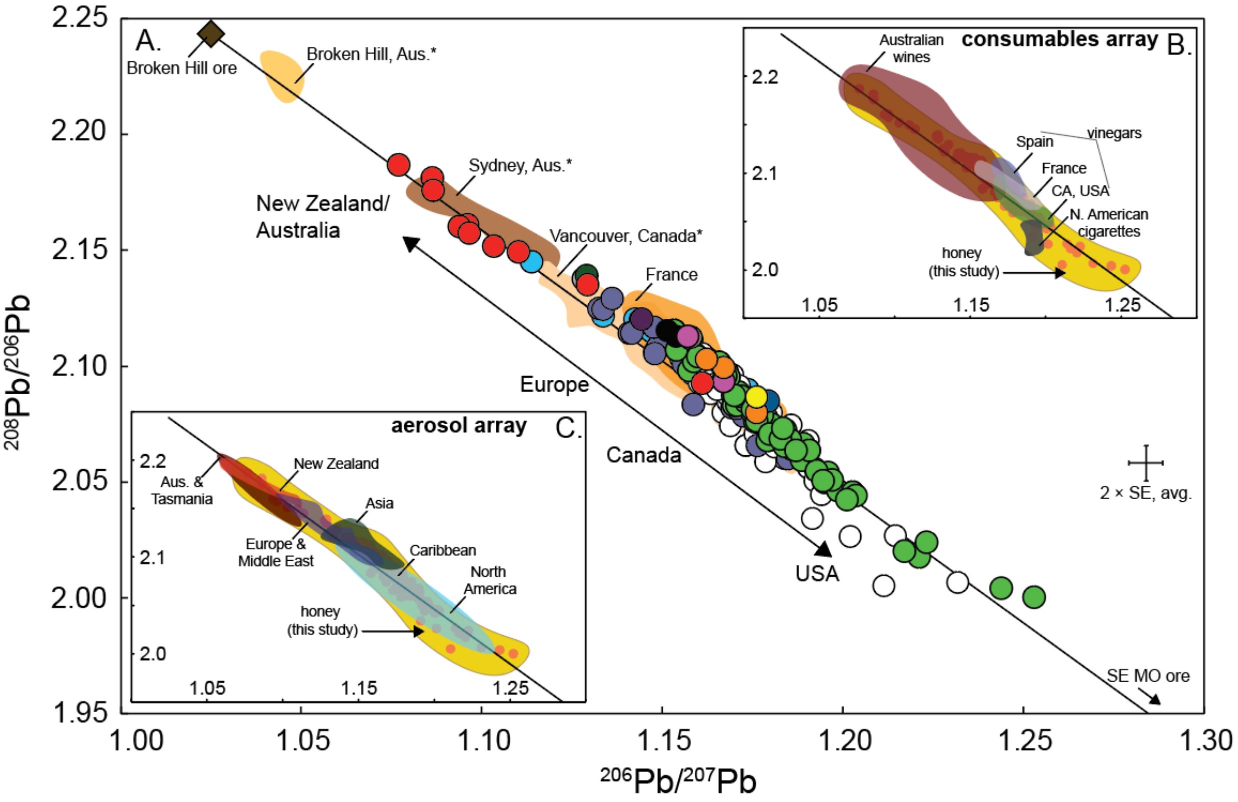

Figure 5. *Panel A*. Lead isotope plot of all honey analyzed in this study, with honey from previous studies included for context, as shaded fields (see Fig. 4 for symbol legend and caption for references). Data field labels denoted with an asterisk (*) include bees (and other hive products) in addition to honey. The Sydney field includes data for bees, honey, and wax from European honeybees (*Apis mellifera*) and an Australian native bee species (*Tetragonula carbonaria*) (Zhou et al., 2018b, 2018a). The line (featured in all panels) is a mixing line between the Broken Hill and Southeast Missouri (SE MO) ore endmembers; a representation of the isotopic compositions of leaded gasolines used globally (Gulson, 1984; Sangster et al., 2000). The average error bar (2 × standard error) is the mean error for all honey (except WI honey, for which error bars are smaller than the symbols). *Panel B*. Lead isotope plot of all honey data from this study (orange symbols in shaded yellow field), with various consumables from around

the globe for comparison: Australian wines (Kristensen et al., 2016), vinegars (apple cider, balsamic, wine, malt, cognac) (Ndung'u et al., 2011), and North American cigarettes (Guo et al., 2015). *Panel C*. Pb isotope plot of all honey data from this study (orange symbols in shaded yellow field), with the global aerosol array superimposed for comparison (Bollhöfer et al., 1999; Bollhöfer and Rosman, 2001, 2000).

The four galena samples from southwest Wisconsin have an isotopic range of 23.1996-23.8820, 16.1133-16.1797, and 42.750-43.281 for $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, and $^{208}Pb/^{204}Pb$, respectively (Table S2). These results are within the Pb isotopic range reported for other MVT Pb deposits from the Upper Mississippi Valley District and are in good agreement with other ores from southwest Wisconsin, which have notably radiogenic compositions (Fig. 7, 8) (Millen et al., 1995).

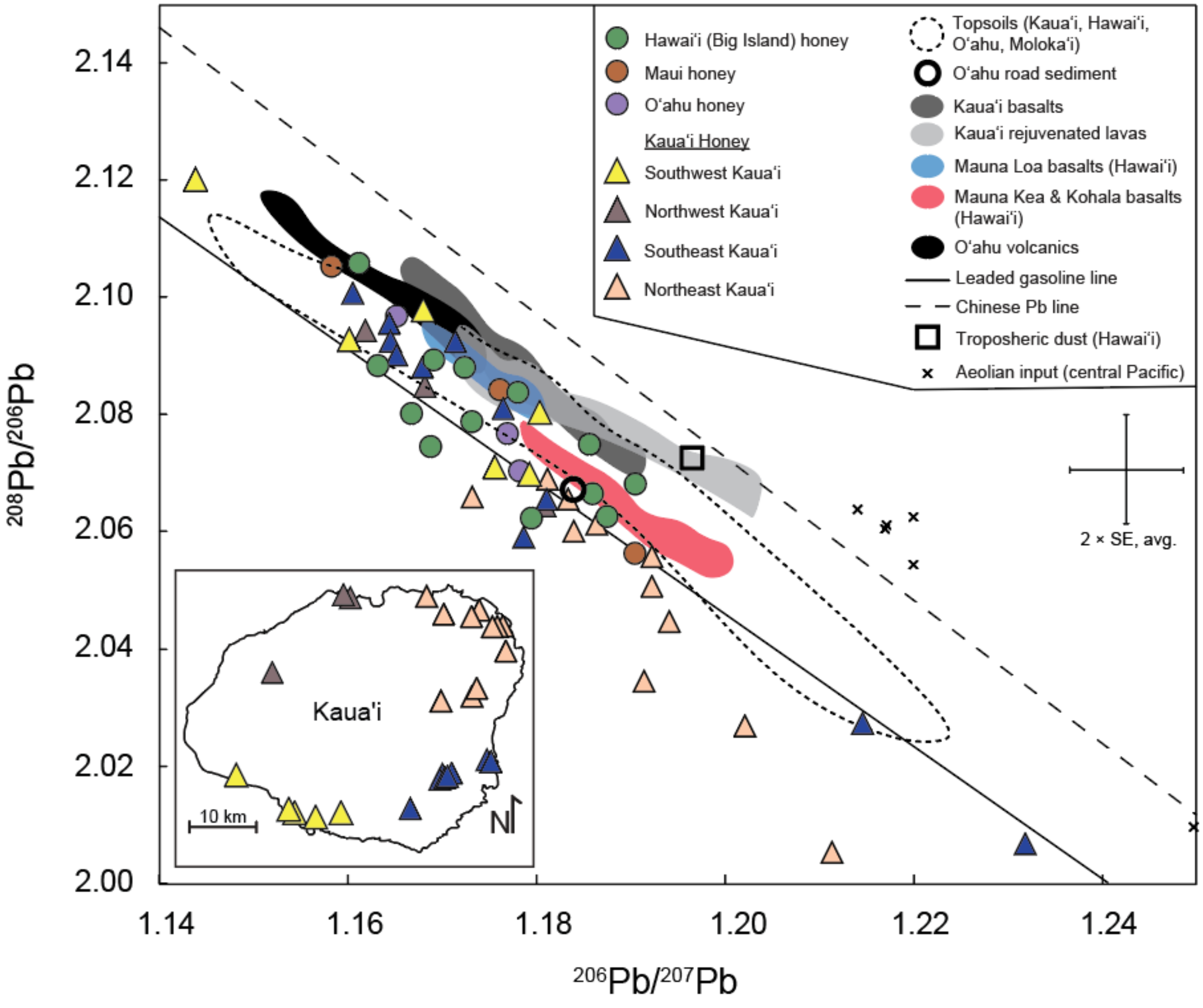

Figure 6. Lead isotope plot of honey collected from four Hawaiian Islands: Hawai'i, Maui, O'ahu,

and Kauaʻi. For context, Pb isotopic compositions of local geology is included: basalts from Mauna Loa, Mauna Kea, and Kohala volcanoes (Abouchami et al., 2000; Hanano et al., 2010; Weis et al., 2011), Kauaʻi basalts and Oʻahu volcanics (Waiʻanae and Koʻolau) (Williamson et al., 2019). Topsoils (dotted field) are compiled from Monastra et al., (2004) and Spencer et al., (1995) and Oʻahu road sediments are from Sutherland et al. (2003). The gasoline mixing line is the same as described in Figure 5. The Chinese Pb line is defined by ores, coals, and fuel emissions from China (Bi et al., 2017; Cheng and Hu, 2010). Other aeolian inputs include samples collected from central Pacific Ocean sites surrounding the Hawaiian Islands (Jones et al., 2000) and tropospheric silicate dust collected on Hawaiʻi (Spencer et al., 1995). Inset map: Kauaʻi honey sampling locations.

4. DISCUSSION

4.1. Eastern North America and the Great Lakes region

4.1.1. New York Metro Area

Cities tend to store more metals (especially Pb, as it lingers in the environment), since they host myriad industrial centers, roadways, ports, etc., in a concentrated area; the size and age of the city compound this effect (Chambers et al., 2016; Laidlaw and Filippelli, 2008; Mielke et al., 2011). Lead concentrations in the NY Metro honey exhibit a similar distribution of Pb in topsoil collected throughout NYC: higher concentrations in areas of high traffic, industrial activity, and in the oldest parts of the city, with the highest Pb concentrations measured in soils from Manhattan, Brooklyn, and Queens (Cheng et al., 2015; Li et al., 2018; Paltseva, 2019). The effect of nearby land use on other metals in honey in these regions is evident: for example, elevated Sb (vehicle brake wear) (Von Uexküll et al., 2005), Zn (galvanized structures, rubber tire crumbs) (Huber et al., 2015), and V (fuel oil combustion) (Zhao et al., 2013) (Fig. 2, S1). In a study of vegetables (root, leafy, and juicy varieties) from urban gardens across NYC, McBride et al. (2014) determined that Pb content is mostly a result of physical contamination of resuspended soil particulates and aerosol deposition, both of which are proven pathways for environmental particulates to make their way into honey (Negri et al., 2015; Pellecchia and Negri, 2018; Zhou et al., 2018b). Thus, typical city activities that contribute to emissions and particulate resuspension, such as heavy stop-and-go traffic, shipping ports, demolition, and construction, are all contributing to elevated levels of metals in NY Metro honey, relative to levels measured in honey collected further from the city center. Similar metal concentration gradients, as a function of distance from the city center, were observed in honey from Metro Vancouver, Canada, and Paris, France (Smith et al., 2020, 2019; Smith and Weis, 2020), with some logical differences (e.g., there is minimal spatial variation in V concentration in the non-port city of Paris, compared to New York and Vancouver that host major shipping ports) (Fig. 2, insets).

The Pb isotopic range of the NY Metro honey ($^{206}Pb/^{207}Pb$: 1.154-1.195) is consistent with ranges previously reported for topsoil, dust, and paints throughout the NY Metro Area. Some honey from this study is slightly (not significantly) less radiogenic than that of outdoor dust collected throughout the city from the tops of pedestrian crossing signals at major intersections ($^{206}Pb/^{207}Pb$: 1.17-1.23) (Caravanos et al., 2006). Our honey range overlaps with that of house paints ($^{206}Pb/^{207}Pb$: 1.141-1.186) and indoor dust samples ($^{206}Pb/^{207}Pb$: 1.166-1.192) collected in Hillsdale, NJ (part of the NY Metro Area) (Jaeger et al., 1998). The NY Metro honey data also falls within the Pb isotopic ranges measured in indoor house dust ($^{206}Pb/^{207}Pb$: 1.134-1.227), interior paints (1.092-1.267), garden soils (1.612-1.200), and street dust (1.155-1.199) collected in Jersey City, NJ (immediately west of Lower Manhattan, across the Hudson River, Adgate et al., 1998), and topsoil from Staten Island ($^{206}Pb/^{207}Pb$: 1.144-1.196, Pribil et al., 2014).

While the Pb isotopic compositions of NY Metro honey fall within the expected isotopic range of the region (Fig. 7), the results do not reveal a predictable geospatial gradient or trend at city-scale, in contrast to previous studies in Metro Vancouver (Smith et al., 2019; Smith and Weis, 2020) and Sydney, Australia (Zhou et al., 2018b). This is likely due to NYC being much larger and older than both Metro Vancouver and Sydney, meaning modern and historical land use, zoning, and industry are comparatively more extensive and convoluted in NYC. Indeed, a similar honey survey, recently undertaken in an even older city (Paris, France), showed a completely predictable Pb concentration gradient (matching the footprint of the fallout from the 2019 fire at Notre-Dame cathedral), while the Pb isotopic compositions of honey exhibited no geospatial trends within the city due to millennia-scale history of Pb use in Paris (Smith et al., 2020). The Paris study and the NY metro honey results imply that the utility of km-scale geospatial resolution of Pb isotopic compositions in honey is not only a function of land use and recent historical development but of the comprehensive geochemical 'baseline' of the region, i.e., all natural and anthropogenic Pb inputs, to date (Matschullat et al., 2000).

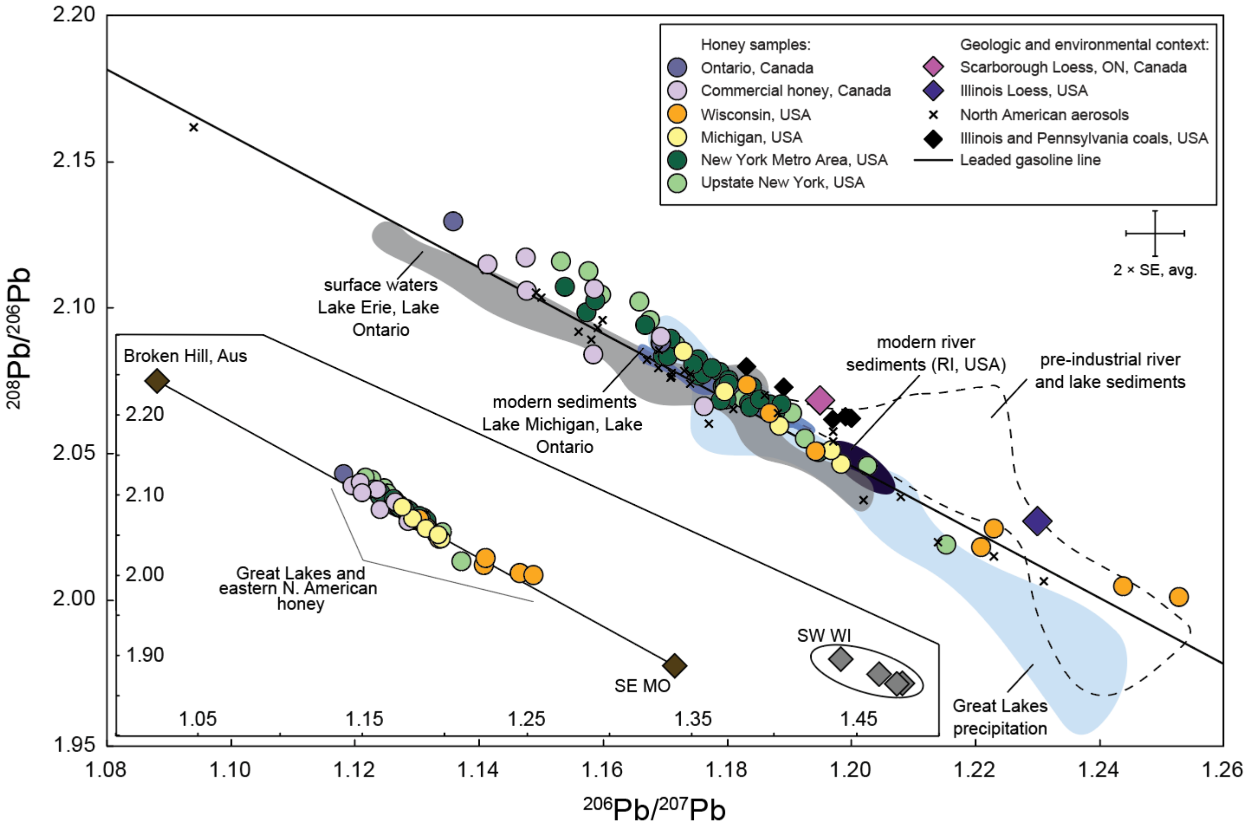

Figure 7. Honey from the Great Lakes region/eastern North America. For commercial honeys from Canada, we only included honey brands that claimed to be of Canadian origin (more specific origin information is unknown). For other honey, if specific origin location is known, it is listed in Table S1. Main plot: Selected local and regional context is provided from the literature, including North American aerosols (Bollhöfer and Rosman, 2001), loess (Biscaye et al., 1997), coals from Illinois and Pennsylvania (Chow and Earl, 1972), Great Lakes region precipitation (Sherman et al., 2015), Great Lakes (Erie and Ontario) surface waters (Flegal et al., 1989), modern (1990s and later) lake sediments from inland lakes in Michigan and Ontario (Cheyne et al., 2018), and modern sediments (1980s and later) from the Pettaquamscutt River (Lima et al., 2005). The pre-industrial field (dashed line) consists of lake and river sediments from the 1700s (Cheyne et al., 2018; Graney et al., 1995; Lima et al., 2005). The gasoline mixing line is the same as described in Figure 5. Average error bars (2 × standard error) are for all honey (except the WI honey, which has error bars smaller than the symbols). Inset: A simplified version of the main panel, zoomed out to show endmembers of the gasoline mixing line and other Pb ores. 'SW WI' is galena from a MVT ore deposit in southwest Wisconsin (Lafayette Co.).

4.1.2. Pb isotopes in USA versus Canadian honey

Lead isotopic compositions of honey from eastern USA and Canada are comparable to Pb isotopic ranges reported in other environmental matrices in that region: precipitation, snow, and modern lake sediments and surface waters, and all honey from this region plots within error of the leaded gasoline mixing line (Fig. 7 and references therein). Honey samples from the USA have Pb isotopic compositions that are more radiogenic than those from Canada ($p_{adj.} <$ 0.001 for $^{206}Pb/^{207}Pb$ and $^{208}Pb/^{206}Pb$). This can be explained by differing industrial histories in these regions and different sources of raw materials for Pb smelting, manufacturing, and recycling in each country. For instance, both countries used some amount of leaded gasoline mixed with tetraethyl Pb (TEL) produced from Southeast Missouri (SE MO) Pb ore, but Canada used only a small percentage in comparison to the USA and sourced the majority of their TEL from British Columbia and New Brunswick (Canada imported < 1 % of Pb ore used for TEL), while the USA supplemented their SE MO Pb with ores from Australia, Mexico, and Peru (Sturges and Barrie, 1987). The USA also had considerably more industrial activity, consuming more Pb and other raw materials than Canada (from ~1750 to the present), resulting in present-day Pb enrichment factors (EF_{Pb}) in lake sediments of ~100 in the Great Lakes region (USA side) compared to EF_{Pb} of ~10 in lake sediments from eastern Canada (Marx et al., 2016).

In the 1980s, when leaded gasoline was still in widespread use, dissolved Pb samples from Lakes Erie and Ontario had more radiogenic compositions when the wind was blowing from the south/USA and were conversely less radiogenic when the wind was blowing from the north/Canada (Flegal et al., 1989). Today, it seems that ongoing sulfide mining and Pb refining in the Mississippi Valley and Quebec (Abitibi Greenstone Belt) continue to ensure that honey from the eastern USA and Canada have differing Pb isotopic compositions, especially since the major metropolitan areas of the USA are downwind of key industrial and mining regions of the mid- and eastern USA, due to prevailing winds (Klink, 1999). This is further clarified when using the ^{204}Pb isotope to examine this disparity; honey from WI plots closer to the USA/MVT Pb ores, as expected based on context from USA and Canadian Pb ores and environmental samples (precipitation, snow, lichens) from both rural and urban areas in eastern Canada (Fig. 8 and references therein).

Earlier studies that focused on city-scale nuance of Pb distribution in honey demonstrated that honey can resolve input from point sources or acute Pb pollution events (Smith et al., 2019; Smith and Weis, 2020; Zhou et al., 2018b), but this localized variation is lost at the regional/sub-continental scale, which better reflects regional-scale processes, i.e., different Pb ore endmembers used in the eastern USA versus eastern Canada in this case. Metal concentrations and Pb isotopic compositions of sediment cores from southern Ontario and northern Michigan recorded the history of local and regional metal input, which, at certain points in time, overwhelmed that of the continental, global leaded gasoline trends of the twentieth century (Cheyne et al., 2018). Presumably, if honey had been collected at a higher resolution near Great Lakes industrial centers throughout the last century (e.g., Toronto, Chicago, Milwaukee, Toledo), it would reveal the local nuances of industrial pollution at each site.

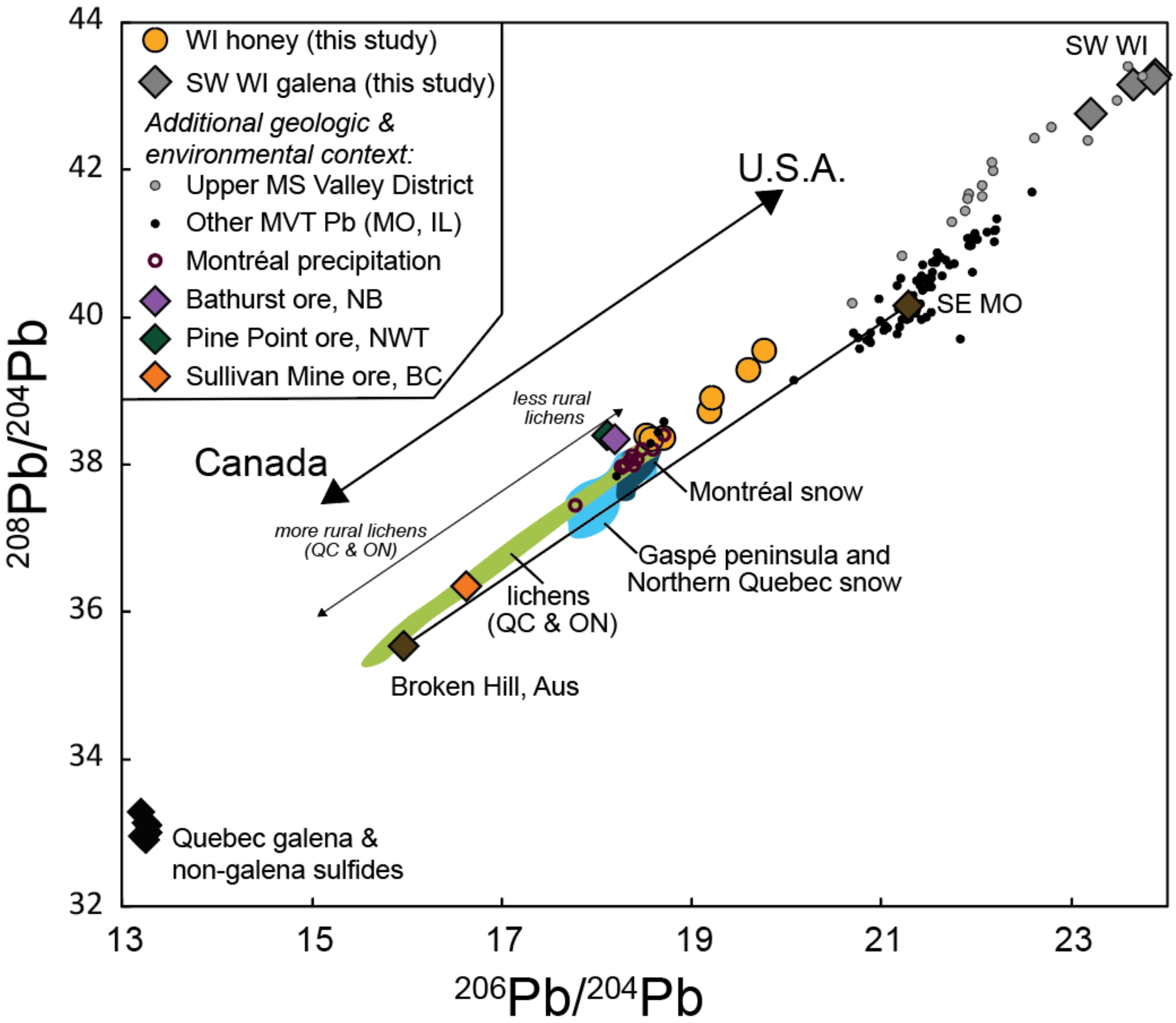

Figure 8. Lead isotope plot featuring honey from south-central WI. The honey plots toward the USA end of the Pb mixing line defined by differing emissions in eastern Canada vs. eastern USA, e.g., after Simonetti et al. (2000a). Reference reservoirs include SW WI galena (this study), Upper Mississippi (MS) Valley District MVT ores (Millen et al., 1995), other USA MVT Pb ores

(Goldhaber et al., 1995), Canadian (Quebec) Pb ores (galena and non-galena sulfides) from the Atibiti Pb sulfide belt (Franklin et al., 1983), and other economically significant Pb ores (mined throughout the past century) in Canada: Pb from the Sullivan Mine (Kimberley, BC), the Pine Point Mine (Great Slave Lake, NWT), and the Bathurst Mining District (northeast NB) (compiled by Sangster et al., 2000). Environmental context includes Montréal precipitation data (Simonetti et al., 2000b), and snowpack and lichen data from Montréal, moderately remote areas (e.g., the Gaspé Peninsula/St. Lawrence Valley), and very remote areas (i.e., along a north-south transect between southern ON/QC and Hudson Bay) (Carignan and Gariépy, 1995; Simonetti et al., 2000a). Error bars (2 × standard error) for the WI honey and southwest WI galena data are smaller than the symbols.

4.2 Kauaʻi, Hawaiian Islands

Major sources of Pb on the Hawaiian Islands are lithogenic (Hawaiian basalts), and legacy anthropogenic (leaded gasoline, leaded paint, discontinued/banned pesticides, e.g., lead arsenate). There is also some aeolian dust input from northeast Asia, confirmed by its continental-derived $^{87}Sr/^{86}Sr$ ratios (too high to be derived from the basaltic, Hawaiian geology) measured in Hawaiian topsoil (Kurtz et al., 2001; Spencer et al., 1995). Lead source apportionment calculations for road-deposited sediments from Oʻahu indicate inputs from gasoline additives of 31-81 % in sediment collected after 1968, the period when anthropogenic Pb input was more easily identified, since TEL content of USA gasoline shifted toward increased usage of the very radiogenic SE MO ores (Sutherland et al., 2003). Compared to honey from the other Hawaiian Islands, Kauaʻi honey exhibits the largest range in Pb isotopes and the most deviation from lithogenic Pb isotopic compositions, including basalts from Kauaʻi and other Hawaiian islands (Hanano et al., 2010; Williamson et al., 2019) and loess transported from the Chinese Loess Plateau and deposited in the central Pacific (Jones et al., 2000). The largest Pb isotopic ranges are observed in honey from the eastern side of the island, where most of Kauaʻi's small population (~72,200 people total) reside, so the observed isotopic compositions are likely a result of current and legacy anthropogenic activity (e.g., backyard scrapping of old vehicles, old paint, legacy gasoline, agricultural activity), especially since any deviations from the global gasoline mixing line (i.e., honey from northeast Kauaʻi) do not plot in the direction of Kauaʻi's natural/geogenic Pb isotopic compositions. For comparison, commercially available honey from Hawaiʻi Is. has Pb isotopic compositions that plot between natural and anthropogenic Pb isotopic compositions (Fig. 6).

Large-scale agricultural activity is likely contributing to the anomalously high metal concentrations observed in Kauaʻi honey compared to honey from Oʻahu, Maui, and Hawaiʻi (Fig. 3, S2). The south side of Kauaʻi is mostly in seed crop and coffee production (Spengler et al., 2019). A previous study of Kauaʻi honey established that concentrations of glyphosate (an organophosphate herbicide marketed as Roundup™ by Monsanto/Bayer AG) measured in honey correlated well with land use: hives near large-scale agriculture operations, roadsides, or golf courses produced honey with higher concentrations of glyphosate (Berg et al., 2018). The regions where Berg et al. (2018) observed the highest glyphosate concentrations in honey are in the south/southwest of the island, where we observe some of the highest metal concentrations (in particular, Pb, Ti, V, Cr, Fe, Ga, Zr, Ba, Fig. 3, S2) observed in honey, not only from the Hawaiian Islands, but in any honey analyzed for this study (Figs. 1, 4, Table S1). Pesticide and fertilizer use as a source for some of these metals should not be discounted, including Cd, Co, Cu, Ni, Pb, and Zn (Gimeno-García et al., 1996). Defarge et al. (2018) measured As, Co, Cr, Ni, and Pb (ranging from 10s to 100s ng/g) as formulant ingredients in glyphosate-based herbicides and other commercially available herbicides, fungicides, and insecticides. Fertilizer, including phosphate fertilizers, sewage sludges, manures, and fly ash (compiled by Kabata-Pendias, 2010 and U.S. EPA, 1999), also introduces metals of foreign origin into agricultural fields. Additionally, three honey samples collected in southern Kauaʻi were from feral hives, one within an abandoned building, all three within 0.5 km of abandoned farming vehicles and equipment.

Another important contributing factor for metal transport is climate. Kauaʻi's central, high-elevation regions experience large amounts of rainfall; up to 1140 cm per year on Mt. Waialeale, among the highest amounts measured anywhere on Earth (US National Weather Service, 2020), which leads to immense amounts of erosion and washout into the valleys (Ferrier et al., 2013) even on the comparatively 'dry' south/southwest side of the island. The Waimea River, which empties into Waimea Bay (mean daily discharge rate of 1.1 m^3/s, 63-years of data, USGS, 2020), is fed by a high-elevation swamp that receives significant amounts of rainfall. Coupled with a humid, tropical climate, extreme precipitation eventually reduces soil pH and mobilizes heavy metals from soils (Spencer et al., 1995). On Oʻahu, others (Andrews and

Sutherland, 2004; Heinen De Carlo and Anthony, 2002) observed that metals of both anthropogenic and lithogenic origin were transported downstream where they gradually accumulated in the upper soil horizons and sediment layers relative to background levels, so a similar transport mechanism is likely on Kauaʻi (which has undergone a similar geologic and soil-forming history) (e.g., Orazio et al., 2007).

A new, modern study on the geochemistry of Kauaʻi topsoil, C-horizon soils, sediments from Waimea Bay (downstream of the Waimea Valley), e.g., after Spencer et al. (1995), and honey would be worthwhile. In particular, sediments would offer the temporal context necessary to reveal whether the downstream sediments contained lower metal contents prior to large-scale farming efforts in the past decades. The ongoing effects of metal and pesticide transport in this region and assessment of the polluting role (if any) that other unique land use features on Kauaʻi (e.g., the US Navy Pacific Missile Range Facility, the Port Allen solar voltaic power plant, and legacy diesel-fed sugar mills) may have on the region should remain the subject of ongoing and future studies. Higher-resolution honey sampling on the other Hawaiian Islands would provide a useful comparison between the Pb isotopic compositions and metal concentrations observed on Kauaʻi versus those in larger Hawaiian urban centers (e.g. Honolulu, Oʻahu).

4.3 Honey and the global Pb array

The Pb isotopic compositions of honey fit predictably on the global Pb isotope array based on the geographic origin of the honey. For example, honey samples from New Zealand have Pb isotopic compositions that are distinctly less radiogenic compared to honey from western Europe, Canada, Hawaiʻi, and the continental USA, while New Zealand compositions are comparable to other samples from Oceania: bees, honey, beeswax, topsoil, and dust from Sydney, Australia, red wines from South Australia (Kristensen et al., 2016), and aerosols from New Zealand, Tasmania, and Australia (Bollhöfer and Rosman, 2000). The least radiogenic Pb isotopic compositions reported for honey, bees, and other hive products were collected in the vicinity of the Broken Hill Mine, Australia (Zhou et al., 2018b), an area noted for providing one of the two Pb ores used to produce the majority of the world's TEL (the other being SE MO ore, at the other end of the Pb isotopic composition spectrum for TEL used globally) (Fig. 5a).

Western European honey from this study (Belgium and Germany) have similar Pb isotopic compositions to previously reported values for honey from France (from Paris and the Auvergne-Rhône-Alpes regions, Smith et al. 2020) and have similar Pb isotopic compositions to wines and vinegars from western Europe (France, Spain, Italy) (Fig. 5b, and references therein). Honey and other products from the USA tend to have the most radiogenic Pb isotopic compositions, with honey from Canada falling between the USA and western European ranges. This is likely due to consumption of TEL from differing sources throughout the twentieth century. There are few continental-scale isoscapes available for Pb, but the honey results match isoscapes mapped using other environmental proxies: the Pb isotopic compositions for topsoil and tooth enamel from the USA are generally more radiogenic than those observed in western European isoscapes constructed using similar proxies (Keller et al., 2016; Reimann et al., 2012, 2011).

Unsurprisingly, the limited honey samples from regions other than North America, Western Europe, and New Zealand (Table S1, Fig. 5a) also plot along the global Pb array. These are the first Pb isotopic compositions reported for honey from Afghanistan, Brazil, Cuba, Germany, Liberia, Taiwan, and Turkey and may serve as a starting point for future studies in those regions. Confirming the geographic authenticity of these honey samples is beyond the scope of this study, but their Pb isotopic compositions fit well with existing Pb aerosol data from the same region/country (Fig. 5). For example, the two Cuban honey blends and three Brazilian honey samples analyzed in this study have Pb isotopic compositions in agreement with ranges reported for aerosols in Cuba and Brazil (collected at the end of the twentieth century) (Bollhöfer and Rosman, 2000).

4.4 Geochemical implications of sampling resolution

A city-scale survey of metals in honey provides insight into factors that influence metal distribution locally, and the corresponding Pb isotopic compositions can provide insight into metal sources. These factors may include local topography, land use, municipal-level environmental policy and regulation, construction, and acute pollution events (Smith and Weis, 2020). For example, the metal concentration data for honey from the New York Metro Area and Kaua'i reveal interesting local patterns of metal distribution. The metal concentrations reported in this study were selected for their utility in geochemical investigations; these metals are most useful on a local scale when combined with a high sampling resolution to differentiate between anthropogenic input versus local background, i.e., best for sourcing metals as opposed to tracing the geologic source of the honey. Thus, these metals are not directly useful for comparison between global regions since there is too much variation in metal mobility as a result of local factors such as plant diversity and soil properties (pH, porosity, organic material, and clay content) (Kabata-Pendias, 2010, 2004). The suite of metals reported in this study may nonetheless supplement future honey surveys that include additional analytes that do assist in resolving the region of origin for honey or to confirm honey authenticity and/or botanical origin. These include certain macronutrients (K, P) (Chen et al., 2014; Zhou et al., 2018c) and lanthanide series metals (Pellerano et al., 2012).

4.4.1 Geochemical 'baseline' and future of Pb isotope use in honey

Honey meets the requirements suggested by Darnley et al. (1995) for adding viable data to a global geochemical database. These requirements include being a commonly available sampling matrix where adequate amounts can be stored indefinitely for future reference. Lead isotopic compositions of wine samples from France (Epova et al., 2020) and Australia (Kristensen et al., 2016) with vintages spanning the twentieth-century exhibit a temporal trend, reflecting the introduction and subsequent phase-out of leaded gasoline in their respective geographical regions. Presumably, if honey archives existed throughout the past two centuries, they would also reflect the key moments of Pb use throughout recent history. Like wine, honey is easily stored for very long periods of time, if not indefinitely. Given that the most recent and advanced spectroscopic techniques (e.g., NMR, FT-IR) are becoming common-place for rapid-throughput authentication efforts (Wu et al., 2017), perhaps archiving of honey will become standard practice, creating a chemical repository for various environmental investigations, i.e., as a record of past environmental conditions and a continuous record of baseline compositions.

Establishing a baseline (natural/background input + anthropogenic input to-date) for a given chemical species is important in geochemical surveys (Matschullat et al., 2000; Smith et al., 2018). Since these data reflect the current baseline for Pb isotopic compositions in honey, they represent a useful tool for future comparison as Pb use shifts. However, confidently identifying the contributions of natural *versus* anthropogenic Pb in consumer products in the modern, global economic climate is becoming increasingly difficult for three key reasons. First, recent (since the mid-1800s) anthropogenic input of Pb into the environment is large (1 to 2 orders of magnitude larger) compared to natural inputs (Han et al., 2002; Nriagu, 1989; Weiss et al., 1999). Second, all anthropogenic sources were originally derived from natural sources – this is obvious but depending on the scale of a given Pb investigation (local/city-scale, regional/continental, global), this concept can conflate Pb mixing models unless possible Pb inputs are identified and quantified at each scale (Flegal and Odigie, 2020). Third, ores and raw materials are shipped globally for refining and manufacturing, then manufactured goods are shipped elsewhere, and post-consumer materials (e.g., Pb batteries) may be shipped internationally yet again for recycling. It is important to note that even raw materials from the same ore deposit can exhibit natural Pb isotopic variability (e.g., Sangster et al., 2000). The food industry has also been globalized, in particular for specialty, non-perishable items like honey where claims of 'terroir' or source environment can be economically valuable, e.g., the manuka honey industry of New Zealand (Carter et al., 2016; Rogers et al., 2014). In general, the honey market remains non-transparent. For example, the distributor of commercially-available honey purchased in North Carolina, USA, for this study stated that the product was a blend of American honey and imported honey but would not disclose any further details about mixing ratios or the geographic source locations of the honey (Pb isotopic composition of this honey is consistent with other North American honey, however).

For honey and other biomonitoring applications, we suggest using Pb isotopes in conjunction with other tools to strengthen sourcing studies. For example, analysis of Sr or Nd isotopes in honey may help distinguish geogenic inputs, e.g., dust and endmember loesses (Grousset and Biscaye, 2005), from anthropogenic inputs (Baroni et al., 2015; Voerkelius et al., 2010). We also suggest more extensive use of the ^{204}Pb isotope in environmental Pb surveys (e.g., Fig. 8) to verify that Pb sourcing interpretations are not oversimplified (e.g., Ellam, 2010). If the goal is to trace the origin of the honey, rather than the source of the Pb and other metals, there is no doubt that a global geochemical database for honey (in which sampling origin is known and knowledge exists regarding anthropogenic and geogenic sources of metals in honey) will supplement and strengthen the existing statistical approaches that use carbon isotopes and trace metal data (Zhou et al., 2018c) and the higher-throughput spectroscopy methods of honey sourcing and authentication, e.g., various infrared and nuclear magnetic resonance techniques (Wu et al., 2017).

5. CONCLUSIONS

This study presents Pb isotopic and metal concentration data for honey at three different scales: local, with high-resolution sampling (NY Metro Area and Kauaʻi), regional (Eastern North America), and global. Local-scale honey data gives insight into local processes affecting metal distribution and aids in identifying point and/or diffuse sources of metals. Metal concentrations in honey indicate potential effects of large-scale farming in and around Waimea on Kauaʻi and typical, city-type distribution gradients of metals associated with human activity (e.g., Pb, Sb, V, Zn) throughout New York City. On these small geographic scales, there is some variation in Pb isotopic composition, but their range is largely contained within the isotopic constraints defined by local sources of Pb, whether natural or anthropogenic. In larger-scale geochemical surveys using honey, metal concentrations are less useful for mapping distributions and identifying point sources and the Pb isotopic compositions reflect regional and global processes related to Pb. A regional survey of Pb isotopic compositions in honey reflects the industry, urbanization, and geology of eastern North America, revealing differences in modern Pb emissions in the eastern USA and eastern Canada. Globally, Pb isotopes in honey roughly reflect the continental origin of the honey: samples from the USA are most radiogenic and honey from New Zealand is least radiogenic. The global trend predictably matches the modern global Pb array, as defined by aerosols, varying regional geologic background, and nearly a century of leaded gasoline use around the world. This study provides a sound starting point for building current baseline data for honey as a biomonitor for metals in the regions discussed here and provides a global framework for Pb isotopic compositions in honey, which is useful context for more localized studies. This study also demonstrates the utility of a community science effort for honey collection.

SUPPLEMENTARY MATERIAL

-Tables S1-S3
-Figures S1-S2

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AUTHOR CONTRIBUTIONS

K. Smith: investigation, formal analysis, validation, writing-original draft, writing-review and editing, visualization, resources **D. Weis:** conceptualization, funding acquisition, writing-review and editing, supervision **S. Scott:** resources, investigation, validation, writing-review and editing **C. Berg, Y. Segal, P. Claeys:** resources, writing-review and editing.

CONFLICT OF INTEREST

The authors declare no conflicts of interest relevant to this study.

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Supplementary Figures

Regional and global perspectives of honey as a record of lead in the environment

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CONFLICT OF INTEREST STATEMENT

The authors declare no conflicts of interest relevant to this study.

Declaration of Interest Form

Declaration of interests

☒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.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: