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# Risk assessment of soil heavy metal contamination at the census tract level in the city of Santa Ana, CA: implications for health and environmental justice†

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(1) Background: exposure to heavy metals is associated with adverse health effects and disproportionately impacts low-income communities and communities of color. We carried out a community-based participatory research study to examine the distribution of heavy metal concentrations in the soil and social vulnerabilities to soil heavy metal exposures across Census tracts in Santa Ana, CA. (2) Methods: soil samples ( $n = 1528$ ) of eight heavy metals including lead (Pb), arsenic (As), manganese (Mn), chromium (Cr), nickel (Ni), copper (Cu), cadmium (Cd), and zinc (Zn) were collected in 2018 across Santa Ana, CA, at a high spatial resolution and analyzed using XRF analysis. Metal concentrations were mapped out and American Community Survey data was utilized to assess metals throughout Census tracts in terms of social and economic variables. Risk assessment was conducted to evaluate carcinogenic and non-carcinogenic risk. (3) Results: concentrations of soil metals varied according to landuse type and socioeconomic factors. Census tracts where the median household income was under \$50 000 had 390%, 92.9%, 56.6%, and 54.3% higher Pb, Zn, Cd, and As concentrations compared to high-income counterparts. All Census tracts in Santa Ana showed hazard index  $>1$ , implying the potential for non-carcinogenic health effects, and nearly all Census tracts showed a cancer risk above  $10^{-4}$ , implying a greater than acceptable risk. Risk was predominantly driven by childhood exposure. (4) Conclusions: findings inform initiatives related to environmental justice and highlight subpopulations at elevated risk of heavy metal exposure, in turn underscoring the need for community-driven recommendations for policies and other actions to remediate soil contamination and protect the health of residents.

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## Environmental significance statement

This study employed a community-based participatory research approach to collect and analyze a large number of randomly sampled soil measurements so as to yield a high spatially resolved understanding of the distribution of heavy metals in the soil, and in turn reduce exposure misclassification. The assessment examines average metal concentrations at the Census tract level and by landuse type, which facilitates an understanding of potential contributing sources of heavy metals in the soil and the association between soil contamination and socioeconomic characteristics.

## 1. Introduction

Heavy metal soil contamination is a global problem due to anthropogenic emissions of metals and their persistence and non-biodegradable nature. Soil-related heavy metal exposure is particularly important in school and residential areas where children play, as well as industrial sites and recreational areas where soil particles can become resuspended into the air. Importantly, low-income communities, communities of color and urban residents incur disproportionately elevated exposures to such metals.<sup>1–3</sup>

Exposure to heavy metals is associated with a range of adverse health outcomes. Evidence of the harmful health effects of lead (Pb) exposure includes asthma<sup>4–7</sup> and adverse neurological and cognitive outcomes in children,<sup>8–11</sup> as well as

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pregnancy complications.<sup>12–15</sup> Other heavy metals such as nickel (Ni) and chromium (Cr) are also associated with asthma and inflammation,<sup>16,17</sup> while cadmium (Cd) has been associated with high blood pressure, hypertension and osteoporosis.<sup>18–20</sup>

Globally, arsenic (As) is one of the most problematic metals, causing peripheral vascular and cardiovascular diseases in chronically exposed populations and linked with type II diabetes.<sup>21–23</sup> Regarding manganese (Mn), Lucchini *et al.* (2007) found an increased prevalence in Parkinson's disease in communities with chronic environmental exposure,<sup>24</sup> and also demonstrated significant deficits in hand dexterity, motor coordination, and odor identification among Mn-exposed adolescents relative to a non-exposed reference group.<sup>25,26</sup> Other metals, including Pb and As, are also known to impact the central nervous system.<sup>22,23</sup>

Kidney damage is associated with Pb, Cd, and (copper) Cu exposures.<sup>22,27</sup> At ambient levels, zinc (Zn) and Cu tend to be the least toxic of the metals described, although health effects still include headaches, dizziness, and irritation of the nose, mouth and eyes for elevated Cu exposure, and cytotoxicity for Zn exposure. Importantly, Pb, As, Ni, Cd, and Cr are known human carcinogens, associated with cancers of the lung, kidney, liver, and bladder.<sup>16,17,21,28</sup>

Sources contributing to soil heavy metal accumulation, and therefore exposure, include lead paint from older homes and structures,<sup>29,30</sup> as well as the historic use of lead in on-road vehicles, agricultural products, and smelting process.<sup>31</sup> Cr and Ni mostly enter the environment through releases by electroplating processes and the disposal of metal-containing waste, while other heavy metals are emitted through a combination of industrial activities such as mining, agricultural inputs (*i.e.* pesticides, fertilizers), fossil fuel and waste combustion, and metal processing.<sup>32</sup>

Evidence of widespread heavy metal exposure and impacts has given rise to policies to bar the sale of certain heavy-metal-containing products (*e.g.* lead paint) in the U.S. and to the listing of the heavy metals As, Pb, Cd, Cr, Cu, Ni and Zn as priority control pollutants by the United States Environmental Protection Agency (USEPA).<sup>33</sup> Despite such measures, households, individuals, and communities continue to incur metal exposures through current and past emissions and products.<sup>34,35</sup>

Although mostly focused on Pb, evidence has shown that heavy metal exposure is not equally realized across the United States. In Oakland, California, McClintock (2012) reported significantly higher Pb levels in West Oakland, made up predominantly of low-income and African American residents, relative to the affluent, predominantly white Oakland hills area. Similarly, Zhuo *et al.* (2012) showed the proportion of Latina/o/x residents within Census tracts to correlate with soil Pb levels in Phoenix, Arizona.<sup>36</sup> In our prior work investigating soil Pb contamination in Santa Ana, CA, Census tracts with a greater proportion of low-income or Latina/o/x residents showed roughly two- to five-times higher soil Pb levels relative to their higher-income or less-Latina/o/x counterparts.<sup>37</sup> Studies have also shown Black<sup>1</sup> and Latina/o/x<sup>1,2</sup> children, and those from low-income households,<sup>1</sup> to consistently show elevated blood

lead concentrations compared to non-Latina white children and children from higher income households.

Although evidence suggests that a similar pattern may be evident as it relates to the distribution of other (non-Pb) soil heavy metals in the context of social and economic factors, this area has been less studied compared to Pb.<sup>38</sup>

Specifically, to our knowledge, few studies to date have carried out a high-resolution assessment of the spatial distribution of multiple heavy metals in an urban environment while also considering the spatial distribution of social and economic characteristics of the resident populations.<sup>39,40</sup> Of the studies we identified, none carried out a quantitative health risk assessment using toxicity values to estimate cumulative risk by Census tract.

In this analysis, we built upon our prior investigation of the spatial distribution of soil Pb and exposure-related social vulnerabilities in Santa Ana by considering the concentrations, distributions, and cumulative health risks related to eight heavy metals that are of particular relevance to public health, including Pb, As, Mn, Cr, Ni, Cu, Cd, and Zn.<sup>38,41–43</sup> This study centers around the following questions: (1) are residential socioeconomic status and racial/ethnic factors at the Census tract level correlated with concentrations of heavy metals in the soil? (2) Given childhood vulnerability to heavy metal exposure and health impacts, are Census tracts with a higher proportion of children correlated with elevated soil heavy metals? (3) Is Santa Ana at a high risk of non-carcinogenic or carcinogenic health outcomes due to soil contamination? (4) Do economic and social vulnerabilities aggregate so as to create heavy metal-related vulnerability hotspots across certain Census tract?

Based on experiences and concerns that have emerged from discussions with our community-academic partners and affected residents, as well as our review of the literature, it was our hypothesis that Census tracts of lower socioeconomic status, with a higher fraction of residents who identify as Latina/o/x, and those with a higher proportion of children have increased soil heavy metal concentrations; that hotspots exist related to cumulative health risk; and that economic and social vulnerabilities to soil heavy metal exposures correlate with cumulative risk.

## 2. Materials and methods

This analysis was carried out as part of the ¡Plo-NO Santa Ana! Lead-free Santa Ana! community-academic partnership. Since 2017, our partnership has been working to understand and intervene upon environmental injustices to promote health equity and economic, political, and social well-being in Santa Ana, CA.<sup>44</sup> Partners consist of Orange County Environmental Justice; Jóvenes Cultivando Cambios (a youth-led cooperative); and faculty and staff at the University of California, Irvine. Importantly, these community and academic partners were all intimately involved in each step of the research process from visualization and conception to data collection and the guiding of central research questions and analyses. Examples of these collaborative processes in action included various in-person workshops that we held both in Santa Ana and at the



university (UC Irvine) during which local residents and volunteers of all ages came together with academic partners in order to discuss issues related to local heavy metal exposure and public health, develop skills related to data collection, and share ideas and questions that would ultimately guide our research. For instance, as a project deliverable, several residents stated an interest in learning whether soil contamination was greater in poor neighborhoods relative to more affluent areas. Based on such input, we then incorporated this research question into our analysis.

During the onset of the COVID-19 pandemic, in-person community workshops and meetings turned into weekly virtual meetings, during which community partners were regularly updated as to the most recent findings concerning data analysis. Further details about this partnership and its origin can be found in our previously published work.<sup>37,44</sup> Data used in this analysis include soil samples obtained by trained personnel as well as demographic information from the U.S. Census Bureau's American Community Survey.<sup>45</sup> This study was classified as exempt by the Institutional Review Board at the University of California, Irvine.

## 2.1 Study region

Located in the southwestern region of California, in the United States, Santa Ana is a densely populated city with an estimated 337 716 residents. With 61 Census tracts, Santa Ana spans 70.6 km<sup>2</sup>.<sup>46</sup> The administrative center of Orange County, Santa Ana is the second biggest city in Orange County and is the eleventh biggest in California.<sup>46</sup> The majority of residents in Santa Ana identify as Latina/o/x (77.3%), followed by Asian (11.4%) and white (9.4%), with a high proportion of immigrant residents (45.2%).<sup>47</sup> As of 2019, Santa Ana includes 78 563 housing units and has a median household income of \$65 313 (2018 dollars).<sup>46</sup>

## 2.2 Field sampling

In Summer–Fall 2018, soil samples were collected across seven landuse types in Santa Ana, CA: arterial roads, parks and gardens, schools, industrial regions, business areas, and residential zones. Since the majority of businesses, schools, and industrial areas were not accessible directly, soil samples were obtained at their boundaries (*e.g.* roadway near facility). When possible within Census tracts, a minimum of six residential units were sampled. Field teams recorded landuse categories and the location of each sampling site using coordinates identified through global positioning system (GPS) readings. Field teams responsible for soil sampling and the recording of landuse types consisted of local community volunteers, including numerous members of the youth community, who were first trained by a field coordinator.

Building upon methodologies by Wu *et al.*,<sup>48</sup> field teams selected sampling locations at each sampling site that were not obstructed by physical barriers. Where possible, field teams marked a three-foot radius, and obtained soil samples from five distinct points (one central point and four other points that were three feet away from the central point) after removing 1 cm of soil (including vegetative matter). At residential units, field

teams drew samples from dripline areas around the home and a minimum of two locations throughout the yard (*e.g.*, front yard, back yard). Between four and five samples were collected from each garden site. In preparation for laboratory analysis, all samples were air dried and sieved with brass screen (#50 mesh, twice; #100 mesh once), which yielded fine soil particles. In a Santa Ana pilot soil analysis, we found lead concentrations in soil to be very similar whether sieving for coarse or fine soil particles. Thus, in the present study, we focused on fine soil particles since this size fraction resembled the heavy metal exposures that pose a particular threat to young children.<sup>49</sup> In total, 1528 samples across 560 different locations were obtained throughout Santa Ana, CA, resulting in a highly spatially resolved characterization of soil heavy metals. For the purpose of establishing baseline soil heavy metal levels, eight soil samples were collected in areas outside of the Santa Ana, in nearby state and regional parks in the county that were relatively natural and unimpacted by local anthropogenic heavy metal sources such as industry, traffic, and construction.

## 2.3 Soil analysis

Soil samples were measured using XRF technology (SPECTRO XEPOS HE Benchtop XRF Spectrometer), a well-established procedure for identifying the concentrations of commonly measured heavy metals in different sampling media.<sup>50</sup> XRF methods have been used extensively in scientific research<sup>39,51,52</sup> and have been shown to give results that are comparable to ICP-MS methods.<sup>50</sup> However, compared to ICP-MS and AAS methods, XRF analysis has the advantage of being non-destructive, which allows for flexibility in doing future analyses using the same samples. The instrument employed in this investigation operates optimally at temperatures between 20–25 °C and undergoes regular multi-channel analysis calibration using standard reference materials on a weekly basis, with global calibration conducted twice per year. Each soil sample was analyzed five times to ensure reproducibility and stability of measurements. Absolute measurement errors were in the range of 0.1% and 2.5% for all elements described in this study, with the exception of As and Cd, which had errors of 9.6% and 20.2%, respectively. To reaffirm quality laboratory analysis, a subset of soil samples ( $n = 18$ ) were subjected to a second round of XRF analysis (five more scans), resulting in a very high correlation ( $r = 1.0$ ). The limit of detection (LOD) using the XRF instrument was very low and varied depending on the element analyzed. LODs for the elements analyzed in this study were 0.1 ppm (As), 0.2 ppm (Pb, Mn, Cr, Cd, and Zn), 0.5 ppm (Cu), 1 ppm (Ni).<sup>53</sup>

## 2.4 Landuse

Some landuse categories were sufficiently similar such that we consolidated them, resulting in more meaningful sample sizes. For this study, garden and park samples were considered a single landuse category labeled “park,” while business and industrial area samples were consolidated under the label “industrial.” Of note, arterial roadway samples are referred to as “roadways.” Accordingly, there were five landuse types in total



in this study including: schools, roadways, parks, residential units, and industrial areas.

## 2.5 Demographics

Population information for Census tracts ( $n = 61$ ) in Santa Ana, CA, were drawn from the 2010 Census. Demographic factors at the Census tract level, including household income, education, race/ethnicity, insurance coverage, nativity, languages spoken, and age were drawn from the American Community Survey (ACS), which is conducted annually. ACS data averaged over five years from 2012–2016 (henceforth, 2016) was employed in this study since averages represent a more stable estimate of community-level characteristics, and since 2016 was the most recent year for which geo-coded ArcGIS files were available.

This study also makes use of a so-called vulnerability index that was developed in our previous work in order to compare social and economic vulnerability with heavy metal concentrations and risk.<sup>37</sup> This index took into account six social and economic factors that could result in a community being at an increased health risk due to heavy metal exposure, including: median household income, percent of housing units occupied by renters, percent of population under age five, percent of residents reporting speaking limited or no English, percent of residents without health insurance coverage, and percent of residents with a college education or higher. Values for each factor were calculated based on quartile distribution rankings, and scaled to range from 0 (low risk) to 1 (high risk). Further details on the development of this index can be found in Masri *et al.* (2020).

## 2.6 Analysis

Summary statistics for heavy metal concentrations were calculated across all soil samples and for groups of samples based on landuse categories, as well as for one (baseline) group of samples obtained outside of Santa Ana. To visually depict the distribution of heavy metal concentrations and estimate their levels between sampling sites, we employed ArcGIS software to carry out simple kriging for each element using variogram covariances that employed 12 lags and a default lag size, nugget size, and partial sill.

To assess the variability in soil metal concentrations and demographic characteristics within Census tracts, we calculated specific demographic indicators including the proportion of residents who identified as Latina/o/x or Hispanic, those who reported speaking no or limited English, residents who identified as immigrant or non-native (henceforth, immigrants), those who did not possess health insurance, residents living in renter-occupied housing, residents under five years old and those with a college education or higher. Once these proportions were calculated, we then matched them with soil samples and sorted them according to their demographic attribute values. This allowed us to separate heavy metal samples into distinct tertiles based on the demographic attributes of the Census tracts from which each sample originated. Details on these methods can be found in our prior work.<sup>37</sup> Given a total of  $n = 1528$  samples, the sample size for each tertile was

approximately  $n = 510 \pm 20$ . To determine statistical significance between sample means, we employed a cutoff of  $p = 0.05$ .

**Enrichment factor.** An enrichment factor (EF) is a geochemical criterion that is commonly used to assess the extent of heavy metal contamination of the soil.<sup>54</sup> It is defined according to the following equation:

$$EF = \left( \frac{C_n}{C_{ref}} \right)_{sample} \div \left( \frac{B_n}{B_{ref}} \right)_{background} \quad (1)$$

where  $(C_n/C_{ref})_{sample}$  represents the concentration ratio of a given metal and a given reference element in a sample, and  $(B_n/B_{ref})_{background}$  represents the ratio of the natural baseline concentration of that metal relative to the reference element. Elements that are most commonly used as reference elements include conservative elements such as iron (Fe) and aluminum (Al).<sup>55</sup> In this analysis, both Fe and Al were used and compared as the reference elements. Baseline values utilized to calculate EF values were derived from the eight previously described baseline samples collected outside of Santa Ana, CA.

To interpret EF values, we employed a five-category system previously introduced by Sutherland and used in other studies,<sup>55,56</sup> which indicates: no or minimal enrichment ( $EF < 2$ ), moderate enrichment ( $2 \leq EF < 5$ ), significant enrichment ( $5 \leq EF < 20$ ), very high enrichment ( $20 \leq EF < 40$ ), and extremely high enrichment ( $EF \geq 40$ ).

**Hazard index.** To characterize the risk associated with the eight heavy metals Pb, As, Mn, Cr, Cu, Ni, Cd, and Zn in urban soil, we employed a methodology recommended by the U.S. Environmental Protection Agency, which allows for the calculation of cumulative risk or chronic daily intake (CDI) across multiple metals according to three potential exposure pathways (ingestion, dermal contact, and inhalation), as follows:

$$CDI_{ing.} = \frac{(Cs \times CF \times IRs \times EF \times ED)}{BW \times AT} \quad (2)$$

$$CDI_{derm.} = \frac{(Cs \times CF \times SA \times AF \times ABS \times EF \times ED)}{BW \times AT} \quad (3)$$

$$CDI_{inh.} = \frac{\left( Cs \times \frac{1}{PEF} \times IRa \times ET \times EF \times ED \right)}{BW \times AT} \quad (4)$$

where  $CDI_{ing.}$  is either child or adult chronic daily intake of metals through ingestion (mg per kg per day),  $CDI_{derm.}$  is the chronic daily intake of metals through dermal absorption (mg per kg per day), and  $CDI_{inh.}$  is the chronic daily intake of metals through inhalation (mg per kg per day). Exposure factors used in these calculations, which differ for children and adults, are the same as those recommended by the EPA, and are presented in Table 1.<sup>57</sup>

To derive either child or adult non-carcinogenic risk associated with each exposure route for each metal, the hazard quotient (HQ) for a given metal was calculated by dividing the CDI values for each exposure route by the reference dose (RfD) (mg per kg per day) for that metal, in turn yielding a unitless value according to eqn (5):





Table 1 Heavy metal exposure factors used in eqn (2)–(4)

Factor	Definition	Unit	Value	
			Child	Adults
Cs	Soil metal concentration	mg kg <sup>-1</sup>	—	—
IRs	Soil ingestion rate	mg per day	200	100
SA	Skin surface area available for exposure	cm <sup>2</sup> per day	2373	6032
AF	Soil-to-skin adherence factor	mg cm <sup>-2</sup>	0.2	0.07
IRa	Inhalation rate	m <sup>3</sup> h <sup>-1</sup>	0.53	0.83
ED	Exposure duration	Years	6	20
BW	Body weight	kg	15	80
AT	Averaging time	Days	365 × ED	365 × ED
PEF	Soil-to-air particulate emission factor	m <sup>3</sup> kg <sup>-1</sup>		1.36 × 10 <sup>9</sup>
CF	Conversion factor	kg mg <sup>-1</sup>		1 × 10 <sup>-6</sup>
ET	Exposure time	Hours per day		24
EF	Exposure frequency	Days per year		350
ABS	Absorption factor	Unitless	0.03 for As, 0.01 for other metals	

$$HQ = \frac{CDI}{RfD} \quad (5)$$

HQ values for each exposure route were then summed together to produce a cumulative HQ value (separate for children and adults). To then characterize the total child or adult non-carcinogenic risk associated with multiple metals exposures, these cumulative HQ values for each metal were summed together to calculate an overall hazard index (HI) expressed as a unitless number, as shown in eqn (6):

$$HI = \sum_{i=1}^8 HQ_i \quad (6)$$

where  $HQ_i$  is the hazard quotient of the  $i^{\text{th}}$  of eight heavy metals in soil. For HI values  $\leq 1$ , adverse non-carcinogenic health effects are considered unlikely to occur, whereas HI values  $> 1$  indicate a potential for such health effects. The series of steps just described (eqn (2) through (6)) were carried out separately for child and adult risk characterization (using their unique parameters from Table 1), the results of which were added together to yield a total HI value. These total HI values were projected onto a city map at the Census tract level in order to give a spatial representation of non-carcinogenic risk. Of note, the extent to which HI values exceed one is not necessarily proportionate to potential health impacts, although higher values above unity are generally more concerning.

To derive child or adult lifetime cancer risk associated with each exposure route for each metal, the cancer risk for a given metal was calculated by multiplying child or adult CDI values for each exposure route by the cancer slope factor (CSF) for that metal and exposure route, in turn yielding a unitless value according to eqn (7):

$$\text{Risk} = CDI_i \times CSF_i \quad (7)$$

where risk represents a unitless probability that either a child or adult will develop cancer over a lifetime;  $CDI_i$  represents the chronic daily intake, or dose, of the  $i^{\text{th}}$  heavy metal (mg per kg per day); and  $CSF_i$  is the cancer slope factor of the  $i^{\text{th}}$  metal via

a particular exposure route, expressed as (mg per kg per day)<sup>-1</sup>. This equation converts the estimated daily intake averaged over a lifetime of exposure directly to a unitless risk of an individual developing cancer. As with non-carcinogenic risk, child and adult cancer risk were summed together to yield a total lifetime cancer risk.

To characterize the total lifetime cancer risk associated with multiple metals exposures, we considered a subset of five heavy metals that are considered carcinogenic according to the California Office of Environmental Health Hazard Assessment (OEHHA). These included Pb, As, Cr, Ni, and Cd. Risk values for each metal were summed together, as shown in eqn (8):

$$\text{Risk}_{\text{Total}} = \sum_{i=1}^5 \text{Risk}_i \quad (8)$$

where  $\text{Risk}_{\text{Total}}$  is the total lifetime cancer risk of exposure to all five heavy metals in the soil, and  $\text{Risk}_i$  is the lifetime cancer risk of exposure to the  $i^{\text{th}}$  of five heavy metals. According to the EPA, cancer risk above  $10^{-4}$ , or one cancer per ten thousand population, which corresponds to the upper-end of the EPA's generally acceptable risk range of  $10^{-6}$  to  $10^{-4}$ , is considered unacceptable.<sup>58</sup>

To obtain a unique HI value and cancer risk at the Census tract level, the previously described equations were applied separately for each Census tract using the average heavy metal concentrations measured across all samples collected within a given Census tract. RfDs and CSFs used in this analysis are presented in Table 2 and include those reported in the EPA's Integrated Risk Information System (IRIS).<sup>59</sup> For some elements, a range of values were reported. In these cases, we used the lower and upper end of the range of RfDs or CSFs in order to calculate separate lower and upper risk estimates (described below). Where IRIS values differed from California OEHHA values, we included the California-reported values when considering the full range of RfDs and CSFs.<sup>60</sup>

To calculate lower-bound HI values and cancer risks for each Census tract, we applied Table 2, making use of the lower values where ranges existed. An additional assumption was that 100% of measured chromium was of the less toxic and non-



Table 2 Reference doses (Rfd) and cancer slope factors (CSFs) for heavy metals

	Reference dose (mg per kg per day)			Cancer slope factors (mg per kg per day) <sup>-1</sup>		
	RfD <sub>ing.</sub>	RfD <sub>inh.</sub>	RfD <sub>derm.</sub>	CSF <sub>ing.</sub>	CSF <sub>inh.</sub>	CSF <sub>derm.</sub>
Pb	0.0035	—	0.0007	0.0085	0.042	0.425
As	0.0003	0.000004	0.00006	1.5–9.5 <sup>a</sup>	12	47.5
Mn	0.024–0.14	0.000014	0.0048–0.028	—	—	—
Cr(III)	1.5	—	0.3	—	—	—
Cr(VI)	0.003	0.000029	0.0006	0.42	510	2.1
Ni	0.02	0.000006	0.004	0.91	0.91	4.55
Cu	0.04	—	0.008	—	—	—
Cd	0.001	0.000003	0.0002	—	1.5–15	—
Zn	0.3	—	0.06	—	—	—

<sup>a</sup> CalEPA.

carcinogenic trivalent form (Cr(III)). In contrast, the calculation of upper-bound estimates made use of Table 2 while applying the upper values where ranges existed. The upper estimate assumed that all chromium was of the more toxic and carcinogenic hexavalent form (Cr(VI)). Our assumption of either 0% or 100% hexavalent chromium for the two scenarios was due to our not knowing the concentrations of these respective forms in the soil of our study area and due to our not being able to identify a common ratio of the these two forms in typical soil, particularly near our region of interest. Thus, we felt this approach was the best way to estimate the potential range of soil contamination and related risk.

In calculating health risk, non-carcinogenic toxicity is understood to arise only above a discrete exposure level, or threshold, whereas carcinogenicity is considered to be non-threshold and therefore exhibits effects linearly even at the lowest doses. These distinctions underly the applications of either a reference dose (non-cancer) or cancer slope factor (cancer). For Pb, while no safe threshold of exposure has been identified for non-carcinogenic risk, we assumed an RfD of 0.0035 mg per kg per day, which is commonly applied in the literature.<sup>41,61</sup> While it was important to include Pb in our assessment of non-carcinogenic risk, this assumption of a threshold response may yield a conservative estimate for Pb-related health risk.

To convert oral toxicity values to dermal values, since oral values are derived from potential (*i.e.* administered) doses that don't account for gastrointestinal absorption, we applied conventional methods that adjust for an assumed GI absorption of 20% across inorganic chemicals.<sup>62</sup> In the case of inhalation, we converted reference concentrations (RfCs) to RfDs using basic arithmetic that assumed an average inhalation rate of 20 m<sup>3</sup> per day and body weight of 80 kg.<sup>62</sup> Since converting to child-specific RfDs using child exposure values is often incorrect given the methods used to derive original RfCs,<sup>62</sup> we extended our adult-derived RfDs to child exposure scenarios in the present analysis. While this approach can foreseeably create uncertainty for child-related exposure, sensitivity analyses using adult-only conversions *versus* conversions unique to children and adults confirmed this decision to be of negligible importance as final risk estimates were virtually unchanged.

### 3. Results

#### 3.1 Spatial patterning of heavy metal soil concentrations

Depicted in Fig. 1 is a map showing interpolated soil concentrations of eight heavy metals derived from kriging. Results relating to Pb concentrations, first published in our original study by Masri *et al.* (2020), illustrate the highest concentrations in the northeastern quadrant and central region of the city, which corresponds with the downtown area of Santa Ana. By comparison, As and Mn exhibited east–west gradients, with higher As concentrations in the east and the opposite pattern observed for Mn. Cr, Cd, and Zn showed similar patterns as Pb, albeit with high concentrations extending southeast all the way to the state route 55 freeway. Cu exhibited similar patterns as Cr, Cd, and Zn, however Cu concentrations reached their highest in the southeast quadrant. Of note, the southeast quadrant of Santa Ana is also adjacent to the John Wayne Airport (see Fig. S1†).

#### 3.2 Descriptive statistics & soil screening levels

Table 3 presents summary statistics for eight soil heavy metal samples and the extent to which either U.S. EPA or California EPA residential soil screening levels (SLs) for the protection of children were exceeded.<sup>60,63</sup> Of note, the mean, median, S. D., Min, and Max values presented for Pb in Tables 3 and 4 are excerpted from our first Santa Ana-based soil study by Masri *et al.* (2020). Where cancer and non-cancer SLs were available, the more protective standard was included in the table. Metals for which at least 1% of samples exceeded federal SLs included Pb (3.9%), As (91.1%), Cr (100%, assuming Cr(VI)), and Ni (100%). CalEPA SLs, which were generally more stringent, were exceeded by Pb (48.2%), As (100%), Cr (100%, assuming Cr(VI)), and Cd (32.6%). For Cr, the proportion of SL exceedances changed drastically depending on whether measured Cr was assumed to be Cr(III) or Cr(VI). Assuming that all measured Cr is the less harmful trivalent form yields 0% exceedance across all samples, compared to 100% exceedance when we assume that all samples are of the hexavalent form.

As a proportion of Pb samples obtained within a single landuse category, roadway, industry and residential samples each





Fig. 1 Spatial distribution of eight heavy metals in the soil of Santa Ana, CA in 2018.

exceeded the 80 ppm California SL at a frequency of approximately 52%, compared to 22% of samples and zero samples exceeding the SL in park and school areas. The 400 ppm federal

SL was exceeded most often by samples obtained in the roadway (12%), compared to residential (4%), industrial (3.4%), park (0.6%), and school (0%) landuse types.



**Table 3** Summary statistics relating to the concentration (ppm) of eight heavy metals measured across 1528 soil samples in Santa Ana, CA, and their associated state and federal screening levels (SLs)<sup>a</sup>

	Pb	As	Mn	Cr	Ni	Cu	Cd	Zn
Min	11.4	0.1	270.1	19.3	16.0	19.7	0.1	76.8
50 <sup>th</sup>	77.8	6.6	646.9	61.2	37.5	56.4	1.4	266.9
Mean	123.1	8.3	663.4	64.9	38.7	67.6	1.7	328.0
Max	2687.0	174.8	8774.0	279.1	170.3	1950.0	23.9	3390.0
S.D.	181.3	9.3	236.0	20.3	9.7	79.6	1.4	233.3
Screening level (SL)				Cr(III)	Cr(VI)			
USEPA	400	0.68 <sup>b</sup>	1800	120 000	0.3 <sup>b</sup>	3100 <sup>b</sup>	71	23 000
CalEPA	80	0.07	—	100 000	0.3 <sup>b</sup>	3000	1.7	23 000
% exceeding SLs								
USEPA	3.9	91.1	<1	0	100	0	0	0
CalEPA	48.2	100.0	—	0	100	0	32.6	0

<sup>a</sup> CalEPA SL values come from CalEPA human-exposure-based screening numbers developed to aid the estimation of cleanup costs for contaminated soil<sup>60</sup> while USEPA SL values come from USEPA's updated 2020 table of regional screening levels.<sup>63</sup> <sup>b</sup> Value for carcinogenic outcomes.<sup>63</sup>

**Table 4** Average (standard deviation) soil concentrations (ppm) of heavy metals by landuse type in Santa Ana, CA<sup>a</sup>

	N <sup>b</sup>	Pb	As	Mn	Cr	Ni	Cu	Cd	Zn
Industry	89	122.6 (164.8)	8.4 (6.2)	<b>736.0 (866.0)</b>	<b>72.1 (25.0)</b>	<b>50.0 (16.0)</b>	<b>291.8 (221.6)</b>	<b>2.8 (3.5)</b>	<b>449.3 (342.7)</b>
Park	161	72.5 (75.3)	7.1 (6.3)	647.8 (128.0)	56.0 (10.9)	37.8 (9.1)	44.4 (14.7)	1.2 (0.5)	201.2 (79.8)
Residential	1173	128.4 (187.9)	8.3 (8.5)	661.0 (109.5)	65.7 (20.3)	37.6 (7.9)	64.4 (32.2)	1.7 (1.1)	338.7 (236.8)
Roadway	76	<b>172.9 (251.1)</b>	<b>11.9 (21.6)</b>	658.9 (127.3)	66.0 (25.4)	43.7 (16.7)	64.0 (34.6)	1.6 (0.9)	313.9 (142.4)
School	10	37.9 (12.9)	9.8 (16.2)	527.9 (40.3)	52.8 (5.6)	34.0 (6.3)	48.1 (14.2)	0.88 (0.5)	279.6 (175.6)
All	1528	123.1 (181.3)	8.3 (9.3)	663.4 (236.0)	65.0 (20.3)	38.7 (9.7)	67.6 (79.6)	1.7 (1.4)	328.0 (233.3)

<sup>a</sup> The landuse type with the highest mean concentration is shown in bold for each column. <sup>b</sup> N, number of samples.

For As and Cd, nearly all and roughly one-third of samples, respectively, exceeded the California SL, compared to 91% and 0% exceedances when considering the federal SL. For As, average concentrations were highest near roadways, but also showed relatively high levels near schools (though school samples were limited to  $n = 10$ ). As a fraction of samples collected within a single landuse type, little variability in As was seen (90–96% of samples exceeding federal SL in each landuse type). Cd showed the highest average concentrations within the industrial landuse type. Samples within this landuse type also exceeded the California SL at the highest frequency (56.2% of samples), compared to residential (33.7%), roadway (32.9%), park (13.7%), and school (0%) samples. Zero Cd samples exceeded the federal SL. For the remaining metals analyzed, the highest average concentrations were measured within

industrial landuse types, and the percent exceedance (see Table 3) of state and federal SLs were binary (0% or 100% of samples exceeding the SL) depending on which SL was considered. Table 4 presents the average concentration of each heavy metal according to landuse type.

Although some metals did not exceed state or federal SLs standards, they were nonetheless found to be enriched in the soil relative to background levels. Table 5 summarizes these findings by showing the percent of samples that were enriched in each of eight heavy metals based on the enrichment factor values that were calculated (using eqn (1)) for each of the 1528 soil samples (using Fe as reference element). Of all metals assessed, those that were moderately or more enriched in the greatest proportion of samples included Zn (~65% of samples) followed by Pb (~50% of samples), with approximately 10% of samples (in both cases)

**Table 5** Distribution (%) of enrichment factor values (based on eqn (1)) for 1528 soil samples across five-category enrichment factor index<sup>a</sup>

	Pb	As	Mn	Cr	Ni	Cu	Cd	Zn
No or minimal enrichment	50.2	71.2	99.8	99.1	99.8	92.2	99.7	34.6
Moderate enrichment	38.2	25.7	0.1	0.9	0.2	6.8	0.3	56.5
Significant enrichment	10.2	3.0	0.1			0.9		8.8
Very high enrichment	1.1	0.1				0.1		0.1
Extremely high enrichment	0.3							

<sup>a</sup> Fe was used as reference element.





showing “significant” enrichment. Arsenic showed moderate or greater enrichment across approximately 29% of samples, with roughly 3% showing “significant” enrichment. By comparison, Cu showed <10% enrichment, whereas Mn, Cr, Ni, and Cd showed virtually no enrichment.

When comparing metal samples in terms of the proportion of SL exceedances and the proportion of enrichment, Pb and As showed both substantial enrichment and SL exceedance. Similarly, Mn and Cu showed agreement in that enrichment and SL exceedances were both found to be minimal. For Cr and Ni, the proportion of SL exceedances changed dramatically depending on the SL used for comparison. Zn was the only metal in which substantial enrichment was found despite not exceeding state or federal SLs. Results were similar when using Al as the reference element instead of Fe, as shown in Table S1.†

Concentration means for each heavy metal were higher than their medians. This, combined with an abundance of outliers above the mean, suggests that the concentration distribution of soil metal was consistently positively skewed. Concentration boxplots for all eight heavy metals in are depicted in Fig. S2† of the ESI.

### 3.3 Social patterning of heavy metal soil concentrations

Fig. 2 presents average metal concentrations and 95% confidence intervals (CI) by median household income at the Census tract level (tertiles). Average metal concentrations decreased with increasing income bracket for seven of eight metals assessed, the exception being for Mn where the opposite trend was observed as Mn concentrations tended to be higher in the western side of the city. Differences in average concentrations between tertiles were statistically significant ( $p < 0.05$ ) for each tertile of Pb, Zn, and Cd, and statistically significant for the first and third tertiles of Mn and Cr.

The largest disparities existed for Pb, Zn, Cd and As, where on average, soil samples obtained in Census tracts where the median household incomes were below \$50 000 had 390%,

92.9%, 56.6%, and 54.3% higher concentrations relative to samples obtained in Census tracts where median household incomes were above \$100 000, respectively. Note, the portion of Fig. 2 relating to Pb concentrations was originally published in our first paper on soil lead in Santa Ana.<sup>37</sup>

Soil samples from Census tracts in the tertile with the lowest proportion of college educated residents had 87.0% and 26.5% higher Pb and Zn concentrations on average relative to those in the highest tertile, respectively. Similarly, soil samples collected in the tertile with the highest proportion of renter occupied housing units had 75.2%, 17.4%, 38.1%, 19.1%, and 33.4% higher Pb, As, Cu, Cd, and Zn concentrations, respectively, compared to samples from the lowest tertile.

Soil samples from the tertile with the highest proportion of residents without health insurance had 96.1%, 17.1%, 14.1%, and 31.9% higher Pb, As, Cd, and Zn concentrations, respectively, and those from the tertile with the highest proportion of children (under age five) had 90.0%, 11.1%, and 27.3% higher Pb, Cd and Zn concentrations, respectively, than to those from the lowest tertiles.

In terms of social patterning, relative to soil samples collected from the lowest tertiles, soil samples collected in tertiles with: the highest proportion of non-English speaking residents had 66.1% and 24.7% higher Pb and Zn concentrations, respectively; the highest proportion of immigrant residents had 96.4%, 18.8%, 20.9%, and 32.4% higher Pb, Cu, Cd, and Zn concentrations, respectively; the highest proportion of Latina/o/x or Hispanic residents had 105.1%, 17.0%, 20.6% and 32.6% higher Pb, As, Cd, and Zn concentrations, respectively. In contrast, the opposite pattern was observed for Mn where the tertile with the lowest proportion of Latina/o/x or Hispanic residents had 11.5% higher Mn concentrations relative to those from the lowest tertile.



Fig. 2 Average heavy metal concentrations and 95% CIs (shown as error bars) of heavy metal soil samples based on the median household income across Census tracts.



### 3.4 Socio-spatial distribution of heavy metal concentrations and health risks

Correlation coefficients of demographic characteristics plotted against lower- and upper-bound estimates of non-carcinogenic (HI values) and carcinogenic risk at the Census tract level ( $n = 61$ ) showed moderate ( $|r| \geq 0.3$ ), statistically significant ( $p < 0.05$ ) positive correlations between non-cancer risk (both lower and upper estimates) and the percent of residents under five years of age ( $r = 0.43$  to  $0.46$ ), the percent of renter-occupied housing units ( $r = 0.41$  to  $0.42$ ), as well as the overall vulnerability index ( $r = 0.31$ , lower risk scenario only). Although statistically significant, the correlation between the upper-estimated non-carcinogenic risk and vulnerability index fell just below ( $r = 0.29$ ) our definition of “moderate” correlation. Statistically significant positive correlations were also found between lower-estimated cancer risk and the percent of residents under five years of age ( $r = 0.31$ ) and the percent of Hispanic residents ( $r = 0.30$ ). A statistically significant, near-moderate correlation ( $r = 0.29$ ) was found between this cancer risk and the percent of renter-occupied housing units. Moderate and significant correlations also existed between average Pb concentrations and the percent of residents under age five ( $r = 0.51$ ), median household income ( $r = -0.38$ ), the percent of renter occupied housing units ( $r = 0.46$ ), and overall vulnerability index ( $r = 0.42$ ). Although significant, the correlation between Pb and the percent of Hispanic residents was just under moderate ( $r = 0.29$ ).

Moderate and significant negative correlations were found between lower- and upper-estimated cancer risk ( $r = -0.38$  to  $-0.30$ ) and the percent of immigrants, as well as between the percent of immigrants and As ( $r = -0.39$ ) and Cr ( $r = -0.39$ ) concentrations. When divided according to the two most

predominant immigrant populations (Latina/o/x and Asian), the percent of Asian residents showed similarly negative (and even stronger) correlations with these variables, while the correlation for Latina/o/x immigrants showed weak and non-significant correlations. Table S2† presents all correlation coefficients relating to average heavy metal concentrations, demographic characteristics, and estimated health risk at the Census tract level.

Shown in Fig. 3 is a map depicting Santa Ana Census tracts according to HI scores that were derived using both lower and upper RfD values (see Table 2) where applicable (Mn only). As shown, depending on whether upper or lower RfDs were applied for Mn, between 35 and 61 (57–100%) Census tracts in Santa Ana showed  $HI \geq 1$ , implying the potential for non-carcinogenic health effects across the majority of Census tracts. The pattern of risk was similar across both lower and upper risk scenarios, with the cluster of Census tracts in the central region of the city, just south of the I-5 and SR-22 freeways, showing the highest HI values. When assessing the average concentration of each heavy metal across all sampling points, the cumulative HI for Santa Ana was calculated to be 1.3 under lower risk assumptions. When upper RfD values were assumed, a cumulative HI of 2.0 was calculated, which is twice as high as the threshold ( $HI = 1$ ) for non-carcinogenic health effects. The HI for the city was driven overwhelmingly by exposure incurred through childhood ( $\sim 9$  times higher than adults) under both upper and lower RfD scenarios, since children have substantially higher soil ingestion rates and lower body weights. The calculated hazard quotients (child and adult risk combined) associated with each metal decreased in the order  $Pb > As > Mn > Cr > Ni > Cd > Cu > Zn$  under the upper RfD scenario and in the order  $Pb > As > Mn > Ni > Cd > Cu > Zn > Cr$  under the lower RfD scenario, with ingestion and dermal exposure being the predominant



Fig. 3 Map of Santa Ana and the hazard index for each Census tract.





Fig. 4 Cancer risk per 10 000 population for each Census tract in Santa Ana.

exposure routes under both scenarios, accounting for approximately 80% and 17% of the HI, respectively. A full list of HI and HQ values corresponding to Fig. 3 for children and adults, and according to each exposure route, can be found in Table S3.†

Fig. 4 presents the lower- and upper-estimated cancer risk per 10 000 population at the Census tract level. As shown, the application of lower CSFs, including the assumption that all chromium is of the non-carcinogenic form (Cr(III)), yields cumulative cancer risks that are above the upper-end of EPA's generally acceptable risk range ( $10^{-6}$  to  $10^{-4}$ ) for 10 of 61 (~16%) Census tracts. When using upper CSFs, and assuming all Cr is of the carcinogenic form (Cr(VI)), all 61 (100%) Census tracts in the city showed cancer risks in excess of the EPA's generally acceptable risk range. When assessing the average concentration of each carcinogenic metal across all sampling points, the cumulative cancer risk for Santa Ana was calculated to be between  $8.5 \times 10^{-5}$  and  $2.7 \times 10^{-4}$ , depending on which set of assumptions were applied. In the case of the latter value, the cancer risk equates to nearly three-times that which the EPA considers generally acceptable. Cancer risk was mostly driven by exposure during childhood, which showed a cumulative risk of approximately three-times that of adults. In terms of each metal, arsenic accounted for the majority of the cancer risk, with a risk that was nearly three-times higher than the next two most high-risk metals (upper CSF scenario). Overall, the calculated risk related to each metal decreased in the order As > Ni > Cr > Pb > Cd under the upper CSF scenario and in the order Ni > As > Pb > Cd under the lower CSF scenario. As with non-carcinogenic risk, ingestion and dermal exposure were the predominant exposure routes under both scenarios, accounting for approximately 80% and 20% of the cumulative cancer risk, respectively. Note that Cr does not show up in the lower scenario ranking since that scenario assumed that all chromium was of the non-carcinogenic form. A full list of cancer risk values

corresponding to Fig. 4 for children and adults, and according to each exposure route, can be found in Table S4.†

Cumulative risk, as well as the order of individual metal-related risk, was heavily affected by the specific cancer slope factor used for As. When using an oral CSF of  $1.5 \text{ (mg per kg per day)}^{-1}$  listed by the U.S. EPA, as opposed to the more protective value of  $9.5 \text{ (mg per kg per day)}^{-1}$  recommended by California Office of Environmental Health Hazard Assessment, overall cancer risk for Santa Ana was calculated to be lower ( $1.2 \times 10^{-4}$ ), albeit still above the upper-end the EPA's generally acceptable risk range. Under this scenario, the relative risk of each metal decreased according to Cr > Ni > As > Pb > Cd. The individual cancer risk remained above the acceptable risk level ( $10^{-4}$ ) for all but seven Census tracts.

Fig. 5 is excerpted from the supplemental section of our prior published study in order to showcase the Census tracts in Santa Ana that ranked highest in terms of their social and economic vulnerability to soil heavy metal exposure.<sup>37</sup> A comparison with Fig. 3 and 4 reveals similar patterning in that the region with the greatest potential health risk based on our risk assessment calculations is also the region where social and economic vulnerability is the greatest; namely, the central region of the map which coincides with downtown Santa Ana. Of note, all freeways and interstate roads surrounding Santa Ana that are described in this paper can be viewed in Fig. 5. Additionally, maps relating to landuse/zoning, total population and household income at the Census tract level are presented in Fig. S3 and S4† of the ESI.

## 4. Discussion

This study assesses the health risk and spatial distribution associated with soil concentrations of eight heavy metals in the





Fig. 5 Santa Ana Census tracts according to their vulnerability index scores.

city of Santa Ana (Southern California) in order to calculate a cumulative hazard index score and cancer risk across the region and within Census tracts. Metal concentrations yielded a high spatial variance, with 57% to 100% of Census tracts showing the potential for non-carcinogenic health outcomes ( $HR > 1$ ) and 16% to 100% showing high risk for carcinogenic outcomes ( $risk > 10^{-4}$ ), depending on whether lower or upper toxicity values were used.

In the case of Pb, nearly half of samples exceeded the California safety recommendation of 80 ppm for soil Pb in play areas. For As, all samples exceeded the state screening level. Other metals such as Cr, Ni, and Cd exceeded at least one screening level, however Cr-related exceedances depended on whether Cr(III) or Cr(VI) was assumed.

Within residential areas, over half of the soil samples had Pb levels above the California EPA safety guideline for Pb in child play areas, and 4% had levels greater than the 400 ppm U.S. EPA standard for play areas. For As and Cd, nearly all and roughly one-third of samples, respectively, exceeded the California SL.

Compared to risk during adulthood, risk during childhood was approximately 9-times higher for non-carcinogenic risk and 3-times higher for carcinogenic risk. This finding is similar to a recent study by Chonokhuu *et al.* (2019) who assessed similar heavy metals in the soil and found childhood risk to be 2- to 9-times higher than that of adults, and similar to results by Gržetić & Ahmed Ghariani (2008) who calculated childhood risk to be 10-times greater than that of adults.<sup>41,42</sup>

These findings are relevant to childhood exposure given that children often play in residential areas. For lead, one Census tract housing more than 650 children under age five had mean

Pb levels above the 400 ppm U.S. EPA standard. Further details on potential child-related Pb exposure in Santa Ana can be found in our prior work (Masri *et al.* 2020). Generally speaking, Census tracts that had a greater proportion of children tended to have higher concentrations of Pb, Cd, and Zn and higher HI scores, implying a greater potential for non-carcinogenic risk. These results underscore a critical public health problem since children are particularly vulnerable to the adverse impacts of metals such as Pb.<sup>9-11</sup> Contaminated soil and the resuspension of soil Pb have been shown to be important contributors to the burden of blood Pb in children.<sup>64-66</sup>

For Pb, soil concentrations from this study were similar to recent reports by Johnston *et al.* (2019) and Wu *et al.* (2010), as described in our prior work.<sup>37,67</sup> Other metals were similarly within the general ranges reported in urban environments across the U.S.<sup>38,39,68</sup> Relative to background soil metal concentrations reported across the U.S. and in California, concentrations reported in this study were elevated by a factor of approximately 6- to 18-fold for Pb, 1.6-fold for As, 2-fold for Cu, 2- to 6-fold for Cd, and 5- to 9-fold for Zn, whereas Cr and Ni concentrations were within the approximate background ranges reported in the literature.<sup>68,69</sup> These findings are generally consistent with our assessment of heavy metal contamination based on enrichment factors, which found Pb, As, and Zn to be most heavily enriched in the city of Santa Ana, relative to local baseline samples.

Additionally, findings suggest increased exposure-related vulnerability among residents of lower socioeconomic statuses. Moderate and statistically significant positive correlations existed between non-cancer risk and the percent of





The southwest quadrant of Santa Ana is also adjacent to the John Wayne Airport, which may contribute to the accumulation of metals in the soil. Copper in particular was found in greatest

Also of note when interpreting our results is that this study did not take into consideration the ingestion of heavy metals through the dietary route. Had we considered this additional exposure pathway, our calculated chronic daily intake levels of heavy metals would have been greater, resulting in higher estimated risk (particularly for metals such as Pb, As, and Cd which have been widely documented in various foods<sup>100–102</sup>). In this regard, while our analysis likely overestimates some aspects of heavy-metal-related health risk through the soil (e.g. use of “upper percentile” ingestion for children), the absence of

## Author contributions

Conceptualization, S. M., A. L., E. V., A. R., J. W.; methodology, S. M., A. L., J. W.; software, S. M.; validation, S. M., M. L.; formal analysis, S. M.; investigation, E. R.; resources, E. V.; data curation, S. M., M. L.; writing—original draft preparation, S. M.; writing—review and editing, S. M., A. L., E. V., M. L., J. W.; visualization, S. M., A. L., J. W.; supervision, J. W.; project administration, J. W.; funding acquisition, A. L., J. W. All authors have read and agreed to the published version of the manuscript.

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## Conflicts of interest

There are no conflicts to declare.

## 5. Conclusions

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