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Small-scale distributions of polycyclic aromatic hydrocarbons in urban areas using geospatial modeling: A case study using the moss *Orthotrichum lyellii* in Portland, Oregon, U.S.A.

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HIGHLIGHTS

- We mapped PAHs using geostatistical modeling on a spatially-balanced sample.
- Traffic emissions and canopy cover were important predictors of PAHs in moss.
- Weather and sampling nuance were also highly influential.
- Models for particle-phase PAHs were stronger than for gas-phase PAHs.
- Results inform urban planning and the design of future moss studies.

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ABSTRACT

Polycyclic aromatic hydrocarbons (PAHs) are widespread pollutants that are costly to measure with air quality instruments. We used a spatially balanced sample of an epiphytic moss, *Orthotrichum lyellii*, to develop fine-scale maps of atmospheric PAHs across residential areas of Portland, Oregon. The unusual abundance of this stress-tolerant species enabled us to systematically collect a large sample ($n = 350$) on a 1×1km grid across the city. This provided the unique opportunity to investigate potential effects of more than 20 factors on PAH concentrations in moss, including emissions sources, tree canopy cover, relative elevation, short-term weather, and nuance of sample collection. We measured 20 PAHs in moss samples and assigned them to 4 groups for analysis based on molecular weight (MW): the Naphthalene, Low, Medium, and High MW groups. Spatial regression models explained a modest amount of variation in PAH concentrations in moss, with spatial structure increasing as MW increased. Motor vehicle emissions were the only PAH source we detected and was highly significant in all models. Deciduous tree cover correlated with lower PAH concentrations in moss for all except the high MW group. Short-term weather and nuance of sample collection, like height on the sampled tree, tree taxonomic family, and whether sample was primarily collected from a branch or bole, had surprisingly large effects on all groups. Overall, results aligned with expectations based on each group's dispersal potential due to MW. All PAH groups were highest over downtown Portland, along major highways, and lowest in parks and outer neighborhoods, although the details varied. Our results supported two common themes in studies directly measuring PAHs: the importance of roads as an emissions source in residential areas, and the potential for trees to reduce atmospheric concentrations. Moreover, the large effects of weather and sampling nuance have critical implications for the design of future studies.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a common class of air pollutants linked to a range of adverse health outcomes, including asthma, cancers, cardiovascular disease, and fetal growth impairment (Boström et al., 2002; Choi et al., 2012). Compared to criteria pollutants, like fine particulate matter ($PM_{2.5}$) or nitrogen dioxide (NO_2), relatively little is known about levels of PAHs in air breathed by the general population (HEI Panel on the Health Effects of Traffic-Related Air Pollution, 2010). As PAHs are by-products of the combustion of organic matter, including fossil fuels and firewood, urban residential areas receive PAHs from many dispersed sources. Concentrations vary over short distances (<0.5 km–1km), creating heterogeneity infeasible to detect with monitoring networks of instruments due to poor spatial resolution (Strum and Scheffe, 2016).

The main network measuring PAHs in the U.S., the National Air Toxics Trends Analysis Stations (NATTS) Network, tracks concentrations using just 27 permanent monitors across the country (Strum and Scheffe, 2016). Due to high monitor costs, smaller-scale patterns are often simulated using dispersion and receptor-based modeling (e.g. Oregon Department of Environmental Quality, 2012). While convenient for visualizing potential air quality scenarios, dependence of these models on emissions inventories is a critical limitation. It's assumed all pollutant sources are known *a priori* and their emission levels accurately quantified, which is unrealistic for complex urban areas (Environmental Protection Agency, 2014). Given the rarity of PAH monitors, modeled scenarios are not validated by much empirical information, if at all. For that, inclusion of less expensive passive PAH monitors (i.e. PUF) can be helpful although equipment costs, vandalism, the need for multiple field visits, and restrictions on drilling into or affixing equipment to public or privately owned trees, quickly becomes limiting and cost prohibitive in urban environments.

Mapping pollutants with biomonitors provides a less deterministic perspective. Moss and lichens function similarly as PAH biomonitors; by lacking roots and an impermeable cuticle, they absorb water and nutrients from the atmosphere including co-occurring contaminants (Capozzi et al., 2020). Using inexpensive assays of pollutant concentrations in their tissue, it is feasible to collect large, spatially dense datasets to help identify new pollution sources and visualize patterns at unprecedentedly fine scales (e.g. Vuković et al., 2016). For instance, cadmium (Cd) extracted from the moss samples used in the present study helped regulators track down a significant source of Cd emissions in Portland, Oregon, neighborhoods that wasn't discoverable using dispersion modeling (Donovan et al., 2016). As moss and lichen interact with the atmosphere differently than humans, however, air instruments are needed to assess potential human health effects. Therefore, we use assays as a hi-resolution screening tool for more effective utilization of instruments and other monitoring resources.

As PAHs exhibit a range of physicochemical properties depending on ambient conditions and the number of benzene rings they contain, they are more challenging to study than heavy metals like Cd. Those with lower molecular weights (MW) have 2–3 rings and mainly exist in the atmosphere as gases, while higher MW PAHs (5–6 rings) occur as particles. Medium MW PAHs (4 rings) can occur in either phase depending on ambient temperature and vapor pressure (Baek et al., 1991). Gas-phase PAHs easily diffuse in and out of moss cells (Keyte et al., 2009), potentially "hopping" to other receptors (Huang et al., 2018). Particle-phase PAHs tend to accumulate externally on moss or lichen surfaces, becoming trapped or potentially washed off by rain (Aboal et al., 2011; Augusto et al., 2013a). Past research suggests moss primarily traps particles in the inhalable fraction (particulate matter diameter ≤ 10 μm; PM_{10}; Tretiach et al., 2011), which includes the fine particle fraction considered most hazardous to human health (i.e. PM with diameter ≤2.5 μm; $PM_{2.5}$; Brown et al., 2013).

Many studies use moss and lichen to characterize spatial patterns in PAH deposition (e.g. Ares et al., 2009; Augusto et al., 2013a; Harmens et al., 2013; Vuković et al., 2016). Likewise, comparisons to field measurements of atmospheric concentrations or deposition report moderate to strong correlations with bioaccumulated concentrations or PAH profiles for both the medium and high MW PAH groups (Aboal et al., 2020; Augusto et al., 2010; Augusto et al., 2013b; Blasco et al., 2006; Domínguez-Morueco et al., 2015; Foan et al., 2015; Loppi et al., 2015; Skert et al., 2010). In contrast, it has been demonstrated that mosses are not good biomonitors of LMW PAHs (Aboal et al., 2020; Capozzi et al., 2020; Domínguez-Morueco et al., 2015; Loppi et al., 2015). For positive associations, however, it remains unclear how generalizable relationships are across environmental conditions, study areas, and organism (moss vs lichen, although see Capozzi et al., 2020). While it is well-known that ambient conditions strongly influence PAH behavior in the atmosphere, our understanding of environmental and situational effects on PAH bioaccumulation is poor (Harmens et al., 2013). For epiphytes, it should be noted that atmospheric levels of PAHs affect bioaccumulation both directly and indirectly via stemflow and throughfall, making characteristics of the sampled tree also important to consider.

Mapping studies usually side-step these unknowns using strict standardization of sample sites and a narrow sampling window to reduce potential sources of variability. Urban studies are particularly challenging because pollutant deposition is often highly complex, requiring many observations to describe it (Gatziolis et al., 2016; Li et al., 2016; Strum and Scheffe, 2016), yet common stressors such as poor air quality and the heat island effect tend to limit biomonitor availability. Most studies use the practical strategy of transplanting material from outside the study area or else sampling in-situ material growing in forested enclaves or under other special sets of conditions (Ares et al., 2012; Orliński, 2002; Vuković et al., 2016). A novel perspective could be gained, however, by focusing on how the natural complexity of the study area affects PAHs in moss. If biomonitor availability weren't limiting, standardization could be relaxed in favor of collecting a spatially-balanced sample that was both intensive and extensive. Then, with geostatistical modeling, important sources of variability could be identified and controlled for statistically - rather than restrictive sampling and *a priori* assumptions about which sources are important.

We took advantage of an unusually abundant epiphytic moss, *Orthotrichum lyellii* (Hook. & Taylor) to produce fine-scaled maps of PAHs across residential areas in Portland, Oregon, and identify important predictors of concentrations in moss. We were able to systematically sample across the city on a 1 km × 1 km grid, with additional, smaller-scale sampling applied at random, to collect an unprecedented amount of data (n = 350). For the first time, we used geospatial statistics to identify and simultaneously model effects of over 20 potential drivers of PAHs in moss, providing new insight into the complexity of environmental influences on moss concentrations. In addition to helping visualize PAH levels across Portland in detail, the models raise considerations for the design and interpretation of future mapping studies.

2. Materials and methods

2.1. Study area and sampling strategy

Portland is a city in northwest Oregon, USA, with a population of 603,000 (U.S. Census Bureau, 2014). It has a temperate oceanic climate with annual mean temperature of 12.5 °C and annual precipitation of 91 cm falling mostly in the winter months. The acrocarpus moss *Orthotrichum lyellii* is the most abundant species across sites in Portland and several other West Coast cities, including locations judged *a priori* as polluted. To our knowledge, we are the first to use this species in a PAH-focused study. We sampled moss in December as past research shows PAH levels in moss, lichens, other vegetation, and the atmosphere, are highest in winter (Augusto et al., 2013b; Baek et al., 1991; Capozzi et al., 2020; Simonich and Hites, 1995). Local air monitoring data confirm that Portland's peak PAHs concentrations occur between December and mid-February (Oregon Department of Environmental Quality, 2012).

We used a sampling scheme with two components. To ensure a spatially balanced sample, we laid out a 1-km grid across the city. We focused on characterizing PAHs in residential areas, so we excluded industrial areas, the airport, and large parks (notably the 2000 ha of Forest Park). We then placed 226 points randomly on a street within each 1-km grid cell and identified the nearest address (Fig. 1). To help predict PAHs in moss at points along the edge of the city, we extended the grid an additional 1-km past the city boundary, where possible. This added another 52 sample locations.

To characterize the spatial correlation and estimate semivariograms for the dataset, we took additional samples at close distance from the grid points (Hasselbach et al., 2005; Zimmerman, 2006). Specifically, we took additional samples within 100 m of 72 randomly selected grid points, called "re-samples." Twelve re-samples were taken at the same point as the first moss sample, and the remaining 60 re-samples were taken between 10 m and 100 m from the first sample location, six at each 10 m increment. The total sample size was 350: 226 grid points, 52 additional grid points in the 1-km buffer, and 72 re-samples (Fig. 1).

Given the large number of samples needed, we collected moss for 3 weeks (December 2–23, 2013). Since weather influences PAH concentrations in both atmosphere and moss, systematic sampling from one side of the city to the other would introduce spatial bias. Therefore, we split the sample randomly into six spatially balanced subsamples. We then used the fastest route between points in a subsample, meaning we looped through the city six times over the three weeks of fieldwork.

We collected moss on hardwood trees from at least 6 distinct clumps above 1-m or higher than the ground to reduce spray from cars. We prepared at least 5.0 g of cleaned, dry moss for laboratory analysis, much smaller than the biomass of *O. lyellii* on most hardwoods (Fig. 2). When there was not a tree with the moss at the randomly selected address, we sampled the nearest suitable tree. Most trees occurred along streets or wayside areas in the public right-of-way. We also sampled trees in parks and on private property with owner permission. If a suitable tree was not available at the required distance for a re-sample, we randomly assigned the re-sample to a different grid point. We recorded sample locations using a high-accuracy GPS (Garmin Oregon 450).

2.2. Moss sample preparation and analysis

Immediately after collection, we stored moss samples at 4 °C in metalized polyester Kapak bags. Each moss stem grades from young, bright green tissue to older, dark brown tissue and rhizoids at the moss base where varying amounts of large particulates and dirt collect. On a lab bench we used scissors and forceps to trim away all but approximately the upper 2/3 of moss stems (Fig. 2). We washed all equipment

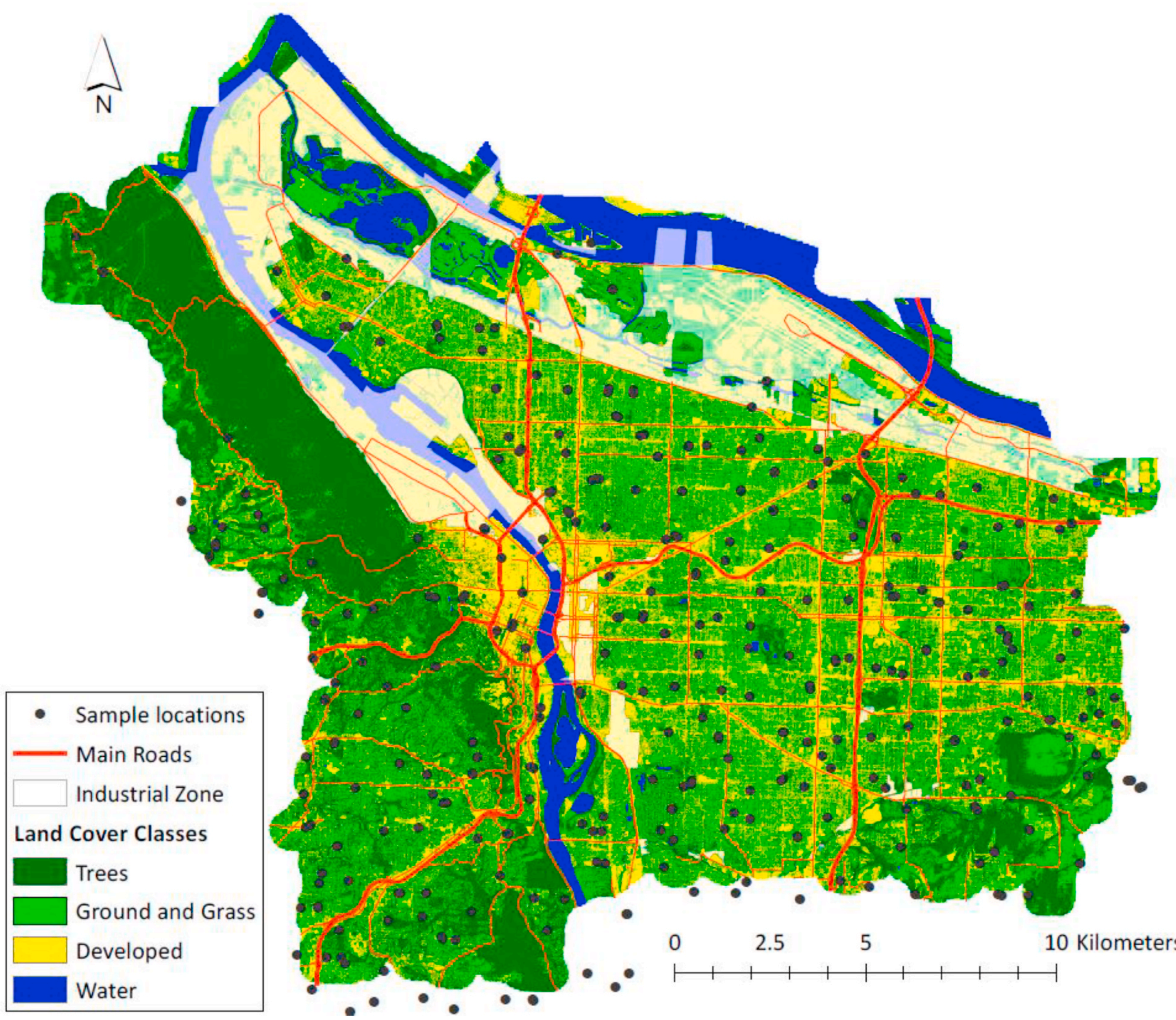

Fig. 1. Map of the study area in Portland, Oregon, showing sampled locations, roads, and land cover classes.

Fig. 2. Photo panel showing the typical environment of a street tree collection site (left, middle) and a close-up of trimming moss fronds for analysis of PAH concentrations (right).

with 70% ethanol between samples. On average, samples were stored for 1 week or less before lab analysis to prevent decay and loss of volatile PAHs.

We used at least 3.5g of moss, dried weight, to extract the EPA's 16 priority PAHs and four others, for a total of 20 (Table 1). Moss samples were desiccated and finely ground using 1–3g of diatomaceous earth. Samples were loaded into 33 mL stainless steel cells, with 1g Florisil® (activated magnesium silicate) from U.S. Silica Company (Berkeley Springs, USA) placed at the outlet of the cell.

An ASE 200 (Dionex, Sunnyvale, CA) was used for pressurized liquid extraction of PAHs (EPA method 3545a) with dichloromethane as the solvent. We pre-heated cells to 100 °C for 5 min to thermal equilibrium then processed with two static extraction cycles. Between cycles, we flushed cells with 50% of cell volume solvent. At the end of cycles, the cell was flushed with high purity nitrogen (N). We concentrated the PAH extract under N at ambient temperature to a final volume of 1 mL.

We analyzed the extract using a method similar to EPA 8270D. Internal Standard recoveries of 50%–150% were considered acceptable for quantifying target PAH compounds. Analytical batch quality assurance and quality control parameters included method blanks, laboratory control standards, initial and continuing calibration verification standards, and matrix spiked samples. Use of the isotopically-labeled PAH analogues (Cambridge Isotopes) provided information on individual compound extraction efficiencies for each sample.

Chromatographic separations were optimized on a Restek Rxi-XLB, 30 m × 0.32 mm x 1.00μm. An Agilent 6890 GC with 5973 Mass Selective Detector operated in full scan mode was utilized for analysis. Calibration was performed using 1 μL injection volumes. Six calibration levels ranging from 0.01 to 20.0 μg/mL were utilized to quantify PAH compounds. Average response factor calibration curves were selected for quantification, with a maximum percent relative standard deviation of 20%.

A primary target ion and two secondary quantification ions were utilized for all target PAH compounds. Identification of compounds was based on retention times determined during initial calibration, and relative response of the primary and secondary quantification ions. The mass selective detector was tuned using decafluorotriphenylphosphine according to EPA 8270 criteria and verified prior to analysis and every 12 h during use.

For quality assurance, we ran a method blank, matrix spike with

Table 1
Summary of PAHs measured in moss. Groups are based on molecular weight (MW). MDL = minimum detection limit.

PAH group	# of rings	MW g/mol	# below detection	Mean mg/kg	SD	Median mg/kg	Max mg/kg	Min mg/kg	MDL μg/kg
Naphtha				45.51	26.61	41.17	320.90	2.78	
naphthalene	2	128.17	2	20.32	11.21	18.20	96.00	1.24	0.24
1-methylnaphthalene	2	142.20	59	6.59	5.03	5.71	32.50	0.10	0.28
2-methylnaphthalene	2	142.20	19	10.82	7.29	9.70	57.30	0.10	0.33
acenaphthylene	3	152.20	92	7.78	10.86	6.64	175.00	0.42	0.42
Low MW				115.57	104.76	91.95	1309.60	11.16	
acenaphthene	3	154.20	113	5.83	7.57	3.47	52.80	0.22	0.38
fluorene	3	166.22	100	7.13	8.39	4.72	62.00	0.42	0.43
dibenzofuran	3	168.19	51	14.51	27.65	7.80	220.00	0.87	0.43
phenanthrene	3	178.23	0	72.64	70.73	59.10	876.00	6.16	0.31
anthracene	3	178.23	12	15.47	23.95	11.15	283.00	0.87	0.34
Medium MW				437.96	510.19	353.15	7540.00	34.69	
pyrene	4	202.25	0	138.76	158.96	112.50	2340.00	11.00	0.19
fluoranthene	4	202.26	0	151.15	196.79	119.00	2900.00	11.80	0.23
benzo(a)anthracene	4	228.29	3	50.71	65.15	39.75	1030.00	1.21	0.15
chrysene	4	228.29	1	97.34	119.46	75.00	1270.00	2.25	0.28
High MW				260.20	300.36	202.24	3950.00	20.63	
benzo(a)pyrene	5	252.31	22	35.90	58.17	27.45	838.00	0.79	0.22
benzo(b)fluoranthene	5	252.31	0	54.47	55.91	44.20	624.00	4.91	0.20
perylene	5	252.32	2	36.94	44.40	28.75	580.00	0.87	0.20
benzo(k)fluoranthene	5	252.32	1	46.47	55.90	36.80	838.00	0.64	0.18
benzo(ghi)perylene	6	276.34	4	64.31	83.59	48.35	1290.00	1.21	0.28

duplicate, and laboratory control sample for each batch of 20 samples. Samples used for matrix spikes were randomly selected from samples with adequate mass for both sample and matrix spiked analysis. Initial and continuing calibration verifications used a three-point calibration not including the calibration blank. Quality control data are presented in the Supplementary Materials section.

2.3. PAH groups

The spatial scale of PAH deposition and chemical toxicity are highly influenced by MW (e.g. Augusto et al., 2009; Geier et al., 2018). For modeling we classified PAHs into four groups based on MW and number of carbon rings (Table 1): the 'naphtha' group (mostly 2 rings), the low MW group (3 rings), the medium MW group (4 rings), and the high MW group (5–6 rings). One PAH (dibenzo(a,h)anthracene) was beneath detection limits for most samples and so was eliminated from further analysis.

2.4. Statistical modeling

We fit spatial linear models to the natural logarithm of PAH concentrations in moss to each of the four MW groups. We log-transformed concentrations because they were highly skewed and strictly positive. We considered three auto-covariance structures (exponential, spherical, or Gaussian) plus a model without spatial covariance, and chose the best fitting model for each PAH group using Akaike's information criterion. We estimated the spatial parameters (nugget, partial sill, and range) using restricted maximum likelihood (REML). To help ensure a global minimum, we used a grid of values centered on the estimated values of the three spatial parameters from empirical semivariograms as starting values in our spatial regression. We used a stepwise variable selection process with a p-value cut-off of 0.1 to select a parsimonious model from the large number of covariates we considered. Note that this approach is the similar to traditional geostatistics and kriging, except that fixed model covariates and the spatial correlation are fitted jointly using maximum likelihood, and the mapped products are based on spatial predictions from the model (Ver Hoef et al., 2001). Our large number of systematically collected moss samples supported use of this different approach seen also in Donovan et al. (2016) and Hasselbach et al. (2005). We estimated the spatial regression model using SAS's MIXED procedure (SAS Institute Inc, 2011).

Table 2
Summary of covariates used to model PAHs in moss.

	Mean (sd)	Min, Max
Sources		
Annual average daily traffic (# cars/day, inverse-distance weighted after square root transformation, 500 m buffer)	182.50 (106.40)	0.00, 562.52
Railroads (%, inverse-distance weighted cover, 500 m buffer)	0.70 (2.09)	0.00, 12.80
Distance to the Portland airport (km)	9.80 (4.40)	0.77, 19.91
PM2.5 emitted from residential wood burning (kg/ha)	26.90 (18.30)	0.53, 116.94
Industrial land cover use (%, 500 m buffer)	2.91 (9.73)	0.00, 83.93
Naphtha-group point source emissions (kg/yr, inverse-distance weighted, 500 m buffer)	39.10 (60.20)	0.00, 350.18
Low MW point source emissions (kg/yr, inverse-distance weighted, 500 m buffer)	0.74 (2.21)	0.00, 13.16
Medium MW point source emissions (kg/yr, inverse-distance weighted, 500 m buffer)	0.35 (0.83)	0.00, 4.60
High MW point source emissions (kg/yr, inverse-distance weighted, 500 m buffer)	0.08 (0.27)	0.00, 1.76
Protectors		
Deciduous tree-canopy cover (%, inverse-distance weighted 500m buffer)	20.70 (9.20)	3.60, 52.20
Conifer tree-canopy cover (%, inverse-distance weighted 500m buffer)	10.80 (7.70)	0.20, 37.30
Relative elevation of sample point (m)	4.10 (29.60)	−62.80, 259.40
Weather		
Mean temperature of the sample day (°C)	1.79 (4.00)	−6.67, 6.11
Mean temperature of the week preceding the sample day (°C)	1.73 (3.31)	−3.57, 6.03
Mean relative humidity of the sample day (%)	82.70 (12.00)	61.00, 97.00
Mean relative humidity of the week preceding the sample day (%)	79.10 (11.30)	61.60, 93.10
Rainfall of the week preceding the sample day (mm)	1.17 (1.33)	0.04, 3.77
Sample characteristics		
Diameter at breast height of the sample tree (cm)	31.0 (14.90)	3.00, 111.80
Sample taken from a branch (% of sampled points)	13.3	
Sample fully or partially facing road (% of sampled points)	84.1	
Sample height on tree (m)	2.19 (0.63)	1.00, 4.00
Sample tree taxonomic family (% sampled points)	*Aceraceae* 28.0 *Betulaceae* 9.2 *Rosaceae* 28.0 Other 34.8	

2.5. Model covariates

Moss concentrations of PAHs are influenced by many factors (Harmens et al., 2013) and the complexity of urban environments complicates the standardization of sampling conditions, especially when collecting from hundreds of locations. Therefore, we considered a large number of covariates in model development (Table 2). Most area-based covariates were calculated within a 500-m buffer around each sampling point, as is common (Ares et al., 2009; Blasco et al., 2011; Environmental Protection Agency, 2014; Li et al., 2016).

2.5.1. Emissions sources

The main anthropogenic sources of PAHs in urban areas include traffic emissions, industrial point sources, residential heating, and trash incinerators for power generation (Baek et al., 1991). The first three are potential sources in Portland's residential areas (Oregon Department of Environmental Quality, 2012). To estimate vehicular traffic effects, we used an estimate of traffic volume (# cars/day), the average annual daily traffic (AADT), calculated for Portland's roadways in 2017 (Oregon Department of Transportation, 2018). *A priori*, we expect traffic near the center of the buffer has a greater impact than that near the edge, so we used inverse-distance weighting for traffic volumes. The AADT was highly skewed, spanning several orders of magnitude, so we used a square root transformation after inverse-distance weighting to meet the assumption of linearity. As other types of transportation may be significant sources of PAHs, we calculated the length of train tracks around each point (also inverse-distance weighted) and Euclidean distance from each sample point to the Portland International Airport.

Emissions dispersing from industrial point-sources could also be significant sources. We used self-reported emissions data from 75 facilities with permits to emit PAHs in Portland (Oregon Department of Environmental Quality, 2012, 2016). For each of the four PAH groups, we calculated their inverse-distance weighted load within 5 km buffers around each sample location. We explored alternative ways of calculating load, including smaller buffers down to 1 km, but analysis outcomes were the same.

As a proxy for PAH emissions from domestic heating, we included variables describing $PM_{2.5}$ emissions from residential wood burning as a primary or a secondary heat source. Data were derived from models based on a survey of 1601 households in the Portland metropolitan area administered by Oregon Department of Environmental Quality (Swab et al., 2019). Standard EPA emissions factors were used to estimate $PM_{2.5}$ emissions at the census block-group level.

2.5.2. Protectors

The PAH-scavenging ability of urban trees is well documented (e.g. Huang et al., 2018; Simonich and Hites, 1995) so we estimated percent canopy cover of deciduous and conifer trees around each sampling point to attempt to capture this effect. Percent cover was inverse distance–weighted within the usual 500-m buffers. Cover estimates were based on Meter-Scale Urban Land Cover (MULC) data for the study area (Environmental Protection Agency, 2012), obtained by processing 1-m National Agriculture Imagery Program imagery (Fig. 1).

The elevation of a sampling point relative to the surrounding terrain could also influence PAH levels by impacting airflow and deposition. Urban dispersion models represent these complex interactions in a variety of ways (HEI Panel on the Health Effects of Traffic-Related Air Pollution, 2010). As a coarse indicator, we used a 0.91-m LiDAR-derived digital terrain model (Oregon Department of Geology and Mineral Industries, 2014) to calculate the relative elevational difference of each sample point to the mean ground elevation of the surrounding 500-m buffer. Positive values meant the sample point was higher than its surrounding area.

2.5.3. Weather

Our sampling strategy aimed to prevent weather-induced bias in PAHs mapping (See subsection 2.1: ***Study area and sampling strategy***). Even so, we accounted for possible short-term weather effects in the models. We used the average temperature, rainfall, and relative humidity during the day and week before each sample was collected as covariates (Weather Underground, 2013).

2.5.4. Sample characteristics

We harvested moss from numerous patches and locations on each sampled tree to avoid microsite variation although moss sometimes clustered in one area. We noted samples taken primarily from the trunk or branches, from the roadside face of the trunk, and samples collected within a narrow height range. We also noted genus of sampled trees and diameter at breast height. We grouped trees into the three most commonly encountered families (*Aceraceae*, *Betulaceae*, *Rosaceae*) and a fourth group representing all other sampled taxa, as an indicator of tree-level effects on PAH accumulation in moss.

2.5.5. Prediction maps

For each PAH group, we created two city-wide maps of concentration in moss, the first based on kriging observed concentrations, and the second on the estimated geostatistical spatial model. Using both approaches, we predicted the log PAH concentration on a 50m grid across the city ($n = 173{,}612$) and calculated values for all significant source and 'protector' variables for points on the grid. For significant variables describing weather and sample characteristics, we assigned the median observed value across the entire sampling period for continuous variables, and the most frequent group for categorical variables.

3. Results

We were able to collect moss at 346 of the 350 sample points. The four missing points were in heavily wooded areas with low road density and high canopy closure. In these conditions, *O. lyellii* only grew high in the tree canopy near branch tips. Most sampled trees were street trees (60%), while the rest occurred in the yard of a residence (18%), wayside areas (14%), near parking lots (9%), or parks (2%). Median distance from the randomly selected address to actual sample location was 92 m. The distance of sampled trees to the nearest roads and residences ranged from 0.1 m to 101 m and 0.5 m–176 m, respectively.

3.1. PAHs summary

The number of non-detects varied widely by individual PAHs and groups, with notably few for the 4-ring (medium) PAHs (Table 1). Acenaphthene and fluorene had the largest number of non-detectable concentrations. The naphtha group had the lowest observed concentrations while mean, median, and range of concentrations for the other groups ranked from low < high < medium MW group. The range of values overall was large, with several PAHs having a minimum of less than 1 mg/kg whereas pyrene and fluoranthene had maximum values greater than 2000 mg/kg. Quality control data and associated discussion are presented in the Supplementary Materials (Tables 1–4).

3.2. Modeling results

Strength of the spatial structure within PAH groups increased as MW increased (Fig. 3) although except for the high MW group, spatial correlation was weak and practically eliminated once significant covariates were included in the model. Model predictive power also increased as MW increased (Fig. 3). Model performance is indicated by the relative decrease in residuals estimated in the semivariogram versus that of the null model with the raw PAH data. The reduction of variance when covariates were included in the model was negligible for the naphtha group (the ratio between the sill of the model for residuals to that of the raw data was 0.93), but more substantial for the low (0.88), medium (0.69), and high (0.79) groups.

Many covariates were inverse distance weighted, which can be difficult to interpret, so we standardized all continuous variables by subtracting the mean and dividing by the standard deviation. The magnitude of regression coefficients can thus be compared as they all denote the effect of a 1-SD increase in the covariate on the natural log of moss PAH concentrations. In other words, the standardized coefficients indicate the strengths of association between a covariate and PAH concentrations in the same scale such that a doubling in value represents a doubling in effect size. Categorical variables were not transformed.

Estimated road emissions (AADT) were the only emissions source represented in the final models (Table 3). It was the strongest predictor of PAH concentrations for all but the medium MW group, but nevertheless had a large regression coefficient for that group. Strength of group associations with AADT, as represented by the standardized coefficients, increased.

monotonically with MW. Coefficients nearly doubled from 0.11 (Naphtha) to 0.21 (High MW), suggesting effect on the heavy group was twice as strong.

Conifer canopy cover, which was less abundant and irregularly distributed in the study area, was non-significant. However, deciduous cover correlated negatively with all but the high MW group. Among the three other groups, strength of the association again increased with molecular weight. Relative elevation (i.e. sampling point higher than the surrounding area) had a lesser but significant negative association with the two heaviest MW groups.

Relatively short term (daily or weekly) differences in weather were highly significant for all groups, although which predictor was important, and the sign of the association, varied by group. The weather effect was greatest for the medium MW group (0.20). That was the one case where a covariate besides traffic was the strongest predictor of concentrations in moss.

One or more sample characteristics were important for all but the low MW group. Like results for weather, important variables for each group varied. The only shared result was higher concentrations of the naptha and medium MW groups measured in moss on trees in the *Rosaceae* family. Other conditions promoting higher concentrations included collecting moss primarily from branches instead of boles (medium MW), from trees in the genus *Aceraceae* (medium MW), and from lower heights on the sampled tree (high MW).

3.2.1. Prediction maps

To standardize effects of sampling idiosyncrasies in the PAH concentration maps, we standardized significant weather and sample characteristics to their median level: mean temperature of the sample day at 2.75 °C, rain for the week prior at 0.66 mm, mean humidity of the sampling day at 88%, mean humidity for the prior week at 77.6% and height on tree at 2 m. We also set taxonomic family to *Rosaceae* and

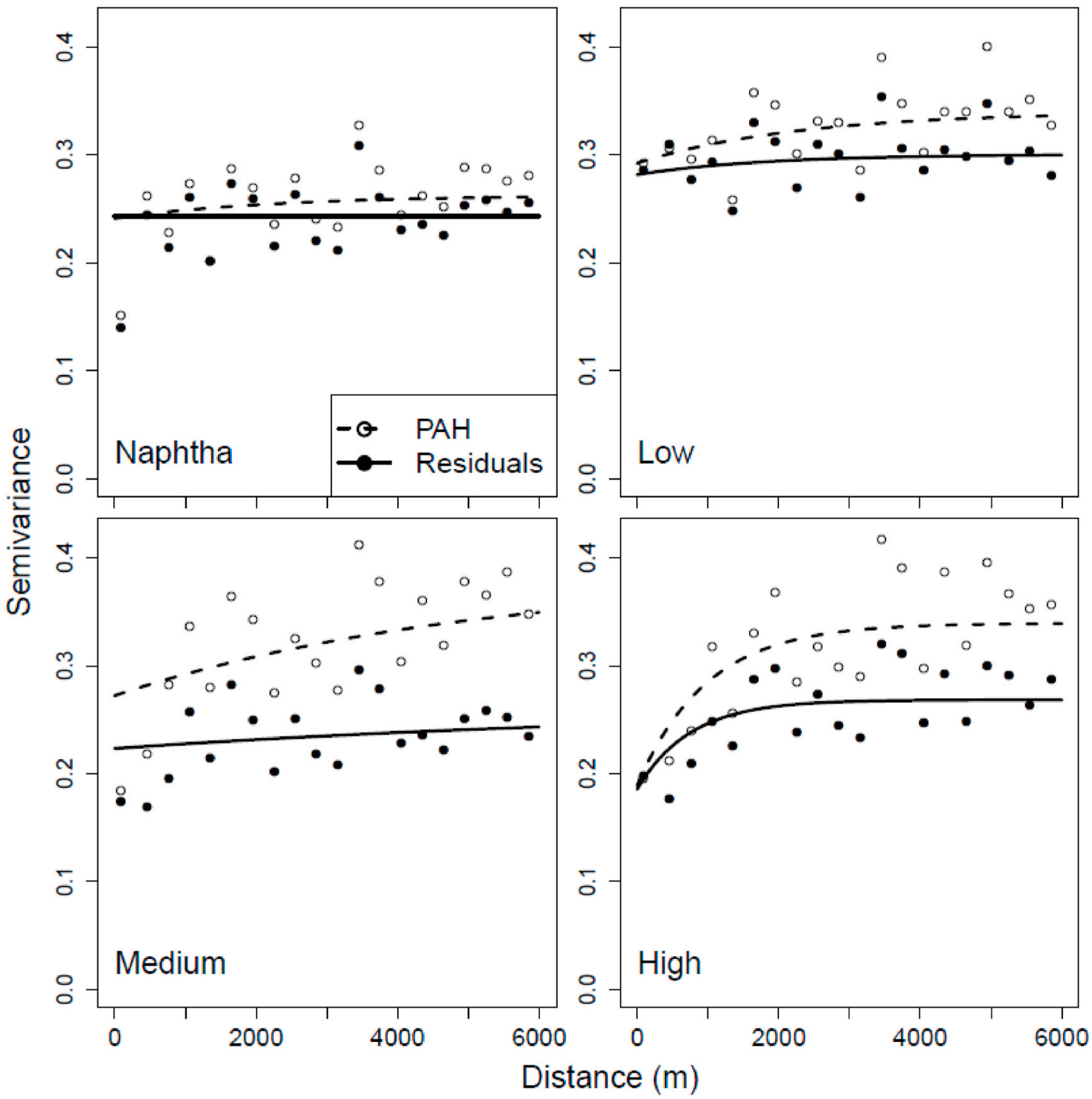

Fig. 3. Semivariograms for the three regression models. Each panel shows an empirical semivariogram of the raw PAH values (empty circles) and residuals after fitting the model (solid circles), together with the spatial auto-covariance model estimated using REML.

sample location to 'collected primarily from a bole.'

Because the absolute magnitude and range of PAH concentrations in the samples differed among PAH groups, we used different scales for mapping concentrations of the naptha and low MW groups versus the medium and high MW groups (Figs. 4 and 5). Predictions for each group were distinctive but shared common features. In the simple kriged maps without covariates, associations with major highways, thoroughfares, and intersections is apparent for all PAH groups, while the modeled maps more sharply articulate roadside gradients. The Interstate 405 loop around downtown Portland, which intersects all major highways, is in all cases a prominent area of relatively high PAHs. Generally, lower concentrations were detected in outer Portland neighborhoods and near large parks. Log-transforming the PAH values helped reveal more heterogeneity within the lower part of concentration ranges, including many additional, small-scale peaks near highway interchanges and smaller road intersections, as well as distinctly low PAH levels in areas with high tree cover.

4. Discussion

Despite the prevalence and well-known toxicology of PAHs, our capacity to monitor with existing technology is limited. The moss maps provide, for the first time, a cohesive depiction of small-scale heterogeneity based on empirical measurements rather than dispersal simulations. The underlying geospatial models allowed us to examine potential effects of a large pool of covariates at the same time, improving salience of the maps and integrating fragmented knowledge about the complex influence of numerous environmental and situational factors on moss-accumulated PAHs. A key advantage of simultaneously modeling covariates is the context gained; i.e. showing which relationships stand after taking all others into account. For instance, model selection of both estimated road emissions and deciduous canopy cover suggests they affect PAHs independently. If modeled singly, it would be unclear whether canopy effects were simply due to a lack of roads or vice versa.

Our models explained only a modest proportion of the observed variability in PAH levels, however, which may be due to influential factors we did not consider, as well as limitations of the datasets we employed (discussed further in subsection 4.5***: Study Limitations***). With only one PAH instrument in the study area, we couldn't validate modeled values, and a comprehensive validation study would be financially infeasible (e.g. the overall cost of this study was roughly equivalent to the annual operating costs of 3 active PAHs monitors). Our confidence in the value of the maps as screening tools thus rests on consistencies with prior PAHs studies as well as whether important

Table 3
Results of spatial regression models for the 4 groups of PAHs. Only associations with $p \leq 0.1$ are shown.

	Naptha Group			Low MW			Medium MW			High MW		
Variable	Std coefficient	Std error	p-value	Std coefficient	Std error	p-value	Std coefficient	Std error	p-value	Std coefficient	Std error	p-value
Intercept	3.6619	0.0313	$<10^{-4}$	4.5500	0.0401	$<10^{-4}$	5.6870	0.1188	$<10^{-4}$	5.3514	0.0387	$<10^{-4}$
Sources												
Annual average daily traffic	0.1057	0.0283	0.0002	0.1296	0.0333	$<10^{-4}$	0.1529	0.0301	$<10^{-4}$	0.2060	0.0313	$<10^{-4}$
Protectors												
Deciduous tree-canopy cover	−0.0478	0.0287	0.0961	−0.0945	0.0353	0.0078	−0.1310	0.0361	3×10^{-4}			
Relative elevation of sample point							−0.0532	0.0270	0.0498	−0.0602	0.0281	0.0329
Weather												
Mean temp. of sample day	−0.0510	0.0270	0.0594									
Rainfall of prior week preceding sample day				−0.0892	0.0298	3×10^{-3}						
Mean relative humidity of sample day							0.2031	0.0266	$<10^{-4}$			
Mean relative humidity of week preceding the sample dayrowhead										0.1183	0.0290	<10-4
Sample Characteristics												
Taken from a branch							0.1519	0.0821	0.0650			
Height on tree										−0.0721	0.0284	0.0115
Tree taxonomic family: *Rosaceae*	0.1067	0.0592	0.0724				0.1481	0.0638	0.0208			
Tree taxonomic family: *Aceraceae*							0.1293	0.0635	0.0424			

drivers in the models, their signs, and relative strengths of association, match *a priori* assumptions about the expected behavior of MW groups in the atmosphere.

The lower explanatory power of models for the naptha and low MW groups, and their relative homogeneity in modeled maps (Figs. 4 and 5), is consistent with the greater dispersal and re-volatilization ability of gas-phase PAHs (Baek et al., 1991). Although many studies detect concentrations of PAHs from the naptha and low MW groups in moss or lichens, recent work suggests moss does not retain lighter PAHs very effectively (Capozzi et al., 2020). Gas-phase PAHs easily cross moss cell membranes (Keyte et al., 2009), making accumulation less efficient than for the higher MW compounds (Harmens et al., 2013; Huang et al., 2018). Some degree of gas exchange during sample preparation in the lab is also possible (Fig. 2; Sucharová and Holá, 2014). To our knowledge, however, relationships to atmospheric gas concentrations have not been observed (Aboal et al., 2020; Domínguez-Morueco et al., 2015; Loppi et al., 2015), making their utility for biomonitoring these groups questionable.

Relationships for the less volatile PAHs are better documented. For instance, Aboal et al. (2020) recently found high correlations between 4, 5, and 6 ring PAHs in standardized transplants of cloned Sphagnum ("moss spheres") versus PM_{10} and bulk deposition measurements (correlation coefficients mostly >0.5 and 0.8, respectively). In a second example, correlations of in-situ lichen to an active particle sampler over 9 months in Portugal ranged from 0.66 to 0.81, with the highest correlation found using a 45-day averaging window (Augusto et al., 2013b; see also Domínguez-Morueco et al., 2015). We'd expect similar outcomes for moss although naturally lower levels of lipids, which adsorb PAHs onto biomonitor surfaces, could cause shorter retention times in moss tissues (see Knulst et al., 1995; Orliński, 2002). On the other hand, the higher surface area of moss may increase particle entrapment, and are reported to contain higher MW PAHs than lichen (Capozzi et al., 2020). Stronger geospatial models and more articulated distribution patterns for the medium and high MW groups in *O. lyellii* is consistent with more limited dispersal ability and thus, deposition closer to emissions sources (Figs. 4 and 5).

4.1. Emissions sources

We found roadside emissions were the dominant PAH source in Portland's residential zones, as is typical in non-industrial urban areas (HEI Panel on the Health Effects of Traffic-Related Air Pollution, 2010, Hwang and Wade, 2008; Kim et al., 2019). While the PAHs dispersal model available for the greater Portland area considers wood burning the biggest source, road emissions were also a major source category (Oregon Department of Environmental Quality, 2012). Steepness of PAH gradients in moss increased with MW, as seen in biomonitor studies focusing on roadside environments (Blasco et al., 2011; Nascimbene et al., 2014; Viskari et al., 1997; Vuković et al., 2016). While we did not explore diagnostic ratios for identifying emissions sources due to questions about dependability (Galarneau, 2008; Tobiszewski and Namieśnik, 2012), three PAHs (Phenanthrene, Pyrene, and Fluoranthene) often considered indicators of traffic emissions (e.g. Alfani et al., 2005; Foan et al., 2010, 2015; Orliński, 2002; Viskari, 2000) were highly abundant in our samples (Table 1).

We were initially surprised no PAH groups correlated with wood burning because residential heating emissions are a major driver of wintertime PAHs levels in urban areas (e.g. Bari et al., 2011; Vuković et al., 2015), including Portland (i.e. Oregon Department of Environmental Quality, 2012; Swab et al., 2019). Simulations from the Portland Air Toxics Solutions dispersal model suggest residential wood-burning explains about 80% of the variability in total PAHs across the greater Portland area. Wintertime burn bans are common in Portland during temperature inversions, which trap fine particulates near ground level, causing acute, unhealthy concentrations (Environmental Protection Agency, 2013; Savolahti et al., 2019).

We attribute the lack of relationship to spatial scale. Low resolution of the emissions data (aggregated to the census block level) and the focus

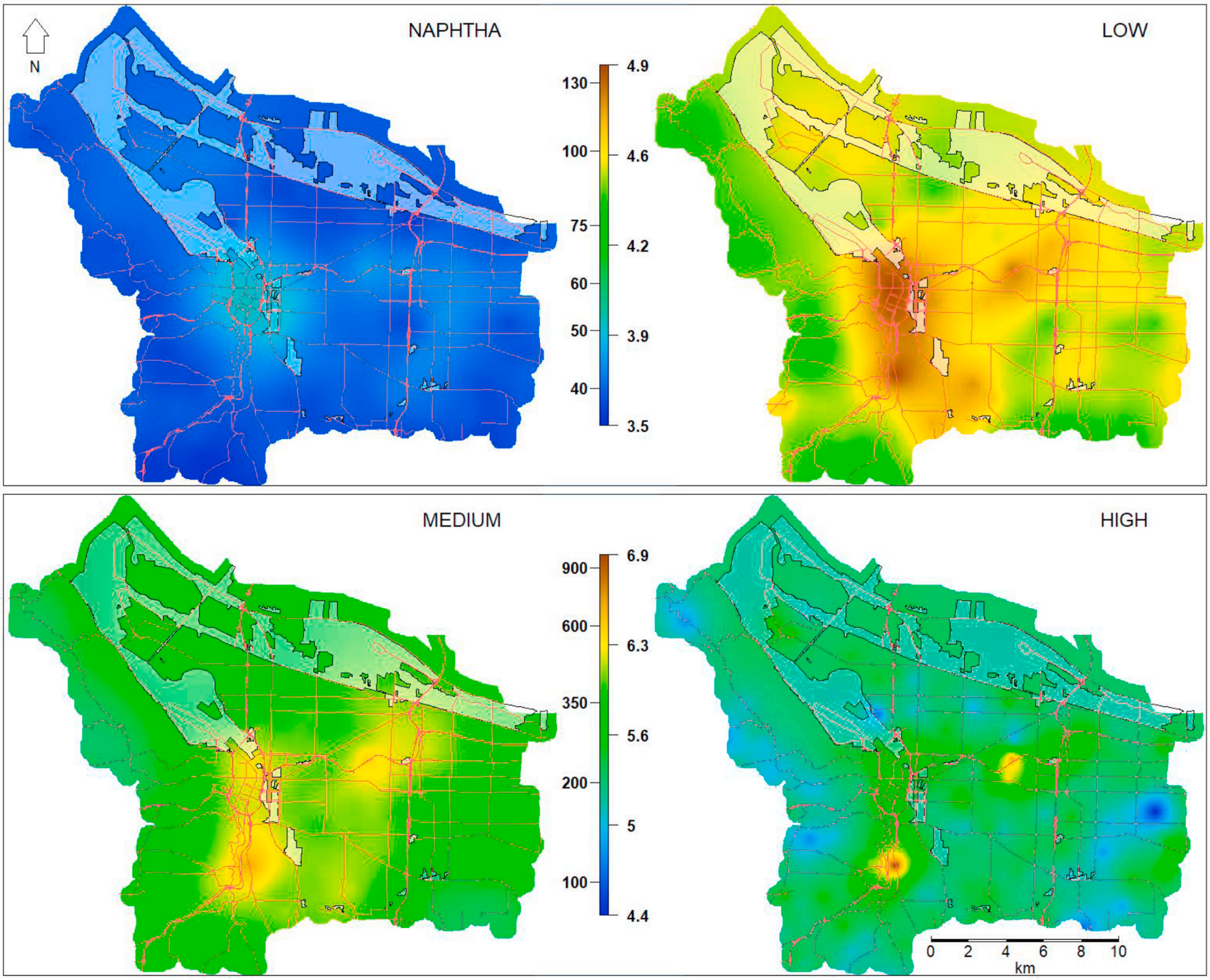

Fig. 4. Simple kriged maps predicting relative amounts of PAHs in moss by MW group. Note that scales used for the lower MW (top) and higher MW groups (bottom) differ to help better visualize variability across the study area. Both untransformed and log-transformed values are shown on the left and right sides of the scale bars, respectively. Industrial areas, which were not sampled for moss, are indicated by black boundaries with semi-opaque fill.

only on $PM_{2.5}$ likely impeded our ability to identify wood-burning as a source. Local-scale emissions from wood combustion can be heterogeneous both spatially and temporally and are thus difficult to describe with confidence (Hellén et al., 2017). Moss collects $PM_{2.5}$ as well as coarser particulates up to PM_{10} (Tretiach et al., 2011), and steeper, more localized gradients of the latter could easily dominate at the fine scale of our sample. While we suspect wood burning emissions are part of the unexplained variability in our models, we have no way to test that presumption at present.

4.2. Protectors

To our knowledge, this is the first biomonitor mapping study suggesting tree canopy has a reducing effect on atmospheric PAHs at the city-scale. Tree canopies are a major part of PAHs cycling (Howsam et al., 2001), and vegetation generally sequesters large amounts of atmospheric PAHs (Simonich and Hites, 1995; Tian et al., 2008), most of which end up in the soil, or, in paved urban areas, as runoff (Nowak et al., 2006; Peng et al., 2012). Canopy sequestration is usually thought of as a function of leaf area (Nowak et al., 2006), with particulate PAHs becoming trapped on the waxy leaf cuticles and gases taken up through stomata (Desalme et al., 2013; Wang et al., 2008). Therefore, it is notable that three of four PAHs groups correlated negatively with deciduous canopy cover even though we sampled moss roughly 1 month after leaf-off. The exception of the heavy MW group fits our expectation of mostly short-range, horizontal (vs vertical) dispersal from adjacent roads (Viippola et al., 2016).

That heavy MW PAHs were higher in moss collected low on the tree also supports this conclusion (see subsection 4.4: ***Sample Characteristics***).

Since moss integrates PAHs over some time period, a legacy effect from before leaf-off is entirely possible. That would mean two things, 1) we underestimate the reducing effect of deciduous canopy, and 2) integration times for *O. lyellii* are longer than the 30 days assumed for mosses based on their low lipid content (Orliński, 2002), or even the 45 day integration period suggested for particulate-phase PAHs in the lichen *Parmotrema hypoleucinum* (Augusto et al., 2013b). It is also likely that some degree of PAH scavenging by deciduous trees persists through the winter (Freer-Smith et al., 2005); while less understood, tree bark surfaces also scavenge PAHs (Rauert et al., 2017; Zhao et al., 2008).

The other 'protective' variable, relative elevation, suggested lower deposition of particle-phase PAHs at sites with high elevation relative to their local surroundings. Presumably there is less local turbulence, and therefore less particle deposition, at higher relative elevations (e.g. hilltops) compared to more sheltered areas where air dispersion is constrained (Kim et al., 2015). Winter inversions in the study area might

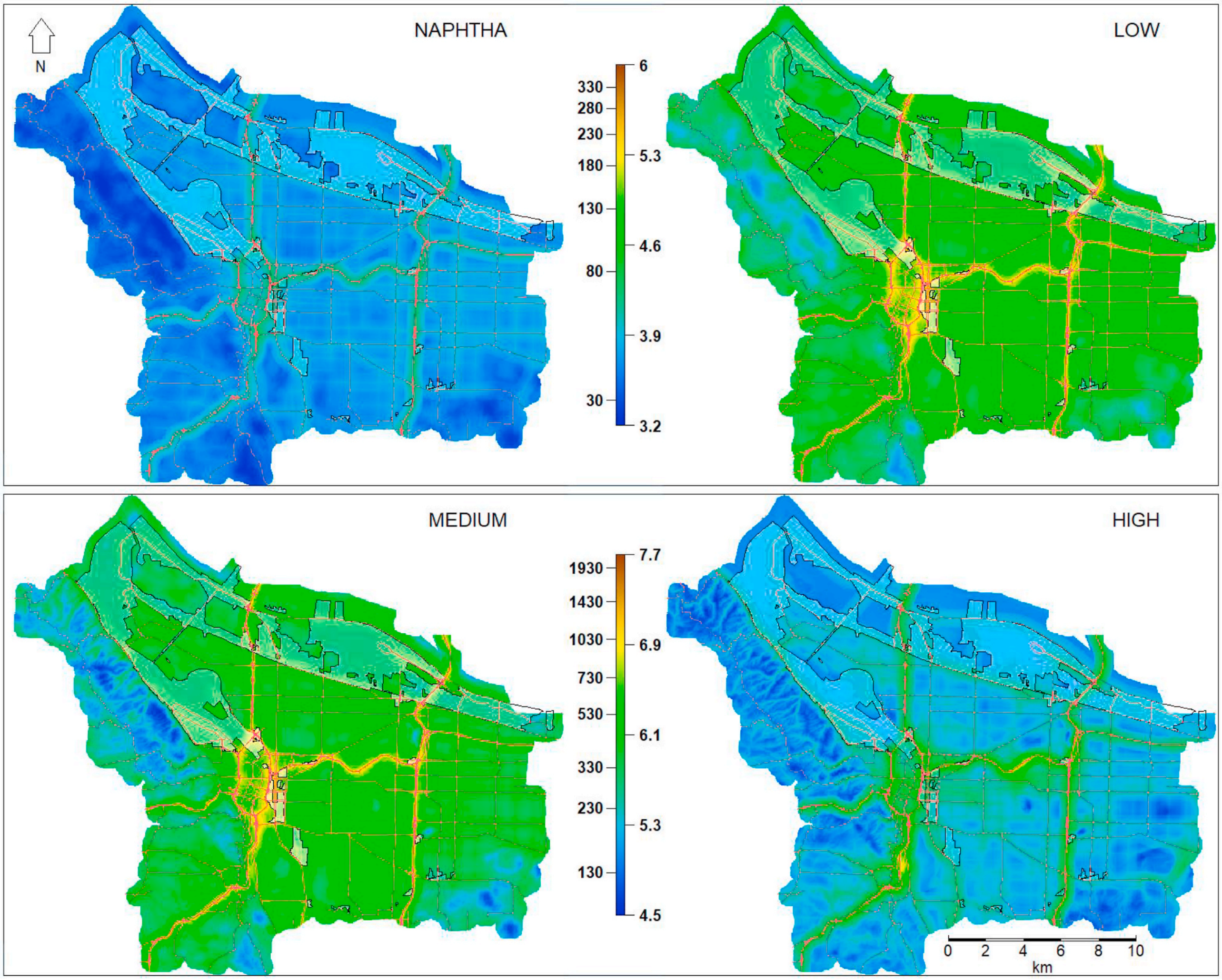

Fig. 5. Modeled maps predicting relative amounts of PAHs in moss by MW group using significant covariates. Note that scales used for the lower MW (top) and higher MW groups (bottom) differ to help better visualize variability across the study area. Both untransformed and log-transformed values are shown on the left and right sides of the scale bars, respectively. Industrial areas, which were not sampled for moss, are indicated by black boundaries with semi-opaque fill.

also increase particulates at lower elevations. Given their high dispersal ability, no detectable effect on the gas-phase PAHs is unsurprising.

4.3. Weather

Despite collecting all 346 samples over a narrow, 3-week range of weather conditions, weekly and even daily conditions were important predictors of PAHs concentrations in moss. Given our meandering data collection process, we can rule out spatial correlation between weather and locality in the sampling grid. Weather effects on PAHs over a short-term period like ours have not been studied (although see Boquete et al., 2011 for heavy metals in lichens). Temporal studies using monthly weather conditions show large seasonal trends in both bio-accumulated and direct PAH measurements (Augusto et al., 2013b; Foan et al., 2015) whereas mapping projects tend to collect fewer samples within in a much shorter window, often 3 days, to limit the potential confounding effects of weather.

The importance of weather is complex because ambient conditions exert numerous, interdependent controls on PAH behavior in the atmosphere, including interactions with vegetation surfaces (Pratt et al., 2018). We examined weather covariates included in prior laboratory and field studies for moss and lichens (Augusto et al. 2013a, 2013b, 2013b; Wu et al., 2014), although for the explicit purpose of untangling their effects, models should allow for interaction terms and encompass a much wider range of weather conditions than we inadvertently did. Therefore, we interpret our weather results broadly, as to whether they make basic sense for each PAH group, with the critical takeaway being the startling importance of weekly and daily weather variability and its implications for PAH mapping studies - even those using short-term sampling windows.

Overall, weather results were generally consistent with past work. Since temperature controls PAH partitioning between the atmosphere and vegetation surfaces for gaseous PAHs (Simonich and Hites, 1995; Franzaring, 1997), the negative association with the highly volatile naphtha group should be expected. A positive effect of humidity on both particulate groups is also intuitive because moisture increases moss capacity to trap particles by greatly increasing surface area and presumably also exposing PAH binding sites on cell walls (Kylin and Bouwman, 2012). While we did not observe the expected temperature effect for the low MW group, the negative effect of precipitation is consistent with atmospheric studies showing rainfall is an effective scavenger for the more volatile PAHs (Arellano et al., 2018; Sahu et al., 2004).

4.4. Sample characteristics

Several local circumstances of how samples were collected had apparent effects on PAH concentrations. The inability to standardize tree species is a common problem for studies using epiphytes, as is the patchiness of biomonitor cover on the tree. The expectation of more within-tree 'microvariability' for particulate PAHs than gas PAHs is supported by our results; all but one significant covariate was identified for the particle-bound groups.

Lower concentrations of high MW PAHs with sampling height on the tree reflects the steep deposition curve for heavy PAHs, supporting our conclusion that short-distance horizontal dispersion in near-road environments may limit effects of surrounding deciduous canopy cover. Accordingly, the high MW group was not affected by characteristics of the host tree as represented by taxonomic family. Accumulation of the medium MW PAHs, however, was appreciably greater on trees from *Rosaceae* and *Aceraceae.* Other studies document high particle entrapment by dense crowns due to higher 'micro-turbulence' (Beckett et al., 2000; Dzierżanowski et al., 2011). *Rosaceae* species were small-statured with markedly dense crowns. *Aceraceae* crowns varied, although canopies of the other commonly sampled family, *Betulacae,* are naturally sparse. Other nuance affecting air flow or water drainage are probably important, as evidenced by higher medium MW concentrations in collections primarily from branches versus boles. Relict effects are also possible given that leaf shape, area, texture, and the nature of epicuticular waxes, are major drivers of PAH uptake by deciduous trees (Dzierżanowski et al., 2011; Howsam et al., 2001). Overall, results suggested *Rosaceae* and *Aceraceae* may be more efficient particulate scavengers and highlighted the considerable influence of host tree and sampling nuance on accumulation in an epiphytic moss.

4.5. Study Limitations

Our study had several limitations. First, our models explained only a modest proportion of the observed variability in PAH levels. Some unexplained variability probably relates to physical factors influencing PAHs dispersion that we could not characterize. For instance, we could not obtain consistent small-scale data on wind speed and direction, parameters which strongly affect pollutant dispersal and transport (Augusto et al., 2013b; Kim et al., 2015; Viskari et al., 1997). Besides relative elevation, we did not account for small-scale aspects of urban topography that may be influential (Wania et al., 2012), such as being located within a street canyon (De Nicola et al., 2013). Particulate matter from residential wood burning may also be an important sporadic source of PAHs that our models did not capture. Arguably, many of these unaddressed factors may be better characterized using dispersion models. While beyond the scope of this study, integrating biomonitor data and dispersion simulations might provide a more nuanced, comprehensive understanding of small-scale pollutant distributions (De Nicola et al., 2013; Iodice et al., 2016), helping overcome the difficulty of connecting emissions to deposition in complex urban environments (Galarneau, 2008, HEI Panel on the Health Effects of Traffic-Related Air Pollution, 2010).

A second limitation is that model and map interpretation may not be intuitive for some users. Small-scale proof-of-concept studies suggest bio-accumulated PAH levels can be equated with atmospheric concentrations (Augusto et al., 2013b; Loppi et al., 2015) across a narrow range of environmental conditions. Our PAH indices, however, predict only relatively higher or lower concentrations in moss, and are not translatable to absolute terms. Using geospatial models on our large-scale, intensive sample, moreover, required us to transform non-normally distributed datasets to meet statistical assumptions of the models. We also frequently used inverse-distance weighting because we felt it better represented the influence of predictor variables on PAHs in moss.

Finally, the standardization we did use (sampling land zoned as residential only) means we have low confidence in predictions for industrial zones. The sampling challenges in those areas (i.e. lack of trees, residences, and accessible public land) are difficult to overcome although if access to trees on private land parcels could be negotiated, *O. lyellii* is common there. On the other hand, we expect our maps represent the interface between residences and the road systems particularly well because site selection, being based on randomly selected addresses and contingent on obtaining landowner permission at the time of visit, led us to sample street trees in the public right-of-way more than half the time.

4.6. Policy and planning implications

While our finding of high PAHs in roadside environments is not new, our empirically based approach to visualizing fine-scale deposition gradients is. Observation of PAHs and other air toxics at human-relevant scales is a critical need for decision-makers in urban areas. While the moss-based PAH maps do not directly indicate where human health is at risk, they can inform city-planning (e.g. location of new bike lanes, parks), land-use on private property (e.g. placement of food gardens and play structures), and generally help prioritize where further monitoring and mitigation efforts are most needed.

The health consequences of living near roads are well-documented (Caiazzo et al., 2013; Chen et al., 2017, HEI Panel on the Health Effects of Traffic-Related Air Pollution, 2010). According to the CDC, about 11.3 million Americans (3.7%) live within 150m of a major highway (Boehmer et al., 2013). In Portland, over 90% of the houses we used for site selection were within 150m of one or more roads, including many smaller roads subject to heavy stop and go traffic. While a road's actual zone of influence depends on a multitude of factors and is difficult to predict (e.g. HEI Panel on the Health Effects of Traffic-Related Air Pollution, 2010, Pant and Harrison, 2013), 150m is a commonly used reference point for where particle deposition is expected to approach background levels (with fine particles and gases dispersing much farther; Batterman et al., 2020; Sucharová and Holá, 2014; Vette et al., 2013). Where possible, small-scale sampling of *O. lyellii* or other biomonitors could help define affected areas more specifically. Other groups in high contact with Portland's road system, besides many residents and drivers themselves, include the large community of bike commuters and a substantial portion of the homeless population. Estimated to be the 4th highest in the country (U.S. Department of Housing and Urban Development, 2018), homeless encampments are commonly established near highways on Department of Transportation land (Bassett et al., 2013) where the roadside pollution burden is high.

The relationship we observed between trees and atmospheric PAHs is policy relevant, as planting trees or other vegetation is feasible and inexpensive in most urban settings. While opinions vary on how to best implement green buffers, trees are already a well-used urban planning tool for reducing a variety of air pollutants (e.g. Janhäll, 2015; Tong et al., 2016; Wania et al., 2012). However, tree-planting is just one of many reduction strategies already underway in Portland. A variety of initiatives and programs, including the city and county's Climate Action Plan, the Central City in Motion Program (Portland Bureau of Transportation), and conversion of buses and other fleets to electric or hybrid motors, are expected to greatly improve roadside conditions. Future moss re-sampling could help evaluate our success over time.

5. Conclusions

Our ability to systematically collect and model a single moss species across Portland enabled us to provide a unique, integrative perspective of PAHs in residential areas. Maps provide a neighborhood-scale view that can inform land use planning and policies, with greater confidence in patterns predicted for the medium and high MW groups. Results may be cross-referenced with traditional modeling tools, like dispersion models, to increase their limited capacity to discover new or unanticipated air quality issues.

Environmental factors to address in future mapping studies include weekly and daily weather variability, tree canopy and associated characteristics affecting air and stemflow, and the distribution of moss across sampled surfaces. While stricter standardization of sampling conditions can reduce ‘noise’ in bio-monitoring studies, the main trade-off, a smaller and more opportunistically collected dataset, limits representativeness of the sample overall and also statistical power to observe important drivers not anticipated or controlled for in the field. While availability of the target biomonitor often dictates sample design, we recommend that future mapping studies challenge established assumptions about which parameters to control when possible. Our ability to observe sources of variability in this study pointed out both the complexity of environmental influences on PAHs in moss as well as our limited understanding of their interactions and mechanisms.

CRediT authorship contribution statement

Sarah E. Jovan: Conceptualization, Methodology, Investigation, Writing – original draft, Writing – review & editing, Supervision, Project administration, Funding acquisition. **Vicente J. Monleon:** Conceptualization, Methodology, Formal analysis, Writing – original draft, Writing – review & editing. **Geoffrey H. Donovan:** Conceptualization, Methodology, Investigation, Formal analysis, Writing – original draft, Funding acquisition. **Demetrios Gatziolis:** Methodology, Software, Formal analysis, Data curation, Visualization. **Michael C. Amacher:** Validation, Writing – review & editing.

Declaration of competing interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at doi: mmcdoino

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