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Spatial predictors of heavy metal concentrations in epiphytic moss samples in Seattle, WA



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HIGHLIGHTS

GRAPHICAL ABSTRACT

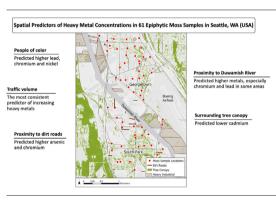
- Epiphytic moss is an emerging, costeffective, approach to identifying air pollution.
- Local youth collected 61 moss samples, analyzed for concentrations of 25 elements.
- We assessed the location-specific spatial predictors of heavy metal concentrations.
- Traffic, dirt roads, industrial corridor predicted higher concentrations.
- Tree canopy predicted lower metal concentrations.

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ABSTRACT

The use of bio-indicators is an emerging, cost-effective alternative approach to identifying air pollution and assessing the need for additional air monitoring. This community science project explores the use of moss samples as bio-indicators of the distribution of metal air particulates in two residential neighborhoods of the industrial Duwamish Valley located in Seattle, WA (USA). We applied geographically weighted regression to data from 61 youth-collected samples to assess the location-specific area-level spatial predictors of the concentrations of 25 elements with focus on five heavy metals of concern due to health and environmental considerations. Spatial predictors included traffic volume, industrial land uses, major roadways, the airport, dirt roads, the Duwamish River, impervious surfaces, tree canopy cover, and sociodemographics. Traffic volume surrounding sample locations was the most consistent positive predictor of increasing heavy metal concentrations of all metals, with statistically significant associations for chromium and lead in some areas. As the distance from dirt roads increased, the concentration of arsenic and chromium decreased significantly. Percent tree canopy within 200 m of sample locations was a significant protective factor for cadmium concentrations. In addition, percent people of color was significantly positively associated with increasing lead,

Abbreviations: AIC, Akaike information criterion; DIRT Corps, Duwamish Infrastructure Restoration Training; DVYC, Duwamish Valley Youth Corps; EPA, Environmental Protection Agency; GWR, geographically weighted regression; OLS, ordinary least squares; ICP-OES, plasma optical emission spectrometry; PAHs, polycyclic aromatic hydrocarbons; VIF, variance inflation factor. * Corresponding author.

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chromium and nickel concentrations. Our findings underscore the potential influence of heavy industry and mobile sources on heavy metal concentrations, the buffering potential of trees in local environments, and persistent opportunity to improve environmental justice.

1. Introduction

Air pollution exposure has a negative impact on multiple health outcomes, including cardiovascular and respiratory diseases, adverse birth outcomes, and mortality (Manisalidis et al., 2020). The Clean Air Act, the Right to Know Act, and other policies have developed in response to increasing knowledge of these impacts. However, there are segments of the population that remain exposed to harmful levels of air pollution. When considering areas in the United States (U.S.) subject to regulatory monitoring (primarily urban), non-Hispanic Black and Hispanic populations are disproportionately exposed to poor air quality (Mohai and Bryant, 2019; Woo et al., 2019).

Documenting and subsequently addressing disproportionate exposures is difficult when regulatory monitoring primarily focuses on regional scale criteria pollutants and is conducted at few stationary monitoring sites in metropolitan areas (Government Accountability Office, 2020). In many airsheds, the deposition patterns of particulate pollutants can be complex due to multiple sources which have significant deposition (and ambient concentration) gradients over relatively small distances (Varela et al., 2014). Networks of air-quality monitors in U.S. cities are too widely-spaced to characterize pollutant patterns, and the dispersion models regulators typically use to visualize smaller-scale patterns rely on emission inventories, and therefore do not incorporate unknown pollution sources. Although local-scale monitoring would be necessary to identify missing sources or smaller scale pollution gradients, air monitoring is typically costly in terms of knowledge and resources. Communities with disproportionate air pollution burden often seek accessible and accurate monitors, and these types of technologies, ranging from "bucket brigades" to wireless sensors, have been under development for decades (Idrees and Zheng, 2020).

The use of bio-indicators is an emerging, cost-effective, alternative approach to identifying air pollution and assessing the need for additional air monitoring. Lichen and moss are the most commonly used bioindicators, and dozens of studies link heavy metals accumulated in their tissues to levels measured in the atmosphere (e.g., see Berg and Steinnes, 1997; Fernández et al., 2007; Messager et al., 2021; Schröder et al., 2013). As both lichen and moss can integrate pollutants over long periods of time (i.e. six months to over a year), they are well-suited to measuring chronic low-levels of pollution, and can be at similar or below the detection limits of conventional air-quality monitors. They might also detect pollution sources that emit intermittently (Donovan et al., 2016). The effectiveness of this approach was demonstrated by Donovan et al. (2016) in Portland, OR using Orthotrichum lyellii (O. lyellii, Hook. & Taylor), a common moss in many Pacific Northwestern cities. This study led to discovery of undetected sources of cadmium pollution in residential areas of Portland, and a related study demonstrated that polycyclic aromatic hydrocarbons (PAHs) found in moss were elevated near traffic-sources, and were reduced in proximity to tree canopy (Jovan et al., 2021). There is some evidence, locally, that moss can be used to assess fine-scale pollution patterns (Messager et al., 2021). The spatial-scale of urban bio-indicator studies (utilizing lichen or moss) is commonly limited by the scarcity of bio-indicators due to poor air quality, the heat island effect, and other stressors (Ares et al., 2012).

This project explores the use of moss samples as bio-indicators of the distribution of metal air particulates in the Georgetown and South Park neighborhoods of the Duwamish Valley in Seattle, WA. This study utilized a community science approach, and prior analyses have identified overall spatial patterns of metals found in moss, as well as internal and external validity (Jovan et al., under review). These neighborhoods, which serve as our study area, are situated on either side of the Duwamish River, the city's

main industrial core. In Seattle, lower-income populations, people of color, and immigrant populations are over-burdened by exposure with traffic-related air pollution (Schulte et al., 2015; Schulte et al., 2013; Su et al., 2010) and air toxics (Abel and White, 2011). These two neighborhoods in particular are home to thousands of residents previously identified as over-burdened with pollution sources (City of Seattle, 2018; Gould and Cummings, 2013; Jovan et al., under review; Washington State Department of Health, 2021). We sought to infer the spatial patterns and area-level determinants of the elemental components of air particulate pollutants within the study area by examining a dataset of the elemental concentrations found in moss samples collected from 61 sample sites. Using exploratory spatial data analyses, including geographically weighted regression (GWR) methods, we assessed the location-specific spatial predictors of the concentrations of each element, and we report in detail on five heavy metals of concern due to health and environmental considerations. Findings from our study will help characterize the potential influence of heavy industry and mobile sources on heavy metal concentrations, and the buffering potential of trees; evidence necessary to address environmental justice issues that plague these types of neighborhoods.

2. Methods

2.1. Study area and partners

The Georgetown and South Park neighborhoods of Seattle are located along the shores of the Duwamish River (see Fig. 1). The river's historic and present industrial activity, active port, and proximity to an airport, railway lines, and major highways all contribute to the contamination of the study area. The U.S. Environmental Protection Agency (EPA) designated a five-mile segment of the Duwamish River that transects the study area as a Superfund site in 2001 due to the presence of arsenic, carcinogenic polycyclic aromatic hydrocarbons, volatile organic compounds and polychlorinated biphenyls (U.S. Environmental Protection Agency, 2014).

The study area (population 4745) contains 2239 housing units, including single and multi-family residences. The neighborhoods comprise two census tracts which house a higher percentage of Hispanic residents than the City of Seattle (20.2% in Georgetown and South Park, compared to 6.7% in the City of Seattle), a higher percentage on non-White residents (53.4%, compared to 36.2%), a higher percentage of people who speak a language other than English at home (28.1%, compared to 21.3%), a higher percentage of people living below the poverty line (23.2%, compared to 11.0%), and a higher percentage of residents 25 and older with less than a high school diploma (16.9%, compared to 5.4%) (U.S. Bureau of the Census, 2019).

This study is the product of a community science (Charles et al., 2020) collaboration between over 15 partners, including local organizations and non-profits, residents, city and federal agencies, and universities. The community science approach prioritized co-producing actionable scientific knowledge, with adult and youth community members leading many project phases. The collaboration is described in detail in Derrien et al. (2020) and Jovan et al. (under review). Key partners in data collection included the Duwamish Valley Youth Corps (DVYC), Duwamish Infrastructure Restoration Training (DIRT) Corps, and the U.S. Forest Service.

2.2. Sampling and data collection

Sampling protocols, laboratory preparation and analysis methods, and quality control procedures are described in detail elsewhere (Derrien et al., 2020; Jovan et al., under review) and were based on protocols from prior studies (Donovan et al., 2016; Gatziolis et al., 2016). We used a

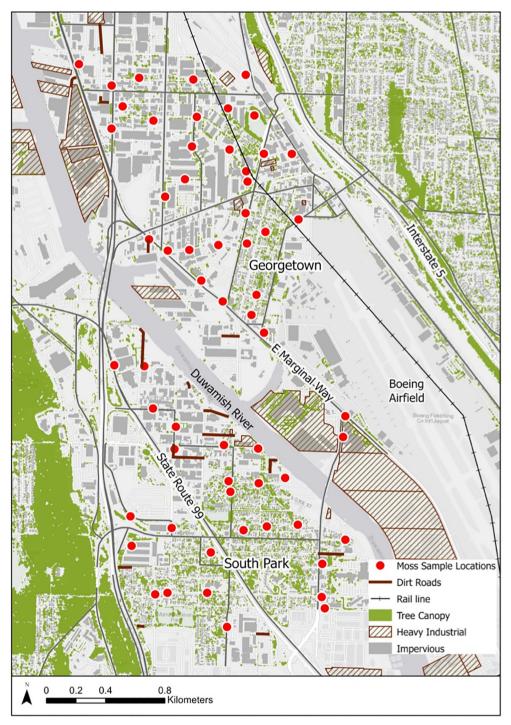


Fig. 1. Overview Map of Study Area, Land Features and Sample Locations (N = 61) in Seattle, WA - Georgetown and South Park neighborhoods (2019).

"train-the-trainer" approach where scientists experienced in leading moss studies trained leaders of the DVYC and DIRT Corps, who then trained all youth DYVC participants (Derrien et al., 2020). DVYC and DIRT Corps participants collected moss samples on four warm, dry days in late spring of 2019 using a quarter kilometer sampling grid (250 m \times 250 m) across the study area. Participants collected *O. lyellii* moss at the nearest tree to the centroid of each grid cell. Most moss samples were collected from trees on streets, but several samples were collected from trees near rivers and in parks.

The DVYC and DIRT Corps led initial sample preparation following laboratory protocols. Samples were then prepared and analyzed, including quality control measures, at the US Forest Service Grand Rapids, MN analytical chemistry laboratory. Full sampling and analysis methods are described in Jovan et al. (under review). Plasma optical emission spectrometry (ICP-OES) was used to quantify the concentrations of 25 elements. We excluded six macronutrients because they are not directly related to pollution, and conducted exploratory analyses on 19 elements, including: aluminum (Al), arsenic (As), barium (Ba), boron (B), cadmium (Cd), chromium (Cr), cobalt (Co), copper (Cu), iron (Fe), lead (Pb), manganese (Mn), molybdenum (Mo), nickel (Ni), silicon (Si), sodium (Na), strontium (Sr), titanium (Ti), vanadium (V), and zinc (Zn). Chromium was not speciated to determine the Cr(VI) fraction (the valence that is highly toxic to humans). Replicate samples (collected at 18 of 61 sampling locations) were used to ensure that measurements were repeatable. Expert samples (collected at 17 of 61 sampling locations) showed acceptable statistical agreement (Derrien et al., 2020).

In this study, we focus on five heavy metals (out of the 19 elements) which are associated with negative impacts on the environment and human health: arsenic, cadmium, chromium, lead, and nickel (hereafter referred to as "priority" metals). Our rationale for selection of these five metals is described in Section S2.2. We made analytical decisions based on preliminary analyses of the priority metals, and we focus our presentation and discussion of results on these metals (although results for all 19 elements are shown).

2.3. Measures

2.3.1. Spatial factors

We estimated various potential spatial determinants of heavy metal concentrations in and near the study area. Variable specifications and data sources are listed in Table S1. Heavy industrial land use could be a source or associated with dispersion of air toxics. We calculated the percent of these land uses within a 500 m buffer surrounding each sample location. Traffic- or goods transport-related emissions could also contribute to metal concentrations found in moss. We therefore calculated the distance (in meters) from each sample location to the nearest major "roadway" (arterial or highway), and the average daily traffic volume (average weekday count) within a 500 m buffer. We chose to calculate traffic volume within a 500 m distance because previous research has demonstrated that traffic-related air pollutants can travel at least 300 m from trafficked-roadways (Zhu et al., 2002). In addition, traffic volume within a smaller buffer size (e.g. 200 m) was highly correlated with the distance from roadway variable. We also calculated the distance (in meters) from each sample location to the nearest airport facility, dirt road, and the Duwamish River, which contains port facilities. Only roads with complete dirt or gravel surface, and not those only with dirt or gravel shoulders, were included in the dirt road dataset. Finally, vegetation can remove gaseous and particle air pollution via uptake and deposition (Nowak et al., 2006). We therefore calculated percent tree cover within a 200 m buffer surrounding each sample location using a landcover dataset (University of Vermont Spatial Analysis Lab, 2016). We chose a 200 m buffer because trees have been shown to mitigate air pollutants, for example in a near-road environment, within close proximity (Tong et al., 2016).

2.3.2. Sociodemographic

We included block-group level percentage of residents with less than a high school education was estimated for each sampling site to assess the potential relationship between educational attainment and heavy metal exposure. We also included block-group level median household income as a proxy for economic status. In addition, we included block-group level percent people of color (defined as population not categorized as non-Hispanic White) as an important demographic indicator in the study area. We derived all sociodemographic data from the U.S. Census Bureau's American Community Survey 5-year Estimates for 2019.

2.4. Statistical analyses

We utilized several descriptive statistics and exploratory spatial data analysis methods to evaluate the varying spatial relationships between the elemental concentrations and neighborhood characteristics. We first computed descriptive statistics, including summary measures and Spearman correlation statistics. We then investigated the presence of spatial autocorrelation using Moran's I statistics (Anselin, 1995).

We then assessed the relationship between metal concentrations and spatial predictors using ordinary least squares (OLS) regression. As a sensitivity test, and to determine final model sample for each metal, we identified outliers using diagnostic plots and ran OLS regression models again with outliers removed.

Next, given the presence of spatial autocorrelation, we considered the local regression approach found in GWR models (Brunsdon et al., 1996)

to assess the spatially varying relationships between the elemental concentrations and the neighborhood characteristics. GWR models allow for the estimated relationship between the elemental concentrations and the neighborhood characteristics to vary over space within sample locations, as opposed to the global assumption of a fixed relationship provided by the naïve ordinary least squares approach. The form of the GWR model is as follows:

$$y = X\beta(u, v) + a$$

where *y* is a 61 × 1 vector of elemental concentrations measured at the 61 sample locations. The independent measures are represented in the matrix *X*, which has dimensions 61 × *p*, due to the *p* neighborhood characteristics considered. The residuals are captured in the vector ε , which are assumed to follow a Normal distribution with mean 0 and variance σ^2 . Most importantly, the local regression coefficients, β , are estimated at each specific location (*u*, *v*), which is the latitude and longitude of the collected sample for each element, respectively.

We first assessed simple spatially-varying bivariate relationships in an effort to establish the pool of potentially informative and meaningful relationships between each elemental concentration and the various neighborhood characteristics. To evaluate these bivariate relationships, we examined the mean, 1st, and 3rd quartile values of the GWR coefficient estimates for each neighborhood characteristic for the 61 sample locations. We selected neighborhood characteristics as potentially informative if the mean, 1st, and 3rd quartile of the bivariate GWR coefficient estimates all had either positive or negative values. Once the pool of objectively significant neighborhood characteristics was established, we assessed the variance inflation factor (VIF) to determine evidence of significant multicollinearity between characteristics (Chatterjee and Price, 1991). If the VIF value was larger than 10, this was evidence of significant multicollinearity and the variable was therefore removed from the list of final neighborhood characteristics for each metal. All regression analyses were conducted in R using the spgwr package (Bivand et al., 2013; Bivand and Piras, 2015).

We calculated summary statistics for the GWR results, including model fit statistics for all locally varying relationships, as well as the percent of the GWR estimates that were statistically significant (p < 0.05). We assessed both the R² and the corrected Akaike information criterion (AIC) (Brunsdon et al., 2000) model fit statistics for both the OLS and GWR models. We also assessed for GWR model improvement using an approximate likelihood ratio test using the bfc99.gwr.test() function in the spgwr package in R (Leung et al., 2000), in which a statistically significant finding based on the F-test indicates improved performance of the GWR over the OLS model (Devkota et al., 2014).

Finally, we present maps of the estimated GWR local regression coefficients and their associated statistical significance for the priority metals of interest. While there are many mapping approaches available to deal with the wealth of information that results from using locally-varying approaches like GWR models, we present maps of the GWR estimates along a shaded continuum of colors to indicate the strength of the varying associations, with a notation of those estimates that are statistically significant.

3. Results

3.1. Descriptive statistics

Fig. 1 provides an overview map of the study area for the 61 moss sample locations where the elemental concentrations were measured. Trees with sufficient moss to sample were more frequently located in residential areas, rather than in industrial areas, such as along the Duwamish River.

Descriptive statistics, including the Moran's I statistic, for concentrations (mg/kg) of the five priority metals found across the 61 moss locations are presented in Table 1. Statistics for the remaining elements are shown in Table S2. There appears to be a number of elements with over-dispersed concentrations, as evidenced by the larger measures of variability (standard

Table 1

Descriptive statistics and Moran's I of metal concentrations (mg/kg) found in moss.

Metal	Mean	SD	Median	Min	Max	Moran's I ^a
Arsenic (As)	1.2	0.7	1.0	0.4	3.2	0.12
Cadmium (Cd)	0.6	0.4	0.4	0.1	1.9	0.05
Chromium (Cr)	16.7	12.8	11.7	4.3	61.1	0.09**
Lead (Pb)	23.6	18.8	18.1	5.9	110.6	0.12***
Nickel (Ni)	7.9	7.9	6.2	2.1	59.0	0.06**

^a * p < 0.05; ** p < 0.01; *** p < 0.001.

deviation and range) compared to the mean values. The majority of the elements had Moran's I statistic values that were statistically significant (p < 0.05), with the exception of arsenic and cadmium of the priority metals and boron, copper, and strontium of the remaining metals. These significant Moran's I values indicate there was spatial autocorrelation, and supports the need to incorporate spatial dependence in the relationships between the elemental concentrations and the neighborhood characteristics.

Descriptive statistics of the neighborhood characteristics are provided in Table 2, where all characteristics also display significant spatial autocorrelation (p < 0.05) with the exception of distance from major roadway.

The correlation between the elemental concentrations found in moss is presented in Fig. S1. Generally, there were strong positive correlations among the 19 elemental concentrations. Cadmium and boron, which were strongly correlated with each other, were weakly (although still positively) correlated with the other elements. These findings mirror those in the previous study by Jovan et al. (under review), which found elements As, Cr, Co, Pb, Ni, Ca, Al, Ba, Cu, Fe, Mn, Mo, Si, Na, Sr, Ti, V, and Zn contributing to a main principal component which explained 76.5% of variation among priority metals, and cadmium and boron contributing to a second, non-significant, principal component.

3.2. Regression analyses

Based on bivariate relationships, we initially selected the following neighborhood characteristics as predictor variables in regression models for each metal: % tree (200 m), % heavy industrial (500 m), % impervious surface (500 m), traffic volume (500 m), distance from major roadway (m), distance from Duwamish River (m), distance from airport (m), distance from dirt road (m), median household income (\$, census block group), % less than a high school education (500 m), and % people of color (census block group). Correlations between neighborhood characteristics are also shown in Fig. S1. We found strong negative correlations between the % heavy industrial and distance from Duwamish River spatial predictors, as well as between % tree cover and % impervious surface cover. We therefore compared model fit between four models: 1) including all covariates, 2) including all covariates except % heavy industry, 3) including all covariates except % impervious surface, and 4) including all covariates except % heavy industry and % impervious surface. We selected Model 4 as our final model because it had the lowest VIF values (see Table S3) most consistent lower AIC and R² values except in comparison with Model 1 (see Table S4), and it avoids correlations between independent variables.

Table 2
Descriptive statistics of neighborhood characteristics.

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Therefore our final model for each metal adjusted for % tree cover, traffic volume, distance from major roadway, dirt roads, Duwamish River and airport, median household income, % less than high school education, and % people of color.

We ran an OLS regression model for each metal using this final variable selection. In addition, we ran OLS models after removing outliers as a test of sensitivity. Based on diagnostic plots, we removed one outlier from As, Cd and Ni, and we removed two outliers from Cr and Pb. Regression coefficients and standard errors for OLS models, with and without outliers, are shown in Table S5. While R^2 and AIC values suggest that removing outliers slightly improved model fit, removing outliers changed the magnitude of estimates slightly, and the direction of the mean differences in metal concentrations per unit increase in neighborhood characteristics was maintained, except for with Ni. Three covariates changed direction due to removal of one, the most extreme (Cook's distance = 1.0) outlier. Therefore, in the interest of maintaining sample size, we removed an outlier from the Ni model but not from the remainder of metals in GWR analyses.

We then fit GWR models for priority metals and remaining elements as a function of spatial neighborhood characteristic predictors. The GWR estimates represent the mean differences in metal concentrations (for priority metals shown in Table 3 and for remaining elements shown in Table S6) per unit increase in each neighborhood characteristic (standardized). When comparing model fit statistics between OLS models (in Table S5) and GWR models (Table 3), AIC values were approximately equivalent between OLS and GWR models for all priority metals. However, R² values were higher for GWR models than for OLS models, indicating that the GWR models could have improved explanatory power than the OLS models. *P*-values for the approximate likelihood ratio test (Table 3) of GWR models were less than 0.05 for all metals except for As, indicating that the GWR model was an improvement over the OLS model, and that averaged OLS associations should not be assumed to be constant across the study area.

The overall directional associations (as well as the percentage of varying estimated relationships that are statistically significant) are presented for the priority metals in Fig. 2, and for the remaining elements in Fig. S2. There were consistent inverse trends in the varying associations between % tree cover and metal concentrations, indicating that as tree cover increased surrounding sample locations, metal concentrations decreased. Distance from the airport and from the Duwamish River were also negatively associated with each of the priority metal concentrations, indicating that as the airport and River were further away from the sampled locations, the priority metal concentrations tended to be lower. There was also negative association between distance from dirt road and all metals except cadmium. The direction of association between distance from major roadway and metal concentrations was mixed.

Traffic volume and % people of color were consistently positively associated with priority metal concentrations. Median household income was inversely associated with all metals, indicating that with higher income, metal concentrations were lower. Percent with less than high school education was inversely associated with all metals except cadmium. While % with less than high school education is typically inversely related, there is

Exposure variable	Mean	SD	Median	Min	Max	Moran's I ^a
% Tree (200 m)	10.8	6.9	8.8	1.4	26.4	0.18***
% Heavy industrial (500 m)	4.6	7.7	1.5	0	36.9	0.14***
% Impervious surface (500 m)	75.6	15.3	83	45	95	0.36***
Traffic volume (500 m)	296,767	225,022	221,101	11,353	1,162,661	0.15***
Distance from major roadway (m)	68.2	74.4	37.5	1.3	335.3	0.04
Distance from dirt road (m)	375.3	285.5	291.3	5.6	1132.1	0.30***
Distance from airport (m)	845.7	470.6	860.8	13	1801.3	0.26***
Distance from Duwamish River (m)	683.8	356.5	562	189	1387	0.23***
Median household income	\$41,422	\$5482	\$40,450	\$36,216	\$52,727	0.28***
% < High school education	23.7	7.8	19	11	41	0.36***
% People of color	44.4	15.5	37.6	21.4	72.6	0.28***

^a * p < 0.05; ** p < 0.01; *** p < 0.001.

Table 3

Mean differences (standard deviation) in five priority metal concentrations per unit increase in neighborhood characteristics (standardized) for geographically weighted regression (GWR) model, and fit statistics for N = 61 sample locations^a.

Exposure variable	As	Cd	Cr	Pb	Ni*
% Tree (200 m)	-0.08 (0.07)	-0.10 (0.06)	-1.46 (1.85)	-1.75 (2.66)	-0.65 (0.71)
Traffic load (500 m)	0.36 (0.07)	0.10 (0.06)	7.16 (1.56)	5.50 (2.43)	1.34 (0.60)
Distance to major roadway (m)	-0.03 (0.06)	0.02 (0.05)	-0.06 (1.47)	0.69 (2.13)	-0.53 (0.59)
Distance to dirt road (m)	-0.33(0.11)	0.05 (0.10)	- 4.26 (3.06)	-4.38 (4.20)	-1.44 (1.13)
Distance to airport (m)	-0.25 (0.11)	-0.10 (0.09)	-6.64 (2.84)	-16.28 (4.1)	-1.64 (1.06)
Distance to waterway (m)	-0.17 (0.13)	-0.17 (0.11)	-5.58 (3.54)	-9.07 (4.93)	-1.51 (1.32)
Median household income	-0.13 (0.09)	-0.12 (0.07)	-1.96 (2.06)	-6.05 (3.15)	-1.13 (0.78)
% < High school education	-0.14 (0.09)	0.01 (0.08)	-3.06 (2.30)	-8.94 (3.48)	-1.35 (0.88)
% People of color	0.20 (0.12)	0.11 (0.10)	6.22 (3.00)	14.26 (4.39)	1.83 (1.13)
AIC	80.38	61.25	457.67	518.61	333.63
R ²	0.69	0.38	0.69	0.54	0.61
F	1.28	1.60	2.13	2.02	1.75
<i>p</i> -Value	0.20	0.05	0.01	0.01	0.03

^a One outlier removed from Ni model.

one census tract in the neighborhood in which they are positively related. It could be that residents in this neighborhood without high school diplomas are employed in high-paying jobs potentially related to a local industry.

The directional associations and percentage of relationships that were statistically significant for the remaining elements are presented in Fig. S2. As with the priority metals, traffic volume was a consistent positive predictor across all elements, and was statistically significant for all except molybdenum, copper, boron and cobalt. Distance from dirt road was consistently inversely associated with all elements, but was statistically significant for copper, aluminum, vanadium and cobalt. Greater distance from the airport was inversely associated with all elements except boron and cobalt, and % people of color was positively associated with all elemental concentrations except boron. Other spatial predictors showed varying relationships, by direction and statistical significance, throughout the study area.

Maps of the varying relationships between each of the priority metal concentrations and the various neighborhood characteristics are presented in Fig. 3a–e. As shown in Fig. 3a, % tree canopy within 200 m of the sample site was associated with stronger decreases in cadmium concentrations in residential areas of Georgetown. This pattern held true with arsenic, chromium and lead, but was not statistically significant.

Traffic volume was a statistically significant positive predictor throughout the study area for arsenic, chromium and lead (Fig. 3b). It was a significant positive predictor of cadmium primarily in the northeastern area of Georgetown close to Interstate-5. Traffic volume was also significantly negatively associated with nickel along the Duwamish River and throughout South Park.

The direction of association at different locations between distance from major roadway and metal concentrations was mixed, and none of the associations were statistically significant (Fig. 3c). The direction of association at different locations between distance from dirt road and metal concentrations was also mixed (Fig. 3d). Greater distance predicted statistically significant lower concentrations of arsenic (at all locations) and chromium (at some Georgetown locations). Finally, greater distance from the Duwamish River predicted lower concentrations of all metals, with statistically significant associations for chromium lead in South Park as well as in the south western area of Georgetown for lead (Fig. 3e).

4. Discussion

We employed a dataset built via a community-powered youth datacollection effort and applied exploratory spatial data analysis methods, including geographically weighted regression models, to determine the location-specific relationships between area-level spatial predictors and heavy metal concentrations found in moss. This study builds upon geographic descriptions of heavy metals distributions found in moss, described

Exposure Variable	Arsenic (As)	Cadmium (Cd)	Chromium (Cr)	Lead (Pb)	Nickel (Ni)
% Tree (200m)					
% significant	0%	3%	0%	0%	0%
Traffic volume (500m)			\rightarrow		
% significant	100%	21%	100%	100%	59%
Distance from major roadway (m)			<hr/>		
% significant	0%	0%	0%	0%	0%
Distance from dirt road (m)					
% significant	100%	0%	13%	0%	0%
Distance from airport (m)					
% significant	100%	0%	72%	100%	20%
Distance from Duwamish River (m)			_		
% significant	0%	0%	8%	33%	0%
Median household income	\leftarrow				
% significant	0%	0%	8%	54%	33%
% < HS education	—			—	
% significant	0%	0%	16%	100%	15%
% People of color			\Rightarrow		
% significant	0%	0%	54%	100%	15%

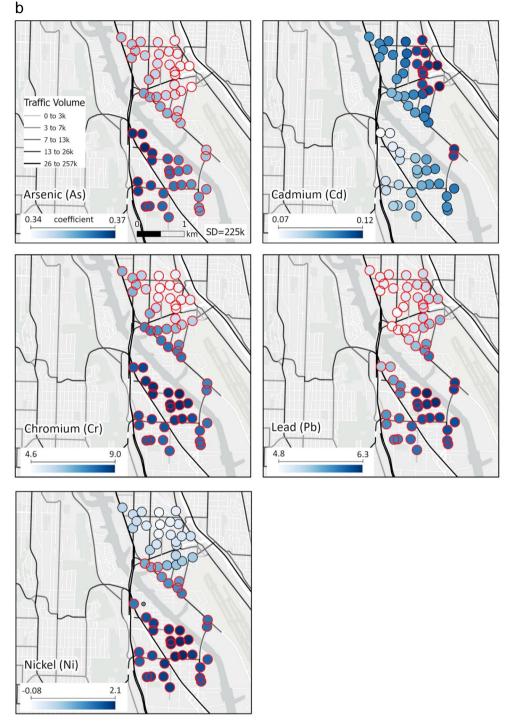
* one outlier removed from Ni model

Fig. 2. Geographically weighted regression (GWR) model estimate direction (of the mean) for five priority metals and percent of 61 sample locations* that are statistically significant * one outlier removed from Ni model.



* one outlier removed from Ni model

Fig. 3. a. Maps of geographically weighted regression (GWR) model coefficient values for heavy metals at 61 sample locations^{*} for the % tree canopy within 200 m predictor. Locations circled in red indicate p < 0.05 statistical significance. * one outlier removed from Ni model b. Maps of geographically weighted regression (GWR) model coefficient values for heavy metals at 61 sample locations for the traffic volume predictor. Locations circled in red indicate p < 0.05 statistical significance. c. Maps of geographically weighted regression (GWR) model coefficient values for heavy metals at 61 sample locations for the traffic volume predictor. Locations circled in red indicate p < 0.05 statistical significance. d. Maps of geographically weighted regression (GWR) model coefficient values for heavy metals at 61 sample locations for the distance from major roadways predictor. Locations circled in red indicate p < 0.05 statistical significance. d. Maps of geographically weighted regression (GWR) model coefficient values for heavy metals at 61 sample locations for the distance from dirt roads predictor. Locations circled in red indicate p < 0.05 statistical significance. e. Maps of geographically weighted regression (GWR) model coefficient values for heavy metals at 61 sample locations for the distance from Duwamish River predictor. Locations circled in red indicate p < 0.05 statistical significance. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)



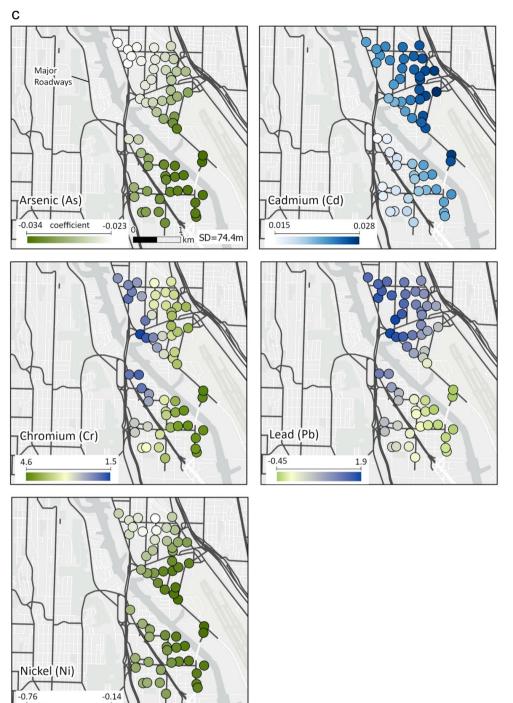


in Jovan et al. (under review), by identifying possible spatial drivers of these patterns. Our findings underscore the potential influence of heavy industrial corridor and mobile sources, as well as the buffering potential of trees in local air pollution environments.

When considering average effect across sample locations, greater traffic volume was consistently associated with higher concentrations of the five priority metals. Nickel is a product of diesel combustion (U.S. Environmental Protection Agency, 1984), and therefore higher concentrations would be expected near heavy traffic. We found that both higher traffic volume and roadway proximity significantly predicted higher nickel concentration along East Marginal Way, along 14th/16th Avenue South and near State Route 99, which are roadways with high levels of

truck-traffic. It could also be that there are nearby emissions related to oil combustion or metal processing in these areas. At the same time, greater tree canopy cover was protective for nickel concentrations in much of the same locations.

While nickel and arsenic are the main heavy metal by-products of fuel (diesel) combustion, statistically significant positive associations with traffic volume occurred throughout the study area also for chromium and lead, and in the northeastern area of Georgetown close to Interstate-5 for cadmium. Other heavy metals produced from vehicle traffic could come from tire wear (cadmium and lead), brake pad dust (chromium), asphalt pavement wear (nickel and chromium), road paint (lead) (Adamiec et al., 2016; Hong et al., 2020; Panko et al., 2018). It could also be that traffic is

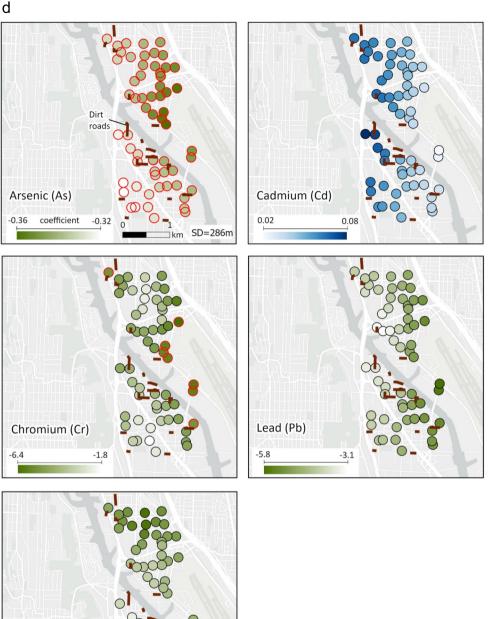




re-suspending dust containing these metals from other, past or current sources (Askariyeh et al., 2020).

Estimates for proximity to dirt roads provided other evidence that metals could be traveling via dust. Closer proximity to dirt roads was associated with higher concentrations of all metals except cadmium, on average. Statistically significant associations for arsenic existed throughout the study area. Closer proximity to dirt roads also predicted higher chromium concentrations (at some locations scattered on the north/east side of the Duwamish River). These findings contribute to other evidence (Jovan et al., under review) that heavy metal concentrations in moss were elevated near the industrial corridor, and were highly correlated with chemical elements indicative of fugitive dust (e.g. Al, Ca, Fe, Si, Sr, Ti). While our investigation relied on a windshield survey of public roadways, a high-quality landcover dataset specifying precise locations of bare earth/soil would be required to investigate potential influence of dust from industrial properties.

Closer proximity to the Duwamish River, which is lined with heavy industrial properties that are potential sources of air toxics emissions, was another consistent predictor of higher concentrations for all metals, on average, with significant associations for chromium in South Park and lead in both South Park and Georgetown. Close proximity to the Duwamish River predicted elevated lead concentrations especially along East Marginal Way in Georgetown, south of Michigan Street. Ship traffic may have contributed to metal pollution near the river through combustion of fuel oils





contaminated with nickel, or coal contaminated with nickel, lead, arsenic, cadmium, or chromium.

-0.58

Nickel (Ni)

-2.5

Another consistent finding was that greater tree cover within 200 m of sample sites was associated with lower metal concentrations in moss. In particular, tree cover was a statistically significant protective factor for nickel concentrations in residential areas in South Park. Urban forests have an established ability to reduce particulate air concentrations both at local and regional scales (Diener and Mudu, 2021), and strategic use of "green infrastructure" is a common mitigation tool in urban planning (Baldauf, 2017). Our results suggest that even modest tree canopy coverage within the studied neighborhoods (approximately 10%) was associated with lower levels of metal particulates pollution. However, only some of these associations were statistically significant, and it is not clear why the significant associations held only for cadmium and not other metals that travel via particulate matter. Larger studies involving greater variability in tree canopy cover are critical since vegetation is a modifiable factor that is relatively easy to manipulate.

Prior studies have established that Georgetown and South Park neighborhoods are overburdened with pollution (Min et al., 2019; Schulte et al., 2015). Our study builds on this finding to show that within these neighborhoods, locations with higher percent people of color had higher measured levels of metal concentrations, on average. Furthermore, the estimated difference in metal concentrations by race was statistically significant at all sample locations for lead. This suggests that race is a consistent



Fig. 3 (continued).

determining factor of inequalities in exposure to harmful air pollutants within Seattle's Georgetown and South Park neighborhoods, and that studies linking measured exposure with health outcomes are warranted.

Our study is subject to a number of limitations. First, it is important to note that our modeling method should be interpreted as an exploratory technique, and not a tool for making specific spatial inferences, especially in light of our small sample size (Páez et al., 2011). In previous studies, small sample size has been found to introduce spurious correlations between local coefficients (Devkota et al., 2014; Páez et al., 2011). In addition, we did not adjust for multiple comparisons, where the possibility of a statistically significant finding by chance increases with more than one test on the same hypothesis (Williams et al., 1999). However, our model comparison statistics indicated that OLS associations should not be assumed to be constant across the study area, and GWR models provided equivalent or better model fit than OLS values for all metals. While it would not be advised to derive specific policy measures based on our findings, our results can be used to guide further investigation.

Second, measurement of metals concentrations in moss samples does not suffice for air pollution data from more traditional monitoring techniques. A parallel monitoring and sampling campaign, which the Puget Sound Clean Air Agency is initiating at this time of this study, would be necessary to translate concentrations from moss to air pollution concentrations, and ultimately to assess human exposures and evaluate health effects. Third, modeling of spatial predictors does not suffice for advanced source modeling that includes accurate data on emissions of these metals. While some emissions are voluntarily reported from some industries in the area, these data give an incomplete picture, and future research could develop projections based on industry codes (such as Standard Industrial Classification codes) and staffing levels for businesses in the study area. Potential confounders that were not included in these analyses include information on local climate conditions, wind flow patterns, soil cover and drying. The potential contribution to airborne metal concentration of soil re-suspension would also be an important area of future investigation.

GWR results for nickel were sensitive to a single outlier. We identified this outlier using Cook's Distance based on OLS regression, indicating that the concentration at this point does not relate to spatial predictors the way it does at other points. Prior analyses suggest that this outlier is unlikely due to measurement error, but rather reflects high air concentrations of nickel (in addition to cobalt) (Jovan et al., under review). Air monitoring near the site and throughout the study area would be required to confirm this localized hotspot. GWR model results without the outlier provides a conservative estimate of spatial predictors and is an accepted method to produce robust results (Fotheringham et al., 2003).

Finally, our sample was collected using a community science approach, and sampling methods could have influenced internal validity of our data. These limitations are described elsewhere (Jovan et al., under review). However, samples taken by youth exhibited a high level of precision (Derrien et al., 2020), and replicate samples taken by expert scientists, while differing somewhat in absolute concentrations, captured similar spatial information (Jovan et al., under review). These measures of sampling precision and accuracy strengthen our confidence that these moss samples are of high quality and were suitable for our GWR analysis. Furthermore, we weigh any tradeoffs in data quality against the value of community leadership, engagement, and empowerment through our community science design.

5. Conclusions

This study provides further evidence of the utility of moss as a viable bioindicator, combined with spatial statistical techniques, in detecting spatial patterns and predictors of airborne metal deposition and semiquantitative concentrations. Our findings underscore the significance of activities in or near the Duwamish River, and the volume of motorvehicle traffic, as predictors of metal concentrations found in moss in the Duwamish Valley. Our results are consistent with the potential importance of urban trees as mitigation measures for metal pollution. In addition, it suggests that people of color are disproportionately exposed to heavy metals – indicating additional evidence of the environmental injustices in these neighborhoods. These results can aid in focusing further investigations into specific sources, relationships with air concentrations, and mitigation measures.

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CRediT authorship contribution statement

MK, LP, AD, and HM contributed to conception and design of the study; SJ, WB and CZ contributed to data acquisition; MK, LP, AD, HM, SJ and CZ contributed to analysis and interpretation of data; MK, LP, AD, SJ, MD and CZ contributed to drafting the article, all authors critically reviewed the article; and all authors gave final approval of the version to be submitted.

Declaration of competing interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.153801.

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