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1 **Low-cost biomonitoring and high-resolution, scalable**
2 **models of urban metal pollution**

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19

20 **Keywords**

21 air pollution, stormwater pollution, automobile traffic, PM2.5, moss, Puget Sound

22

23 **Abstract**

24 As the global toll on human lives and ecosystems exacted by urban pollution grows, planning
25 tools still lack the resolution to identify priority sites where toxic pollution can be most
26 efficiently averted at a spatial scale that matches funding and management. Here we tackle this
27 gap by demonstrating novel scalable methods to monitor and predict urban metal pollution at
28 high resolution (< 5 meter) across large areas (10,000-100,000 km²) to guide pollution reduction
29 and stormwater management. We showcase and calibrate predictive models of Zn, Cu, and a
30 synthetic index of pollution for the Puget Sound region of Washington State, U.S., a densely
31 urbanized yet important ecosystem of conservation interest, and exemplify their transferability
32 across the entire United States. We leveraged widely and freely available datasets of car traffic
33 characteristics and land use as predictor variables and trained the models with biological
34 monitoring data of metal concentrations in epiphytic moss from > 100 trees based on new rapid
35 and low-cost protocols introduced in this study. Our model predictions, showing that 50% of the
36 total Cu and Zn pollution across the Puget Sound watershed is deposited over only 3.3% of the
37 land area, will allow cities to effectively and efficiently target toxic hotspots.

38

39 **1. Introduction**

40 The global expansion of urban areas over the past century has led to widespread
41 pollution, yielding a cascade of impacts on ecosystems and human health ¹⁻⁴. Car ownership
42 underlies much of this impact as the world's fleet continues growing, exceeding one billion
43 vehicles in 2015 ⁵. Ambient air pollution is now considered one of the leading causes of
44 premature death ⁶⁻⁸. Additionally, precipitation flowing over impervious surfaces transfers a
45 complex mixture of contaminants to streams and waterbodies — metals, polycyclic aromatic
46 hydrocarbons (PAH), pesticides and other pollutants emitted by urban activities are washed off
47 from streets and parking lots or leached from roofs and other construction material by
48 stormwater runoff and subsequently carried into nearby ecosystems ⁹. This stormwater runoff is
49 the fastest growing source of water pollution in many areas ¹⁰. With nearly 80% of the
50 population of the world projected to live in cities by the end of the 21st century ³, and the number
51 of vehicles expected to double or triple by 2050 ¹¹, it is imperative to tackle the negative effects
52 of urban dwelling.

53 Metals are among the pollutants of highest concern in urban areas due to their
54 persistence, bioavailability and toxicity ¹²⁻¹⁴. High concentrations of metals threaten aquatic
55 ecosystem functioning and biodiversity, as well as human health ^{10,15}. In cities, metals are
56 commonly reported pollutants, reaching waterways directly through atmospheric deposition and
57 indirectly through rain runoff by being leached or picked up as dust after depositing and building
58 up on impervious surfaces ¹⁶. Road dust naturally contains metals from eroded soil but
59 anthropogenic sources like building and road materials, vehicle brake, tire wear, and tailpipe
60 emissions are the main non-point sources of metal pollution in urban areas ¹⁷⁻¹⁹.

61 High-resolution estimates of pollutant sources are required to mitigate exposure to toxic
62 compounds by identifying the specific locations and associated site characteristics where the
63 deposition of metals is greatest²⁰. Pollution predictions must also be available for large
64 geographic areas to match the spatial scale of funding, management, and some environmental
65 issues⁸. Low-resolution (coarse) models cannot identify emission hot spots, as pollution sources
66 are averaged over large spatial units of analysis, and models developed for small spatial extents
67 may not match the ecological scale relevant to species targeted by management¹⁰. However, the
68 few models that focus on metals usually lack the combination of resolution and scale for this
69 purpose (but see²¹⁻²³). Therefore, high-resolution, scalable models that describe the relative
70 magnitude of metal pollution sources across space are needed to guide investments across large
71 areas.

72
73 Further complicating the training and validation of high-resolution models of urban metal
74 pollution is the dearth of monitoring data at the appropriate temporal and spatial scales. For
75 stormwater management, infrastructure siting requires long-term average metal emission
76 measures in flowing and standing water bodies, yet common monitoring data usually represent
77 snapshots in time subject to high sub-daily, daily, and seasonal variability²⁴. Air quality
78 monitoring networks, on the other hand, measure the concentration of metals in Nfine (< 2.5 μm
79 diameter – PM_{2.5}) and coarse (2.5–10 μm diameter – PM₁₀) particulate matter²⁵⁻²⁷ through costly
80 protocols. Consequently, the geographic density and temporal grain of metal pollution
81 monitoring is currently insufficient to train fine scale models of pollution, and most developing
82 countries lack the resources to install such monitoring infrastructure. Affordable approaches to
83 assess metal pollutant emissions integrated over long time periods thus hold the potential to

84 greatly facilitate air pollution and stormwater management across large spatial scales and in
85 areas with limited resources^{28,29}.

86

87 The aim of this study was to develop scalable methods to monitor and predict urban
88 metal pollution at high resolution to guide pollution reduction and stormwater management.
89 Here, we showcase and calibrate these methods for the Puget Sound region of Washington State,
90 U.S., a densely urbanized yet important ecosystem of conservation interest³⁰. We modeled long-
91 term average pollution levels for two metals of concern, Zn and Cu, as well as a pollution index
92 combining the pollution levels of these two metals with Pb. The model was trained with an
93 extensive set of passive biological monitoring data. We measured metal concentrations in
94 epiphytic moss collected by in-situ portable X-Ray Fluorescence (XRF) spectroscopy for > 100
95 trees based on new protocols introduced in this study. Lastly, we exemplify the utility of the
96 models by assessing its ability to predict metal air pollution across the U.S.

97

98 **2. Materials and Methods**

99 *2.1. Overview*

100 We first compiled spatial data for six of the main variables that have been reported to
101 drive the spatial distribution of urban metal pollution³¹⁻³⁵: land use; motor vehicle traffic
102 volume, speed, and congestion; road gradient; and over-ground public transit frequency (TOC.1).
103 Second, we generated pollution source “density maps” that simulate the cumulative and diffuse
104 emissions from each road-based source of pollution (TOC.2). In these source maps, each location
105 is assigned the distance-weighted sum of the value of the variable (e.g. traffic volume) across the
106 entire area within a buffer around that location. This allowed us to account for the cumulative

107 effect of the variable from multiple roads around every location while accounting for pollution
108 concentrations decay away from each road. Third, we measured the concentrations of metals in
109 epiphytic moss at 87 sites across the greater Seattle area (WA, U.S.) using portable XRF
110 (TOC.3). Fourth, a set of multiple regression models were developed using the density maps
111 values and land use at the sampling sites as candidate predictor variables and the XRF
112 measurements as response variables (i.e. training data, TOC.5). Three models were developed
113 separately for Zn, Cu, and the synthetic pollution index (PI). Zn, Cu, and Pb were selected as our
114 pollutants of study as these elements are among the most commonly studied and reported metal
115 pollutants in urban environments³⁶⁻³⁸. Zn and Cu, in particular, are closely associated with
116 automobile traffic volume and congestion through the emission of rubber tire dust and brake
117 wear dust, respectively^{18,37}. The three trained models were subsequently used to make spatially
118 continuous predictions of the relative concentrations of each pollutant across the entire study
119 area (TOC.6). Lastly, we predicted Zn pollution around every air quality monitoring station from
120 the United States Environmental Protection Agency (EPA) that recorded ambient air
121 concentrations of Zn in the form of fine particulate matter (TOC.7). This allowed us to assess
122 how well our predictions fit standard measurements of long-term average air pollution. See
123 Appendix 1 for a detailed description of the methodology and data sources.

124

125 2.2. *Study area*

126

127 The Puget Sound region encompasses 41,500 km² of upland, freshwater, estuarine and
128 marine habitats, and currently supports a large and increasingly urban population from
129 Vancouver, British Columbia (Canada) to Olympia, Washington (U.S., Fig. 1). Population
130 projections suggest that human numbers in the greater Puget Sound region will increase by two

131 million in the next 30 years, and Seattle, Washington, the largest city in the region, is one of the
132 fastest growing in the country ³⁹. With over 40 species of birds, mammals, fishes, plants and
133 invertebrates currently listed as threatened, endangered, or as candidates for state and federal
134 endangered species lists, Puget Sound is considered a “hot-spot” of extinction risk ³⁰.
135 Importantly, some of these imperiled species, such as Chinook salmon
136 (*Onchorhynchus tshawytscha*) and killer whales (*Orcinus orca*), are regional icons that have
137 been commemorated in art, culture and tradition for millennia (e.g. ⁴⁰).

138

139 *2.3. Candidate predictor variables: data sources and pre-processing*

140 We used the Percent Developed Imperviousness dataset from the 2016 Multi-Resolution
141 Land Characteristics Consortium's National Land Cover Data (NLCD, spatial resolution of 30 m)
142 ⁴¹ as a continuous measure of land use intensity. Because the effects of urban land-use on air and
143 water pollution are diffuse, two spatially smoothed imperviousness layers were also generated
144 from this dataset with 3x3 pixel mean and median filters.

145 A single spatial road network dataset with traffic volume (Annual Average Daily Traffic)
146 and speed was compiled from the U.S. Department of Transportation (DOT) and the U.S. Census
147 Bureau (Table S1). The slope of each road segment in the network was calculated from the 3D
148 Elevation Program Digital Elevation Models (Table S1) at the highest resolution available for
149 every location by first smoothing the elevation dataset and then computing the ratio of each road
150 segment's elevation range and length. To quantify traffic congestion, we computed the average
151 traffic flow across the area of interest based on data from the Bing™ Maps REST Services
152 Application Programming Interface (API). To do so, we built a custom tool similar to Tostes and
153 colleagues' ⁴². We applied our tool to obtain traffic conditions throughout the Puget Sound

154 watershed area from November 28th, 2018 at 08:00 am to December 5th, 2018 at 08:00. For Air
155 Quality Monitoring Stations, the tool was run from April 9th at 18:00 2019 to April 16th at 18:00
156 2019, resulting in 169 hourly snapshots.

157 Maps of over-ground public transit frequency were generated for the Puget Sound
158 watershed and within the vicinity of Air Quality Monitoring Stations based on static General
159 Transit Feed Specification (GTFS) data from the National Transit Map (Table S1) — the largest
160 seamless compilation of GTFS data in the U.S. at the time of this study. The result provided us
161 with a single seamless layer of the average weekly number of times that a public transit vehicle
162 went through every location across the landscape, thus integrating across all agencies, modes of
163 transport, routes, and schedules.

164

165 *2.4. Pollution spread models*

166 To simulate the spread of pollutants away from roads and thus the cumulative pollution
167 from multiple roads, we generated pollution source density maps of public transit frequency,
168 road gradient, and traffic volume, speed and congestion. For each density map, the value at each
169 location was calculated as the distance-weighted sum of all pixel values within a maximum
170 distance from that location, whereby the weight attributed to each pixel was determined by a
171 distance-decay function. To encompass various possible rates and functional forms of metal
172 particle deposition, we generated 24 candidate density maps for each of these five road-based
173 variables based on a set of 24 different distance-decay functions, each function illustrating a
174 different hypothesized pattern and distance of pollution spread (see Appendix 1). The resulting
175 120 continuous maps of pollution sources were subsequently used as candidate predictor
176 variables to select and train the models of metal pollution across the Greater Seattle area.

177

178 *2.5. Data collection and model training*

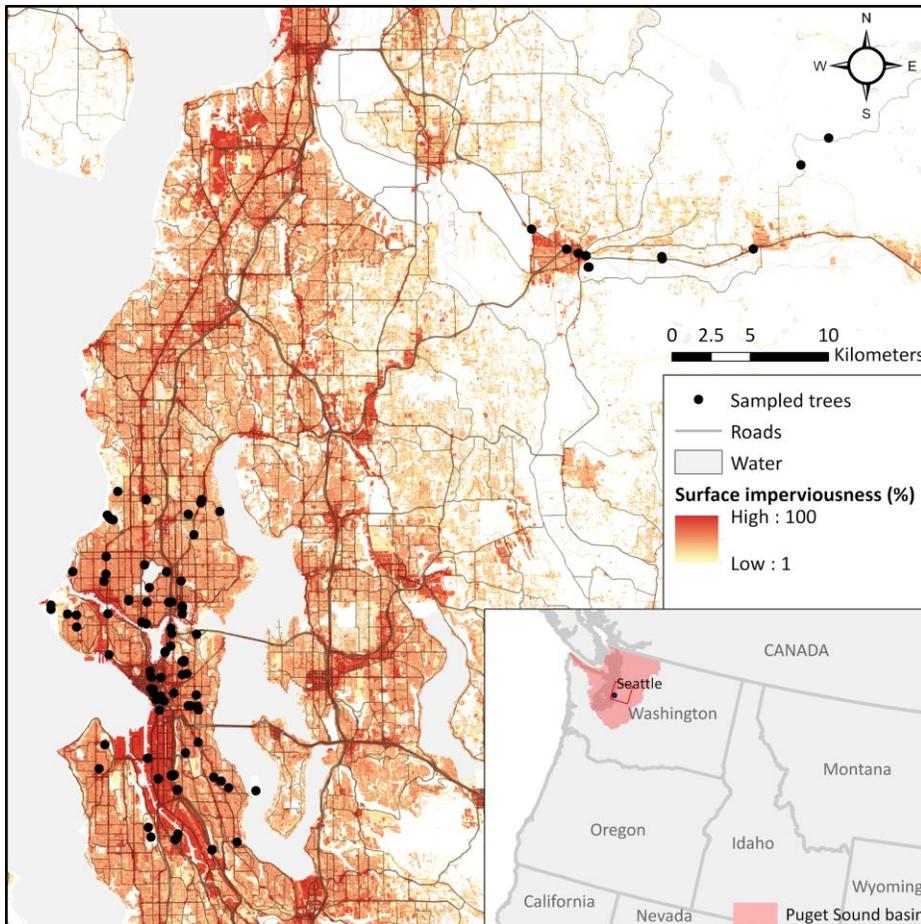
179 We used the elemental composition of epiphytic moss on trees from 87 sites determined
180 by portable XRF to train and validate our predictive models of pollution (Fig. 1, see Appendix 1
181 and [Messenger et al. companion paper] for more information on sampling design and protocols).
182 Mosses have been widely used in Europe as biological monitors of metal pollution for decades,
183 because they readily accumulate pollutants over time, reflecting long term pollution levels ^{43,44}.
184 Unlike plants, they lack roots and a cuticle, so they absorb nutrients from the atmosphere over
185 their entire surface and all cells are directly exposed to wet deposition ⁴⁵. Pollutants in dust also
186 accumulate on their surface through dry deposition ⁴⁶. Their high cation exchange capacity
187 results in moss cells passively trapping pollutants, including metals ⁴⁵. As a result, mosses more
188 readily accumulate atmospheric metals than vascular plants, and metal concentrations in moss
189 pseudo-tissues are not as influenced by local variations in soil metal levels ^{29,47}. In addition,
190 mosses are ubiquitous in the study region, in both rural and urban settings, and persist even in the
191 most polluted urban sites. Leveraging these characteristics, multiple studies have successfully
192 used elemental analysis of mosses to assess the spatial distribution and sources of urban and
193 motor vehicle traffic-related metal pollution with high specificity ^{28,31,48}.

194

195 For this study, a new protocol of rapid in-situ elemental analysis by portable XRF was
196 developed. With this method, photons are emitted from an X-ray tube in a portable instrument
197 whose measuring window is directly applied against moss mats growing on tree bark. The
198 emitted photons interact with the atoms within the moss sample, which in turn emit photons at
199 energy levels that are specific to each element. Photons from the moss atoms are then received,

200 counted, and converted by the instrument into a spectrum of energy with clearly identifiable
201 peaks whose size reflects the relative quantity of each element in the sample ⁴⁹. Portable XRF is
202 now a common tool whose performance is comparable to more conventional methods like
203 benchtop XRF and inductively-coupled plasma-optical emission spectrometry (ICP-OES) for the
204 analysis of hazardous metal concentrations in soils and plants ⁵⁰⁻⁵². We show in [Messenger et al.
205 companion paper] that, compared to quantitation by ICP-OES, pXRF enables quantitative
206 screening of Cu (pseudo- $R^2 = 0.75$) and Zn (pseudo- $R^2 = 0.81$), and qualitative screening of Pb
207 ($R^2 = 0.63$), with high measurement replicability. While epiphytic moss has previously been
208 used for urban metal pollution mapping ²⁸, this is, to the authors' knowledge, the first time that
209 portable XRF was used for biological monitoring of fine-scale spatial patterns in ambient metal
210 pollution.

211
212 Although 87 different sites were selected for sampling, measurements were separately
213 taken from two closely adjacent trees in a quarter of the sites to assess small-scale variations in
214 metal concentration. Moss mats were thus sampled on a total of 109 trees (measurements from
215 single trees, rather than sites, are hereon used as our unit of analysis). Sampling took place in
216 July-August 2018 and March-April 2019. As various studies have shown that air-moss metal
217 concentration relationships vary among species ⁵³, measurements were taken from a single moss
218 species, *Orthotrichum lyelli* Hook. & Taylor growing on hardwood tree species. Portable XRF
219 assays were obtained from three different moss mats on each tree using a Bruker Tracer III-SD
220 (T3S2606, Bruker Elemental, Kennewick, WA, U.S.). Spectral data were processed and
221 normalized to yield a comparable 'concentration' index across samples using standard analysis
222 protocols (See Appendix 1 and [Messenger et al. companion paper] for details on the protocol) ⁵¹.



223

224 **Figure 1.** *Spatial distribution of sampled trees used in model development (black points, after*
 225 *outlier removal: $n = 107$), roads (grey lines, excluding local roads) and surface imperviousness*
 226 *(National Land Cover Dataset 2016) across the Greater Seattle area (Washington State, U.S.),*
 227 *the most densely urbanized region of the Puget Sound watershed (pink polygon in inset map).*

228 **[1.5 column-fitting image]**

229

230 We developed three separate models for Cu, Zn, and a pollution index. These multiple
 231 regression models were developed through model selection⁵⁴, all using the density map values
 232 and imperviousness at the sampling sites as candidate predictor variables and the element
 233 concentrations as response variables (see Appendix 1 for a detailed description).

234 The pollution index was computed with the following equation for each sampled tree i :

235
$$\text{Pollution index}_i = \left(\frac{[\text{Cu}]_i}{[\text{Cu}]_{ref}} \times \frac{[\text{Pb}]_i}{[\text{Pb}]_{ref}} \times \frac{[\text{Zn}]_i}{[\text{Zn}]_{ref}} \right)^{1/3} \quad (\text{Equation 1})$$

236 Where $[\text{Element}]_i$ is the element's normalized net count rate averaged over the three XRF assays
237 at that site and $[\text{Element}]_{ref}$ is the background normalized net count rate (after within-site
238 averaging) for that element recorded at a reference site away from pollutant sources.

239 After selecting a reduced set of predictor variables through preliminary selection, a
240 systematic model selection was conducted to develop a multiple regression model for each
241 response variable⁵⁴. We then assessed whether including sampling season (summer 2018 or
242 spring 2019) as a fixed effect improved the model, to take in account the potential influence of
243 temperature and precipitation on moss pollution retention⁵⁵⁻⁵⁷. The model residuals were also
244 evaluated for spatially collinearity; we built spatial simultaneous autoregressive error models
245 (linear regression models accounting for spatial error structure) when appropriate⁵⁸.

246

247 *2.6. Model predictions and fit to air pollution measurements*

248 Using the trained models described above, we made spatially continuous predictions of
249 pollution for the Puget Sound region and around every air quality monitoring station operated by
250 the EPA across all states of the contiguous U.S. The latter allowed us to assess whether our
251 estimates of metal concentrations from a model trained with local bio-monitoring data could be
252 related to more standard measurements of air pollution beyond our focal area. We selected all air
253 quality monitoring sites in the conterminous U.S. with at least 200 measurements of Zinc PM_{2.5}
254 ($\mu\text{g}/\text{m}^3$) from 2014 to 2019 and extracted the Zn concentration estimated by our model (Zn_{pred})
255 for each of these sites. As spatial and temporal variations in air pollution are strongly influenced
256 by local climatic and meteorological factors, we also computed 37 variables identified as key
257 drivers of PM_{2.5} through 50th percentile quantile regressions by Porter and colleagues⁵⁹. To

258 assess the adequacy of Zn_{pred} for predicting $Zn PM_{2.5}$ records, we compared two partial least-
259 square regression (PLSR) model of $Zn PM_{2.5}$ (after logarithmic transformation): the first only
260 included meteorological variables as predictors whereas the second included those 37 variables
261 together with Zn_{pred} .

262

263 **3. Results and Discussion**

264 *3.1. Observed distribution and drivers of metal pollution in the study area*

265

266 XRF assays of epiphytic moss revealed that numerous metals were prevalent throughout
267 the Puget Sound watershed. Their concentrations covaried with urban land use and motor vehicle
268 traffic, often ranging in concentration by an order of magnitude above background levels
269 (Appendix 2). Metals detected by our sampling included Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sr, Ti,
270 Zn, and Zr. Notably, Zn concentration in the most polluted site was 23 times higher than in the
271 most isolated area, whereas Cu varied by a factor of 10 throughout the area. The concentrations
272 of metals measured by XRF sampling also covaried among elements (Appendix 3, Fig. S1). This
273 further justified our focus on a reduced set of elements (Cu, Zn, and Pb) that could serve as
274 indicators for a broader suite of metal pollutants. Clusters of elements revealed common
275 physicochemical properties, patterns of spatial dispersion, and potential sources. High
276 concentrations of Zn, Co, Fe, Zr, Cr, and Ti tended to co-occur ($0.6 < \text{Pearson's } r < 0.9$), Cu
277 exhibiting slightly different distributional patterns, whereas Pb was clustered with As. The
278 observed correlations among metals can be attributed to their co-occurrence in automotive parts
279 and in crustal dust, as traffic leads to non-exhaust emissions through both abrasion (i.e. of brake
280 pads and tires) and dust resuspension^{18,60}.

281 Applying our rapid and low-cost biomonitoring protocol across a dense array of sites, and
282 combining these measurements with widely available spatial data on potential pollution sources,
283 allowed us to shed light on the traffic and land use characteristics most responsible for the
284 occurrence of different metals in urban environments. This analysis showed for example that Zn
285 and Cu concentrations are both strongly correlated with traffic speed ($r > 0.5$). Zn also strongly
286 covaries with traffic volume and congestion ($0.6 < r < 0.7$) whereas Cu is correlated with bus
287 transit volume ($r > 0.6$, Appendix 3, Fig. S2). The distance-decay functions used to produce the
288 density maps of pollutant predictors that were most correlated to metal concentrations further
289 hinted at the relative distances to which different metals disperse away from their source of
290 emission. For instance, Zn is slightly more correlated to traffic speed and volume density maps
291 generated with 100 m kernels compared to those based on 200 m kernels or larger. On the other
292 hand, Cu is most correlated to density maps produced with the largest kernels of dispersion for
293 every pollution predictor. Closely associated with tire wear, Zn in dry deposition is usually
294 associated with larger particle size ($> 10 \mu\text{m}$) whereas Pb, Cd, Ni and Cu are mostly present in
295 smaller particle sizes ($< 10 \mu\text{m}$)³⁷. As particles $< 10 \mu\text{m}$ behave more like gases, diffusing along
296 concentration gradients to deposit on surfaces, they tend to travel further than coarser particles
297 whose deposition rates are more gravity-driven, explaining the differences in spread patterns we
298 observed between Cu and Zn⁶¹.

299

300 *3.2. Model training and validation results in the Puget Sound watershed*

301

302 The models of pollution developed here — for Zn, Cu, and a pollution index — relied on
303 only two to three predictors each, and all yielded estimates with average percentage errors from
304 20 to 30% (Fig. 2). These three models explained 65%, 64% (78%) and 57% (72%) of the

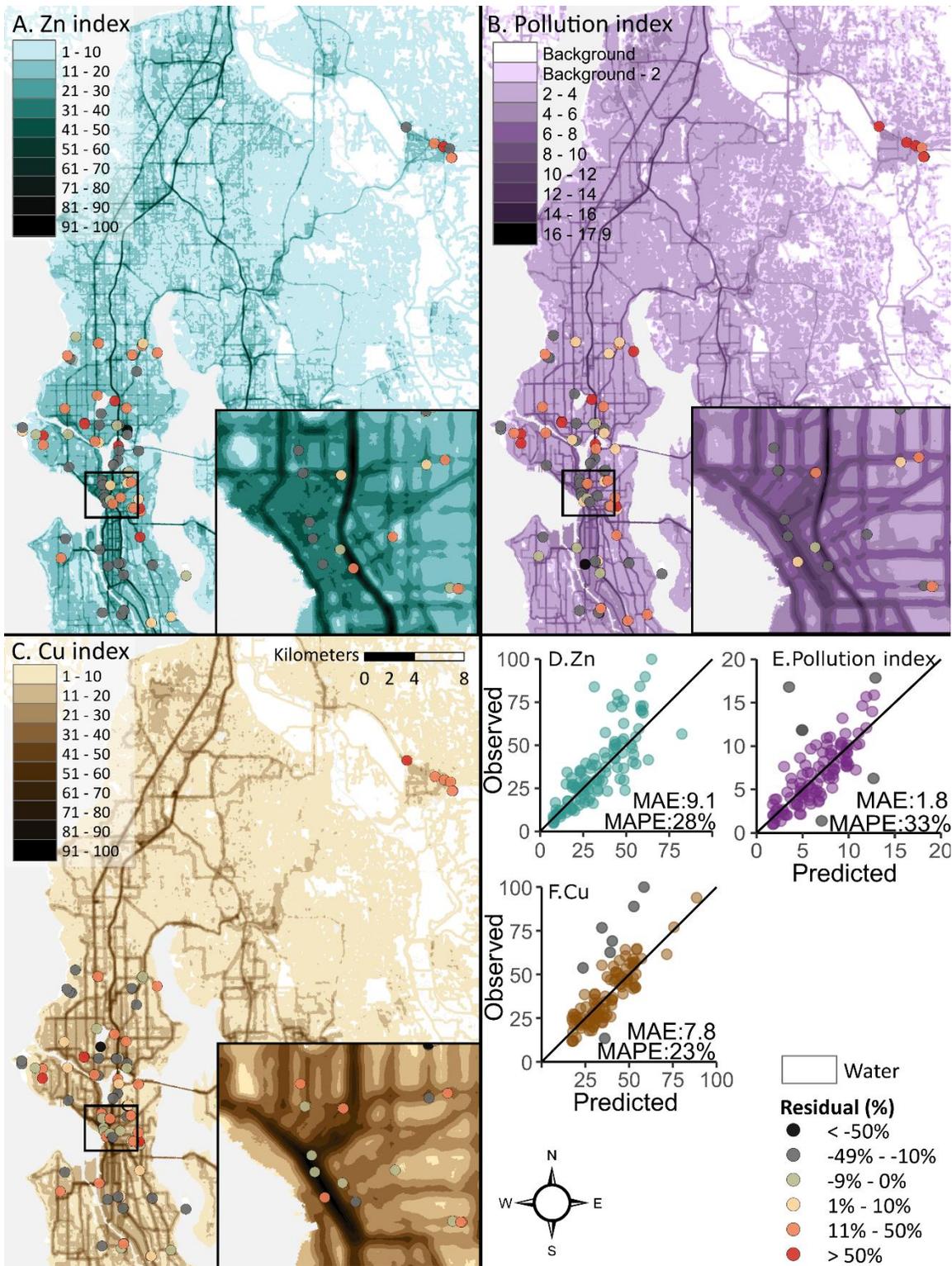
305 variability in Zn, Cu, and pollution index across sampled sites, respectively, based on pseudo-R²
306 computed with (and without) outliers (Fig. 2). For a full list of candidate models, performance
307 and diagnostic tests, refer to Appendix 4. To achieve normality and homoscedasticity of
308 residuals, the regression models of Cu and pollution index were trained without outliers (grey
309 points in Fig. 2 scatterplots). Sampling season did not have a significant effect ($p > 0.05$) in the
310 models. Regression coefficients from autoregressive error models with inverse distance spatial
311 weight matrices based on one nearest neighbor were used for model predictions in all three cases,
312 as Lagrange Multiplier robust diagnostic tests revealed spatial error dependence in the initial
313 simple linear regression models. The metal pollutant concentrations were thus estimated
314 according to the following regression models (based on Zn and Cu values maximum-
315 standardized to 100):

316
$$Zn = \exp(2.042 + 0.417 \cdot volume_{\log 100}^{1/4} + 0.012 \cdot imperviousness_{smooth}) \text{ (Equation 2),}$$

317
$$Cu = 17.522 + 5.808 \cdot transit_{\log 200}^{1/2} + 0.142 \cdot imperviousness_{smooth} \text{ (Equation 3),}$$

318
$$Pollution\ index = 1.783 + 0.048 \cdot volume_{\log 100} + 1.690 \cdot transit_{linear 100}^{1/3} + 0.031 \cdot imperviousness_{smooth} \text{ (Equation 4),}$$

319 where $volume_{\log 100}$ is the value from the density map of average annual daily traffic created with
320 a 100 m-log kernel, $transit_{\log 200}$ and $transit_{linear 100}$ are produced with bus transit density maps
321 with 200 m-log and 100 m-linear kernels, respectively, and $imperviousness_{smooth}$ is based on a
322 3x3 median filter of imperviousness.



323

324 **Figure 2.** Prediction maps and scatterplots of observed vs. predicted metal pollution for (A, D)

325 Zinc, (B, E) Pollution index, and (C, F) Copper (respectively). Map values for Zn and Cu are

326 *minimum-maximum standardized to describe the spatial distribution of metal concentrations*
327 *across the area, 0 reflecting “background” metal concentrations presumably due to local crustal*
328 *deposits at the least impacted site, and 100 corresponding to the highest predicted value across*
329 *the Puget Sound watershed. For all models, Mean Absolute Percentage Error (MAPE) and*
330 *Mean Absolute Error (MAE, of the standardized indices) were below 30% and 10, respectively.*
331 *Residuals show percentage error of predictions at sampled sites based on XRF-measured metal*
332 *pollution. Insets show the heavily trafficked downtown area of Seattle adjacent to a section of the*
333 *Interstate Highway 5 where a dense array of measurements were conducted.*

334 **[2 column-fitting image]**

335

336 Our results echo previous studies relating motor vehicle traffic characteristics with
337 measured air, soil, and stormwater pollution. Horstmeyer and colleagues³², for instance,
338 document the greatest Pb, Zn, and Cu concentrations in the topsoil of vegetated infiltration
339 swales at sites with high traffic volume or with frequent braking and acceleration, at crossings,
340 roundabouts and in stop-and-go traffic. Cu and Zn concentrations in mosses along transects of
341 nine roads also increased with traffic density and the incline of the roads in Austria³¹. While
342 additional predictors could have been added to increase the performance of our model, we
343 instead focused on using widely available, free-to-access data to maximize the scalability of our
344 model and its transferability to varied geographical contexts. The fact that our models including
345 bus transit best predicted Cu was a surprising finding, but we conjecture that bus transit routes
346 reflect major transportation axes also used by heavy commercial motor vehicles (i.e. trucks) and
347 thus buses do not solely account for the pollution patterns. Pb was not as well predicted by traffic
348 and land use as Zn and Cu. This lower predictive ability goes against our expectations as Pb,

349 despite having been phased out of gasoline for several decade, remains a prevalent component of
350 motor vehicle parts ^{18,62}. Point-source pollution spreading over large distances and the age of
351 surrounding buildings may be more strongly driving the spatial distribution of this pollutant ⁶³.
352 For the development of more local models, we therefore posit that additional sources of metal
353 pollution to account for, when available, should include: heavy-duty motor vehicle traffic ³²,
354 building density and age ⁶³, parcel-level land use (e.g. parking lots ³²), road surface material ⁶⁴,
355 commercial railways ⁶⁵, and locations of point-source pollution ²⁸.

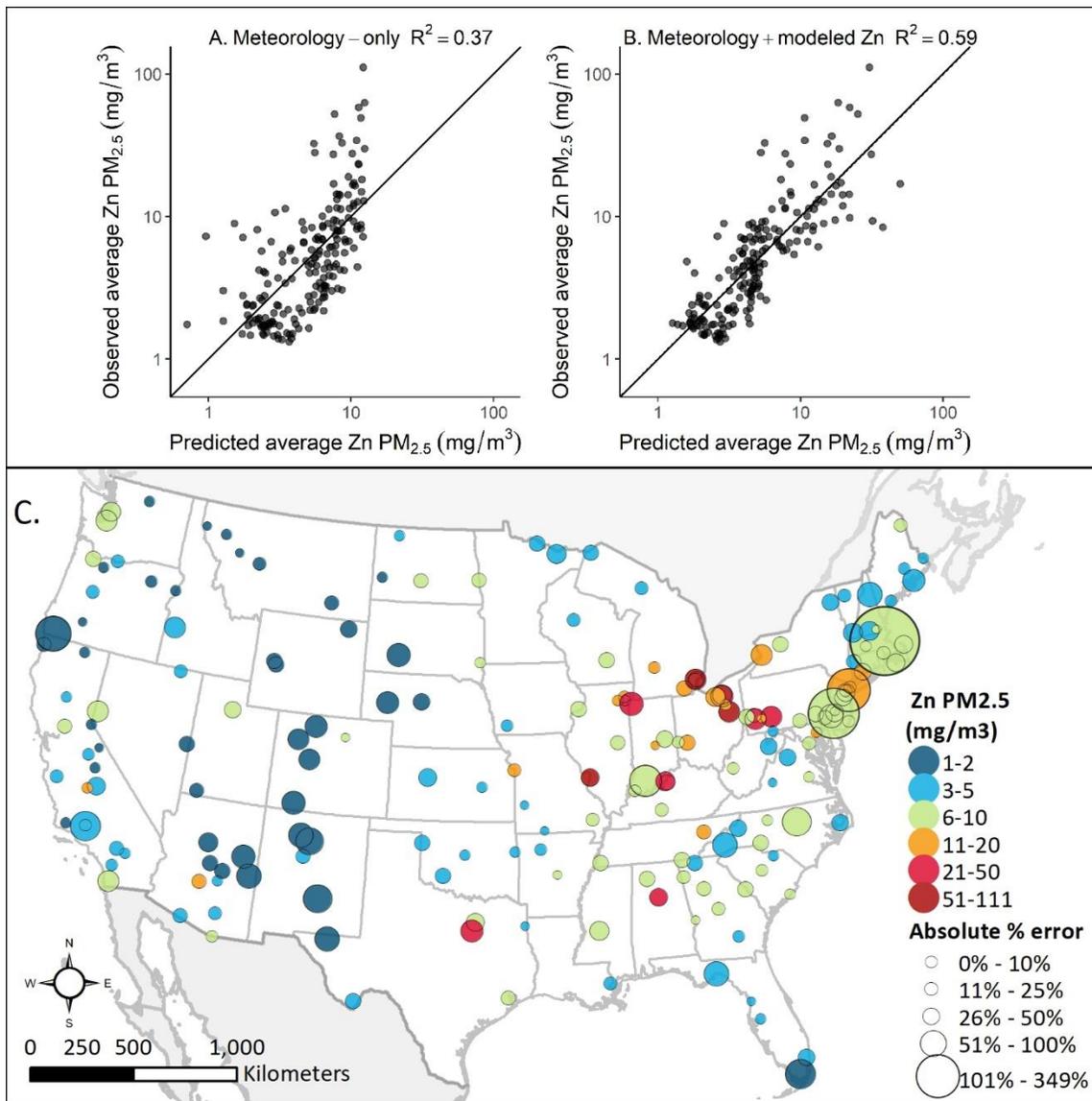
356

357 *3.3. Model transferability: comparison to nationwide monitoring data*

358

359 Although our models were trained with metal concentrations in epiphytic moss from across
360 the Puget Sound watershed, they could also predict air metal pollution at the continental scale
361 with relative accuracy. Together with long-term meteorological and climatic predictors, our
362 estimates of Zn pollution explained 59% of the variability in long-term average Zinc PM_{2.5}
363 ($\mu\text{g}/\text{m}^3$) across all 195 air quality stations with at least 200 daily observations (2014-2019) across
364 the U.S. (Fig. 3). Here we focused on assessing the ability of our models to predict Zn in fine
365 particulate matter (PM_{2.5}). However, Zn, Cu and Pb emissions are associated with a range of
366 particle sizes that might be better predicted by our model (e.g. PM₁₀ and coarser ^{18,66}). In
367 addition, although the goal of our US-wide analysis was to assess the relevance of our model
368 estimates regarding long-term air metal pollution, more complex models accounting for temporal
369 (weekly and seasonal) and spatial autocorrelation could achieve greater predictive ability.

370



371

372 **Figure 3.** Predictive performance of (A) a meteorology-only partial least-square regression
 373 (PLSR) and (B) a PLSR combining meteorological variables and the Zn index estimated by the
 374 model developed in this study as predictor variables for (C) long-term average Zn PM_{2.5} (color,
 375 mg/m³) recorded at 195 air quality stations across the United States from 2014 to 2019. The
 376 black diagonal lines (A and B) are 1:1 lines. The points in the map (C) are sized according to the
 377 absolute percentage error of the [meteorology + modeled Zn] PLSR relative to measured Zn
 378 PM_{2.5} at the stations. [2 column-fitting image]

379 The biomonitoring method and modelling framework demonstrated here provide a unique
380 combination of scale and resolution. They can be leveraged for fine-scale decision-making to
381 control a family of pollutants that is rarely modeled yet whose threat to the environment is
382 increasingly recognized ¹⁰. A large portion of urban metal pollution is initially emitted, or
383 resuspended, to the atmosphere before settling on the road surface and being washed off by
384 stormwater^{35,67,68}. Therefore, moss biomonitoring and the associated predictive models can
385 inform stormwater management in identifying hot spots of metal emission by these sources
386 located upstream from waterbodies of interest. Although moss has been used across Europe to
387 map large scale patterns of pollution ⁶⁹, only a handful of studies have demonstrated the use of
388 bryophytes for assessing fine scale patterns of pollution ³¹. Using portable XRF, we were able to
389 assess the concentration of 12 metals across 87 sites in about five minutes at each location,
390 allowing for fine-scale assessment of the spatial distribution of pollution. By contrast, there are
391 only two stations with long-term Zn PM_{2.5} data in the Puget Sound watershed. The models can
392 also be further adapted with moss measurements from other locations of interest. Because we
393 trained our models based on widely and freely available datasets of pollution predictors, this
394 framework can be leveraged to estimate metal pollution at high resolution (here ~ 5 m) in other
395 contexts and across large areas. Outside of the United States for example, the road network from
396 the Open Street Map dataset, a global crowd-sourced collaborative mapping project, is nearly
397 complete ⁷⁰, and our novel tool for assessing the degree of traffic congestion using Bing TM could
398 be applied in > 70 countries at the time of this study ⁷¹. Traffic congestion data could also be
399 more easily obtained through the establishment of custom user agreements with companies
400 specializing in the collection of traffic data (e.g. INRIX®). Lastly, sub-models and variable

401 substitution can be used to accommodate cases where predictor variables may be unavailable
402 (e.g. traffic volume) as multiple variables performed well in predicting pollution (Appendix 4).

403

404 *3.4. Sources of errors and uncertainty*

405 Multiple sources of uncertainty are associated with the framework introduced here, stemming
406 from the use of moss biomonitoring, XRF measurements, as well as in the predictor variable
407 data. The main source of uncertainty in moss-based biomonitoring is that metal levels in moss
408 pseudo-tissue do not reflect a passive and steady accumulation over time but a dynamic
409 equilibrium between concentrations in the environment, moss growth, and meteorology⁵³. We
410 review sources of uncertainty in moss measurements in Appendix 5, including precipitation,
411 proximity to saltwater, pseudo-tissue age and growth rate. Field sampling was conducted across
412 two different seasons due to maintenance requirements for the XRF instrument — while we did
413 not find a significant effect of season in our models, we recommend that future studies conduct
414 all sampling within a single season or re-sample the same sites across different periods of the
415 year to better capture seasonal variability⁴⁴. Due to these uncertainties, moss biomonitoring is
416 best suited for assessing the spatial distribution of pollution in relative terms rather than
417 determining absolute levels for the evaluation of pollution criteria compliance. Regarding field
418 XRF measurements, the main uncertainty resides in the heterogeneity of moss samples growing
419 on trees, resulting in unequal densities of pseudo-tissue and depth of measurements. Nonetheless,
420 the low level of tissue differentiation and the lack of a cuticle greatly reduces variability in XRF
421 measurements of moss compared to vascular plants⁵¹. One additional limitation of XRF is that
422 the prevalence of some commonly used tracers of non-exhaust traffic pollution (e.g. Sb,
423 Ba^{60,72,73}) cannot be accurately determined at the same time as those discussed in this study;

424 these elements' characteristic energy lines occur outside of the region that the instrument was set
425 to measure (through the use of a yellow filter, see Appendix 1 for details on protocol). Owing to
426 its affordability and speed, the resulting ability to compute averages over replicated
427 measurements, and the minimal disruption of samples involved compared to conventional
428 methods (e.g. sample extraction, transport, processing, digestion), we believe that this protocol is
429 a significant advance towards high resolution monitoring of urban metal pollution.

430

431 *3.5. Predicted metal pollution across the Puget Sound watershed*

432 With the assurance that our models satisfactorily described the spatial patterns of the
433 three metal pollution indicators, we made predictions of metal pollution across the entire
434 ~40,000 km² of the Puget Sound watershed. These estimates show that the pollutant footprint of
435 human development spans across the entire landscape, with 27% of the watershed's land area
436 (excluding all 30x30 m pixels identified as water by the National Land Cover Dataset) exhibiting
437 metal concentrations above background levels for at least one of the indicators. Cu affects the
438 greatest area in the region, as expected from the larger size of the distance-decay function for bus
439 transit (Equation 3) in its computation. Our work also reveals that metal pollution is very
440 localized: 50% of the total pollution deposited on land across the Puget Sound watershed is
441 deposited over only 3.3% of the land area for both Cu and Zn (Fig. 2 and 3).

442

443 *3.6. Conclusion*

444 Effectively and efficiently combating pollution generated from motor vehicles is a major
445 challenge facing natural resource managers and urban planners. In this study, we demonstrate
446 the utility of new biomonitoring techniques that can provide unique insight into the spatial

447 distribution of pollution sources at a fraction of the cost of usual approaches. Intensive sampling
448 of epiphytic moss with pXRF revealed the existence of Zn and Cu emission hotspots in densely
449 urbanized areas of the Puget Sound watershed with metal concentrations exceeding background
450 levels by an order of magnitude. Combined with open access datasets to train predictive models,
451 our inexpensive, rapid, and rigorous approach allows managers to identify and mitigate toxic
452 metals at fine spatial scales over large geographic regions. Here we applied these models to
453 predict the concentration of metals at ~5 m resolution across the Puget Sound watershed and
454 around every air quality monitoring station in the contiguous U.S. With increasingly limited
455 resources, this new approach holds the potential for cities to target toxic hotspots, thus
456 maximizing environmental returns on mitigation investments.

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460

461 **Data availability**

462
463 All scripts used in this study are available for reuse (<https://github.com/messamat/traffic> for
464 Bing™ traffic flow analysis, https://github.com/messamat/stormwater_samplingPy for GIS
465 analysis and mapping, and https://github.com/messamat/stormwater_samplingR for sampling,
466 XRF data post-processing, and statistical analysis). All data are also available at [figshare URL].

467
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481
482 **Disclosures**

483 The authors declare no competing financial or other interest.
484

485 **Associated content**

486 See the accompanying Supporting Information for the full methodology (Appendix 1), a table of
487 the selected measured elemental concentrations and land use characteristics at sites sampled by
488 XRF (Appendix 2), cluster and correlation analysis results depicting the relationships among
489 selected elemental concentrations measured in epiphytic moss by XRF and land use and vehicle
490 traffic pollution predictors (Appendix 3), model development tables (Appendix 4), and a
491 literature review of the sources of uncertainty related to moss biomonitoring of ambient metal
492 pollution (Appendix 5).

493

494

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