Predicting springtime herbicide exposure across multiple scales in pacific coastal drainages (Oregon, USA)

Kaegan M. Scully-Engelmeyer [*], Elise F. Granek

Department of Environmental Science and Management, Portland State University, 1719 SW 10th Ave. SRTC 218, Portland, OR 97201, USA

ABSTRACT

Identification of non-point sources of watershed pollution such as pesticide runoff is challenging due to spatial and temporal variation in landscape patterns of land use and environmental conditions. Regional case study monitoring investigations can document region-specific conditions and processes to inform managers about pesticide movement through watersheds. Additionally, modeling field-collected data within these contexts can be used to predict pesticide presence in un-sampled areas. During a 45 day period in the spring of 2019, we sampled sixteen coastal watersheds in Oregon, USA for current-use water-borne herbicides commonly used in forestland vegetation management. At 80 % of sampling locations, at least one of four commonly used herbicides was detected in integrative passive water samplers, with hexazinone and atrazine most commonly detected. In this study, we use total accumulation of detected compounds to compare relative detections with upstream management and environmental watershed variables using multiple linear regression. An additive effects model was developed using slope, herbicide activity notified during the sampling window, and recent clearcut harvest notifications to predict variation in total herbicide accumulation ($R^2 = 0.8914$). The model was then applied to predict concentrations in un-sampled watersheds throughout the Oregon's coastal region at three watershed scales using Hydrologic Unit Codes (HUCs) 8, 10, and 12. Regional differences in predicted values were visualized using choropleth maps. Subwatersheds (HUC12) were grouped by subbasin (HUC8) and base mean predicted values were compared to further quantify regional differences. Models predicted that south coast sites have higher than average herbicide concentrations, which aligned with field-collected data findings.

1. Introduction

Offsite movement of pesticides throughout watersheds is a universal concern for managers and scientists, especially in light of research on sublethal effects of low dose exposures to aquatic organisms. Approaches to understand risk in these contexts vary, but a central challenge is collecting sufficient data at appropriate scales and time intervals in a cost-effective manner to make informed decisions about how pesticides affect aquatic ecosystems. Monitoring results from field collected data can be useful not only to inform managers about transport within the sampled locations but also to predict concentrations in un-sampled areas through modeling (Holvoet et al., 2007). Commonly in landscape scale research, multi-site comparisons and empirical modeling are implemented to record the influence of natural and anthropogenic variables - such as land-use, on in-stream conditions (Allan, 2004; Allan et al., 1997; Turner and Gardner, 2015). Such efforts can support better understanding of cumulative effects of land management practices on

water quality, specifically pesticide concentrations in watersheds of differing sizes.

Investigation into cumulative effects of intensive forestry on water quantity have found significant relationships between the scale of operations and their contribution to water quantity deficits in downstream waterways (Perry and Jones, 2017). Substantial research has focused on cumulative effects of many types of forestry practices (road construction, clearcutting, planting, etc..), but less is understood about the effects of multiple chemical applications within watersheds and the transport of chemical mixtures away from application sites (Clark et al., 2009; Norris et al., 1991). Pesticide application on forestlands is often downplayed in comparison to agricultural applications based on the frequency of occurrence (herbicide applications take place 1–5 years after clearcutting versus multi-annual applications on agricultural lands) until replacement conifers are established. Most research concerning chemical applications on forestlands is focused on site-level effectiveness, and data gaps remain on the effects of chemical

applications across larger spatial scales or multiple watersheds within regions (Neary and Baillie, 2016). Exploring the effectiveness of management practices at the site scale provides valuable and critical information, but looking at other larger scales may provide more accurate information on exposure by organisms within a watershed. Chemical applications in forestlands and agricultural lands take place in concert with other land use practices across the landscape, and should be considered within these contexts (Metcalfe et al., 2019; Norris et al., 1991). Additionally, valuable and protected resources are found across multiple watershed scales, highlighting the importance of looking beyond site-scale impacts to understand catchment or watershed level effects.

During late winter and spring, pre-emergent and site preparation herbicide treatments are commonplace in Oregon's coastal forestlands. Chemical site preparation treatments accompany mechanical, manual, and fire-based methodologies as vegetation control measures that take place within the first year of the original cutting before reforestation occurs (Rose and Haase, 2006). Once trees have been planted, pre-emergent or "dormant applications" are utilized to control competing vegetation before conifer bud break takes place in late spring (Peachy, 2020). Competing vegetation targeted in these applications range from herbaceous grasses and ferns to early successional woody species such as vine maple and alder. Dormant applications are commonly applied in mixtures to target a variety of early successional vegetation (Table 1). Rainfall during spring months in Oregon's Coast Range is substantial, and many compounds used in vegetation management during this period are rainfall activated products. Resultant runoff events following forestland pesticide application are generally characterized as episodic exposures, wherein "pulses'' of higher chemical concentrations move downstream followed by decreasing concentrations (Louch et al., 2017). The majority of forestry specific monitoring in the region has occurred during foliage applications occurring in the summer and fall months (Caldwell and Courter, 2020; Dent and Robben, 2000; Louch et al., 2017), with monitoring during spring runoff understudied. Despite the low number of spring season studies on forestlands in Oregon, the highest levels of pesticides are frequently observed during springtime runoff periods (Hapke et al., 2016; Kelly et al., 2012).

During spring and early summer in Oregon, changes in water temperature cue reproduction in several freshwater and estuarine species (bivalves, pacific lamprey, etc…) that inhabit coastal watersheds (Allard et al., 2017; Meeuwig et al., 2005). Since reproduction and larval life stages of aquatic organisms are considered the most sensitive to chemical contaminants (Bringolf et al., 2007; Cope et al., 2008; Perry and Lynn, 2009), understanding in-water concentrations of current-use herbicides during time periods coinciding with spring spray is critical to assess relative threats to non-target aquatic species.

Integrative sampling is a valuable method to explore in-water pesticide presence from pulsed exposures during a fixed timeframe, to detect hydrophilic compounds easily missed in grab sampling, and to capture compound mixtures to identify diffuse contaminant sources (Alvarez, 2010; Metcalfe et al., 2019). Since seasonal and annual monitoring across the Coast Range is time consuming and limited by funding constraints, modeling existing monitoring data can extrapolate measured concentrations to unsampled areas. Modeling results, though simplified representations, can predict exposure at multiple scales and guide future monitoring efforts addressing exposure from cumulative or mixed effects.

A previous phase of this project explored herbicide runoff during the spring spray season (six week deployment) to understand differing exposure of bivalves to current-use forestry pesticides based on management regime (Scully-Engelmeyer et al., 2021). Using integrative passive water sampling, we detected four current-use herbicides downstream from actively managed catchments, which, along with biomonitoring efforts, allowed us to examine bivalve exposure in Oregon coastal watersheds (Scully-Engelmeyer et al., 2021). We explored watershed variables related to management and physical characteristics to explain variation in herbicide detections in passive water samples and found that slope and active notifications for aerial herbicide application during the deployment window were the two best individual predictors of total herbicide accumulation in passive water samplers. In this study our goal was to demonstrate an application of publicly available management data to explore the scale effects of management intensity in watersheds on predicted herbicide exposure in downstream waterways. Here we develop a multiple linear regression model to explain relative pesticide concentrations and: (1) identify whether management variables can be used in combination with watershed variables to explain the variation in detected concentrations, (2) assess spatial variability in modeled predictions of the relative presence of herbicides in un-sampled coastal watersheds, and (3) identify the scale effects and regional patterns in measuring predicted concentrations. Additionally, we examine detected herbicides in the context of other protected and valuable aquatic resources in the study location. We expect that variables related to herbicide use and watershed slope in upstream forestlands will best predict downstream concentrations detected in POCIS sampling, and that regional differences in measured pesticide concentrations will be reflected in predicted concentration values.

2. Methods

2.1. Study location

The largely forested Coast Range region of Oregon encompasses the majority of Oregon's coastal watersheds (Spies et al., 2002). The defining feature is the Coast Range Mountains, which separate the coastal watersheds from the inland portion of the state, both topographically and climatically (Franklin and Dyrness, 1973). Unlike other regions in Oregon, drainage basins in the Coast Range (aside from some sections of the Umpqua) are dominated by forestland from headwater to mouth (Spies et al., 2002). This unique geographic scenario provides a valuable and unique opportunity to explore how forestland management practices affect watershed health at multiple scales, without excessive confounding factors from widespread interspersed agricultural or urban

Table 1

Herbicides commonly applied during spring months in forestlands during vegetation management applications (site preparation and pre-emergent (Peachy, 2020)).

Herbicide Compound Name	Common Product Names	Target vegetation	Application rate (active ingredient per acre)
2-ethylhexyl ester of 2,4-Dichlorophenoxyacetic acid (2,4-D)	Weedone LV-4, Weedone LV-6	Broadleaf weeds and woody plants	1–2 lb
Atrazine	Aatrex 4L, Atrazine 4L, Atrazine 90	Grasses and herbaceous plants	3–4 lb.
Clopyralid	Transline	Herbaceous plants	0.19–0.49 lb.
Glyphosate	Rodeo, Roundup	Grasses and broadleaf weeds	1.5–3 lb.
Hexazinone	Velpar L, Velpar DF	Herbaceous and woody plants	1–3 lb.
Indaziflam	Esplanade F	Broadleaf weeds and grasses	0.73–1.46 oz. (not–exceed 10 oz./a of product annually)
Sulfometuron-methyl	Oust, Oust XP	Grasses and broadleaf weeds	1.5–3 lb. 0.375–0.94 oz.
Triclopyr	Garlon 4 Ultra	Woody plants	<6 lb. ae (triclopyr) = 6 quarts

land uses.

2.2. Passive water sampling

Sixteen catchments associated with four main watershed areas were selected for passive water sampling to encompass a range of active forestland management across multiple scales and different latitudes in the Coast Range (Fig. 1). Integrative passive water sampling was utilized to capture episodic chemical exposure in selected catchment areas. Polar organic chemical integrative samplers (POCIS) were deployed (three replicate disks per sample) for six weeks beginning March 26–29, 2019; samplers were retrieved in identical deployment order. POCIS samplers use two microporous membranes (0.1 μm pore) to continually capture water soluble organic compounds from the water column in a solid phase extraction resin (Oasis HLB sorbent) during their deployment period. Upon retrieval, POCIS disks were sent to Environmental Sampling Technologies (EST; Missouri) for extraction. Composited ampules (three disks per ampule) were then sent to Anatek labs (Idaho) for pesticide analysis of commonly used forestry compounds (Supplementary Material (SM; Table S1). Field replicates were deployed at three randomly selected locations to assess method variance, and field and laboratory blanks were implemented to assess unintended contamination during field work and processing. Deployment, retrieval, and quality control measures were implemented in accordance with the guiding document on POCIS monitoring developed by the United States Geological Service (USGS) (Alvarez, 2010). Detailed processing and extraction information can be found in Scully-Engelmeyer et al. (2021).

During the POCIS deployment period, a severe spring storm blanketed south coast watersheds, raising river levels and causing flooding and landslides (FEMA 4452-DR-OR). Upon receding, POCIS canisters at two sites (west fork Millicoma River: MA.1, and north fork Smith River: SH.1) were partially stranded on the bank where they had been deposited while river levels were elevated. Oasis HLB media were still intact in those canisters, so they were processed and included in the results. The submerged sampling interval for those canisters cannot be determined, so concentrations may under-represent exposure over the 45 day sampling period. Additionally, the membranes and HLB media in the Euchre Creek canister (Siletz River: SZ.2) were destroyed during the deployment period, restricting analysis of sampling results at that site.

2.3. Model development

2.3.1. Catchment characterization

Catchment areas above sampling locations were delineated using USGS's online StreamStats application: Streamflow Statistics and Spatial Analysis Tools for Water-Resources Applications (Version 4). Delineations calculated basin characteristics within catchment areas using continuous parameter grids based on 30 m Digital Elevation Models (DEM) (Cooper, 2005; Risley et al., 2008). Variables such as annual precipitation, slope, and elevation were calculated in this way (Table 2).
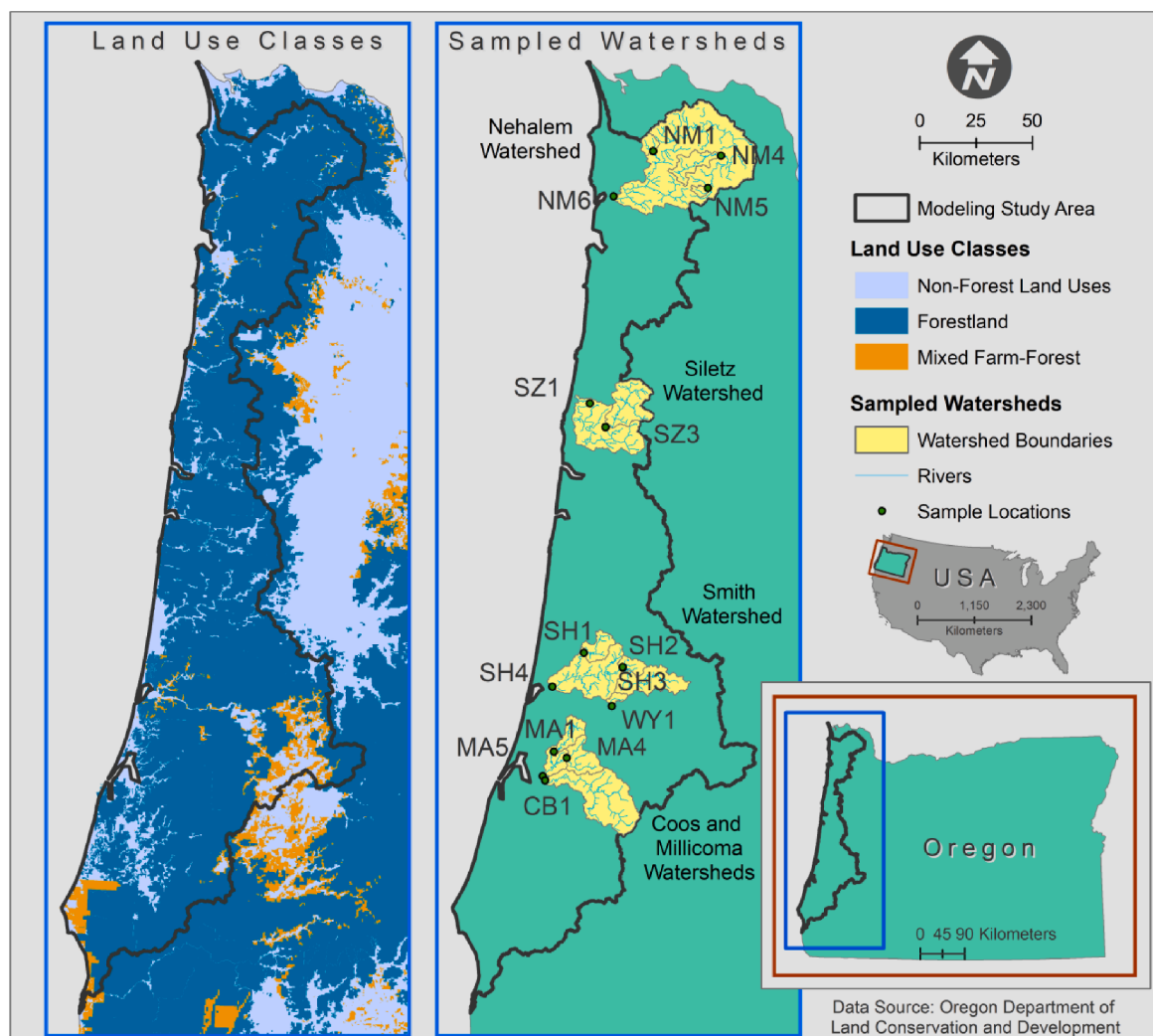

Fig. 1. Simplified land use zoning classes and watershed areas sampled using integrative passive water samplers. Outlined area shows modeling study area.

Table 2

Watershed characteristics - including physical variables calculated above each sampling location and management variables at each location - used in regression analyses. dv = dimensionless variable, km2 = square kilometer.

Watershed Characteristics	Abbreviation	Unit
Physical Variables		
Area	area	Km^2
Steep slopes (slope above 40 %)	slp_abv	%
Road density	rd_den	dv
Drainage density (Σ stream length / watershed area)	drn_den	dv
Forest loss	floss	%
Stream temperature change (between deployment and retrieval)	avtemp_c	Celsius
Average annual precipitation	precip_cm	centimeters
Management Variables		
Area notified for clearcut within 1 year of deployment	cc1yr	%
Area notified for clearcut within 3 years of deployment	cc3yr	%
Area notified for herbicide application during deployment	allherb_dep	%
Area notified for aerial herbicide application during deployment	aerial_dep	%
Area notified for herbicide application within 1 year of deployment	allherb_1yr	%
Area notified for aerial herbicide application within 1 year of deployment	Aerial_1yr	%

Additionally, drainage density and length of roads were automatically calculated during delineation (Cooper, 2005; Risley et al., 2008). Arc-Map version 10.7 was used to determine and export additional characteristics above sampling locations based on catchment delineations from StreamStats. Forest loss data ((Hansen et al., 2013) version 1.7) was imported to ArcMap and converted to polygons. Forest loss from 2016 to 2019 was selected, clipped within study watersheds, and exported. Oregon Department of Forestry hazard slope shapefiles indicating slope above 40 % were used to develop a steep slope variable (Table 2).

Notifications regarding management activities taking place on state, private, and tribal lands are recorded and publicly available through the Forest Electronic Reporting and Notification System (FERNS), and activities on federal lands are accessible through the U.S. Forest Service Activity Tracking System (FACTS) database and a separate online record system for U.S. Bureau of Land Management (BLM) lands. Notification data available in the FERNS dataset outlines types and date ranges of planned management activities, implementation methods, and potential chemicals proposed for use (in the case of pesticide application notifications). Additionally, polygon and line shapefiles, available from the Oregon Department of Forestry's spatial data library, contain notification identification numbers matching pesticide application notifications available from FERNS. The exact date and precise chemical mixtures used in the final activity are not included in this notification data. FERNS notification data were sorted and filtered in excel to encompass the desired timeframes and activity types, then categorized into watershed variables for analysis. Sorted data were imported into ArcMap and joined with FERNS polygons based on identification number; only matching records were retained. Polygons were then re-selected based on desired activity type to exclude irrelevant activities that were inadvertently retained under the same NOAP id number during the first step. Remaining polygons were aggregated (using the Dissolve tool) and clipped to watershed boundaries; the Identity tool was used to compute the variables within study watersheds. Final polygons for each variable were catalogued, exported, and prepared for regression analysis.

2.3.2. Best fit model development

Multiple linear regression analysis was chosen as it offers a simplified and clearly interpretable estimation of variable/response relationships. Independent variables (Table 2) were scaled, and the dependent variable was square root transformed to meet regression assumptions.

Correlation matrices were used to investigate relative correlation between total accumulation in water samples and environmental variables as well as multicollinearity of environmental variables. Additive relationships were explored using manual forward selection stepwise multiple linear regression until coefficient of determination explained close to 90 % of the variation. Since scale is one of the primary output explorations, it was critical to rule out watershed size as a predictor in developing the model. The final model assumptions of normality and multicollinearity were tested using a Shapiro test of residuals and variance inflation factors (VIFs). Remaining model assumptions of skewness, kurtosis, and heteroscedasticity were tested using the Global Validation of Linear Models Assumptions (GVLMA) package. Model validation was done using the leave-one-out cross validation method (LOOCV). LOOCV, a configuration of k-fold cross-validation wherein models are developed for each data point in the input dataset, was chosen for its utility in working with small datasets.

2.4. Model application

Based on the best fit model, independent explanatory variables were calculated and projected across the entirety of the Coast Range province. Hydrologic Unit Code (HUC) catchments at 8, 10, and 12 digit scales from the Watershed Boundaries Dataset (WBD) were then overlaid above Coast Range watersheds, defining the study area. Within the 10 and 12 digit scales, HUC unit boundaries used in model analysis were restricted to catchments containing a complete drainage area to avoid misapplication of model output on HUC units representing partial watershed context (Omernik et al., 2017). This method was applied to avoid misrepresentation of downstream HUC segments as complete watersheds when they are more accurately defined as partial catchment units. HUCs modeled using this selection method represent complete catchments at small (HUC 12), medium (HUC 10), and large (HUC 8) scales within the Coast Range. Ratios of each predictor variable were calculated separately within each HUC across the three scales and exported to excel. Variable values for each catchment were then used to calculate the predicted concentration within each HUC unit based on the best fit model formula.

2.5. Model output analysis

2.5.1. Comparing model output across scales

Final model variables were calculated within each HUC scale across the study area, exported to excel, and imported to Rstudio (version 4.0.4) to calculate predicted values. Predicted values within each catchment across the three scales were displayed in choropleth format across the study area to visually explore patterns of predicted exposure at the three scales investigated. Non-parametric Kruskal-Wallis one-way analysis of variance was used to compare the predicted values at the HUC 8, 10, and 12 digit scales, and relative distribution was explored via density plots.

2.5.2. Exploring regional differences in variables and model outputs

Ratio values of each predictor variable, calculated within each watershed scale, were displayed in a series of choropleth maps of the area to explore regional differences among predictor variables across scales. Density plots were used to compare relative distributions of each predictor variable among scales. Predicted values projected within HUC boundaries across the coast range were displayed via choropleth mapping to visually explore regional differences in predicted exposure. HUC 12 catchments were then grouped into HUC 8 categories to explore how predicted values at the small catchment scale match up within larger drainages/subbasins across the study area. Kruskal-Wallis analysis of variance was used to compare predicted values in the smaller catchments (HUC 12 subwatersheds) across the HUC 8 subbasins (as the grouping variable).

3. Results

3.1. POCIS deployment and detections

Of the fourteen herbicides and one surfactant included in POCIS canister analyses, four commonly applied herbicides were detected (hexazinone, atrazine, sulfometuron methyl, and metsulfuron methyl). Herbicides were detected at 80 % of sample locations (Table 3). Detections ranged from 1.16 to 936 ng/POCIS, averaged 277 ng/POCIS, and varied across locations (Table 3). Concentrations were not detected in field or laboratory blanks.

3.2. Model development

Correlation matrices and Pearson's correlation suggest strong relationships between total detected herbicide concentrations in POCIS samplers and upstream watershed variables, as well as collinearity among variables (Appendix 1A & B). Manual additive multiple regression analysis determined that a model with three independent variables best predicted total herbicide accumulation in passive water samplers without violating multicollinearity assumptions. A multiple linear regression was determined to predict total herbicide accumulation based on watershed characteristics including within the last year (cc1yr); (F (3, 8) = 31.1, p < 0.000), with an R^2 of 0.8914. POCIS predicted concentration = 15.016 + (3.854 *slp_abv) + (5.212* allherb_dep) + (4.855 *CC1yr), where all variables are measured as percentages of upstream catchment areas and were significant predictors of total concentration. Variable inflation factors (VIF) for final variables were 1.460, 2.001, and 1.463 for slp_abv, allherb_dep, and CC1yr respectively, indicating no multicollinearity issues with covariates. Cross validation using LOOCV resulted in a model root mean squared error of 4.567 ng/POCIS, a mean absolute error of 3.783 ng/POCIS and an R^2 of 0.8358.

Overall, variables within HUC 12 watersheds displayed the largest ranges across all categories, followed by HUC10 and HUC8 scales (Fig. 2A, B & C, Table 4). Though ranges varied widely between scales, no significant differences were seen among HUC group means for any of the predictor variables based on Kruskal—Wallis tests (Table 4).

3.3. Model predicted concentration values

Predicted concentrations based on the best fit multiple regression model produced values ranging from 0.1 to 2445.1, and averaged 299.6 ng/POCIS across all categories (Table 4). Similar to predictor variables,

Table 3

Herbicides detected in POCIS samples. Sample locations are organized from north to south along the coast. SMM = sulfometuron methyl, MSM = metsulfuron methyl, RL = reporting limit. RL = 1 ng/POCIS for each compound shown.

Sampling Location	ng/POCIS				
	Atrazine	Hexazinone	SMM	MSM	Total Accumulation
NM.1	11.93	<RL	1.8	<RL	13.7
NM.4	6.05	1.09	<RL	<RL	7.1
NM.5	<RL	<RL	<RL	<RL	<RL
NM.6	<RL	<RL	<RL	<RL	<RL
SZ.1	<RL	38	<RL	<RL	38
SZ.3	<RL	14	<RL	<RL	14
SH.1	<RL	11.6	1.55	<RL	13.2
SH.2	131	816	36.3	1.4	984.7
SH.3	139	212	1.92	<RL	352.9
SH.4	164	103	2.78	<RL	269.8
WY.1	466	963	1.16	<RL	1430.2
MA.1	<RL	<RL	<RL	<RL	<RL
MA.4	185	117	<RL	<RL	302
MA.5	253.3	117.3	<RL	<RL	370.6
CB.1	232	138	<RL	<RL	370

the largest ranges were seen in HUC12 watersheds, followed by HUC10 and HUC8. No significant differences were observed between watershed scales (Table 4, Fig. 3B). Predicted values varied geographically, with the highest values seen in the southern portion of the study area across all three scales (Fig. 3A). Comparisons of HUC 12 predicted values grouped by HUC 8 catchment indicate regional differences in predicted concentrations, wherein predicted values in the Coos watershed were significantly higher than the group mean, and those within Wilson-Trask-Nestucca were significantly lower (Fig. 4). The highest overall predicted values were seen within sub-watersheds of the Umpqua watershed.

4. Discussion

4.1. Passive water samples and independent variable correlation

Concentrations of four commonly applied current use forestry herbicides detected in passive water samples during the spring of 2019 ranged across watersheds and at least one compound detected above reporting limits in 80 % of the samples (12/15 of samples; Table 3). Correlation matrices indicated many correlative relationships between total accumulation in samplers and independent watershed characteristics, as well as among watershed variables. In many instances catchment size is an important predictor in aqueous pesticide concentrations (Schulz, 2004), but in this case watershed size was not correlated with total accumulation in POCIS canisters, signifying that an exploration into factors across multiple scales would be appropriate for these data (Appendix 1A). Another explanatory variable that did not correlate with accumulation was road density, which is important to note as roadside spray activities are considered a potentially confounding source of herbicide runoff in watersheds (Huang et al., 2004; Massoudieh et al., 2005) (Appendix 1A).

4.2. Final explanatory variables

Multiple linear regression revealed that watershed variables steep slopes and notified herbicide and clearcut activity best predicted herbicide accumulation in passive water samplers. LOOCV analysis determined a mean absolute error of 3.783 ng/POCIS, suggesting a relatively low magnitude or error in the predictive capacity of the final model. Watershed slope is an important factor in determining runoff potential within watersheds (Dabrowski et al., 2002; Zhang and Zhang, 2011), so it's significance in predicting pesticide exposure is logical. Additionally, small scale watershed research indicates that steep slopes significantly increase herbicide loss due to runoff (Müller et al., 2004). Herbicide concentration correlated with notified clearcut activity during the previous year, suggesting that site preparation treatments (which occur within the first year post-harvest, before reforestation (Rose and Haase, 2006)) may have contributed to herbicides detected in integrative samplers. Herbicide applications notified during the deployment period was the final predictor in our multiple regression model. Based on the time of year, active notifications during the sampling window (March-May) were likely comprised of pre-emergent (dormant) applications to help established plantations, as well as site preparation treatments.

Final model variables displayed spatial variability (observable in Fig. 2) suggesting regional differences in management (recent clearcuts and herbicide usage) and physical watershed characteristics (slope) within the Coast Range. Steep slopes were most prominent in the north coast watersheds at the HUC 10 and 12 scales near the Kilchis and Wilson rivers (Fig. 2A). Notified herbicide activity was highest in south coast watersheds, especially in tributaries of the Smith, Siuslaw, and Umpqua Rivers (Fig. 2B). Clearcuts notified within the previous year were noticeable throughout the study area, with the highest percentages seen in the Nehalem watershed in the north coast, Siletz watershed in the mid coast, and near the Coquille and Sixes rivers in the south coast (Fig. 2C). The combined additive effects of these variables across the
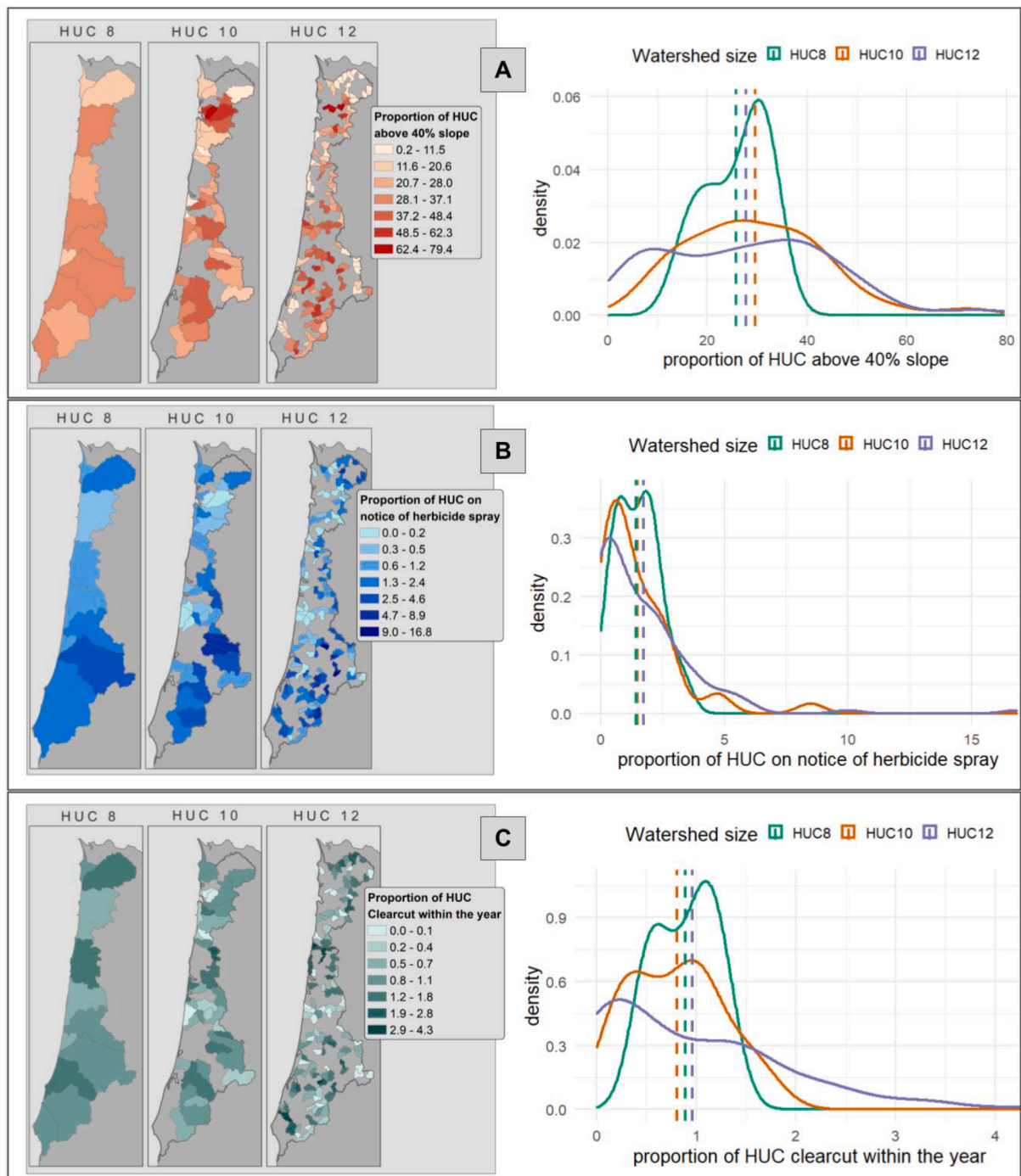
Fig. 2. Percentage of each catchment with steep slopes (A), herbicide notifications during deployment window (B), and clearcuts within a year of deployment (C) were calculated across three HUC scales within the study area. Density distributions reflect un-transformed variables.

landscape served as indicators of predicted herbicide concentration based on the measured sampling window. Across the three scales, the widest ranges of variables were observed within the HUC 12 watersheds followed by the 10 and 8 scales. This is not surprising since smaller catchments are more prone to dominance by single land use types/features, which can translate to higher and lower values of these variables. At larger scales, the complexity of the landscape has a dampening effect on the range of individual variables, as they are averaged across the entire watershed. Across scales, mean values for each variable were not significantly different (Table 4).

4.3. Model outputs/predicted concentration values

Similar to individual independent variables, predicted concentration values based on regression model output displayed regional differences in high values. Catchments in the Umpqua, Coos, and Smith river watersheds displayed the highest values at the HUC 12 scale, followed by tributaries within the Alsea and Sixes river watersheds (Fig. 4). At the HUC 10 scale, the upper Smith River had the highest predicted value followed by a number of other headwater catchments in the central and south coast. HUC 8 predicted herbicide concentrations were highest in south coast watersheds. Data structure of predicted concentrations was similar to predictor variables, wherein HUC 12 catchments displayed the

Table 4
Summary statistics for final predictor variables [steep slopes above 40 percent (slp_abv), area notified for herbicide application during deployment (allherb_dep) and area notified for clearcut within 1 year of deployment (cc1yr)] and predicted values in each HUC level.

Watershed size	Predictor variables: $\bar{x}$ (range)			Model predicted values (ng/POCIS)
	slp_abv (%)	allherb_dep (%)	cc1yr (%)	
HUC8	25.7 (15.5–33.0)	1.5 (0.4–3.2)	0.89 (0.5–1.3)	294.6 (99.5–516.8)
HUC10	29.6 (5.6–72.2)	1.5 (0–8.5)	0.8 (0.1–1.8)	289.0 (17.3–1301.8)
HUC12	27.8 (0.2–79.4)	1.7 (0–16.8)	0.96 (0–4.28)	303.5 (0.1–2445.1)
Overall	28.1 (0.2–79.4)	1.65 (0–16.8)	0.9 (0–4.28)	299.6 (0.1–2445.1)
Kruskal—Wallis	H(2) = 0.704, p = 0.7033	H(2) = 0.315, p = 0.8538	H(2) = 0.316, p = 0.8542	H(2) = 2.1409, p = 0.3428

largest ranges of values, followed by HUC 10 and 8 scales (Fig. 3). Despite differences in range, differences among scales were not significant (Table 4), which is not surprising given the nested nature of the HUC watersheds in the study area. Predicted concentrations calculated across scales based on watershed slope, herbicide activity, and notified clearcuts highlights the importance of looking at potential impacts to aquatic ecosystems from a landscape pattern perspective, beyond the site level.

Subwatersheds (HUC 12) grouped by subbasin (HUC 8 scale) allowed for quantification of regional differences in predicted values (Fig. 4). In our analysis, South coast watersheds had higher average predicted concentrations than mid or north coast watersheds, but Coos was the only HUC8 group significantly higher than the base mean, and the Wilson-Trask-Nestucca was the only watershed group with significantly lower predicted concentrations (Fig. 4). Regional patterns from this analysis are similar to field-collected data, wherein south coast locations exhibited higher on average concentrations compared with mid and north-coast counterparts. These observations may represent the amount of active management taking place in southern watersheds or could be an artifact of spray timing/management differences between the areas.

4.4. Other aquatic resources across scales

Considerations of the spatial configuration of landscape variables (land use, management, environmental characteristics) are critical in understanding anthropogenic activities threatening watershed water quality, ecological processes, and aquatic resources (Lee et al., 2009). Within the context of the Oregon Coast Range, watershed scale aquatic resources exist at multiple points along stream networks, and are therefore influenced by upstream conditions at multiple scales. Interpreting potential impacts to these resources at the scales in which they are found is challenging, especially given the wide range of ownership, management, and physical watershed characteristics in upstream drainages. Study results suggest that the potential for both higher and lower herbicide exposure is greater at smaller watershed scales, but overall watershed size does not impact the average exposure among the three scales investigated. Our investigations provide predicted concentrations at established HUC scales, but on the, resources exist independent of established scale boundaries such as the HUC system. Fig. 5 offers a subset of Oregon Coast Range aquatic resources, such as drinking water sources (surface and groundwater), salmonid runs, and aquaculture areas within watersheds, which are influenced by catchments of various sizes. Drinking water originating from surface water is a good example of a resource that, though permitted and collected at a specific point, is influenced (and potentially threatened) by upstream catchment characteristics such as land uses and practices (Lari et al., 2014). As indicated in Fig. 5, herbicide detections at sampling locations varied along the coast, with the highest values seen at the south coast sites. Furthermore, this figure illustrates the overlapping nature of detection sites and other aquatic resources within the Coast Range.

4.5. Model applicability

This investigation into springtime herbicide exposure across multiple scales in coastal watersheds is one of many potential avenues of inquiry into non-point source pesticide pollution, and like many monitoring and modeling efforts is limited by available data. Our sampling window characterizes one time period, and though results are useful in explaining relationships between upstream variables and observed concentrations, considerable inter-annual variation in management activities throughout the Coast Range introduces uncertainty about the suitability of our model to other timeframes or regions. Inconsistency in management regimes applied to Oregon forestlands based on developments in ownership, guiding regulations/practices, and technology throughout time present a complicated picture of the landscape ecology in coastal watersheds. Harvest rotations for contemporary intensive forest management are generally 30–50 years long, and over the timeframe of one harvest cycle, updates to methodology and regulations can evolve. Our results provide insight into herbicide movement through the water column during a 45-day deployment period, and associated catchment variables that can predict concentrations in this
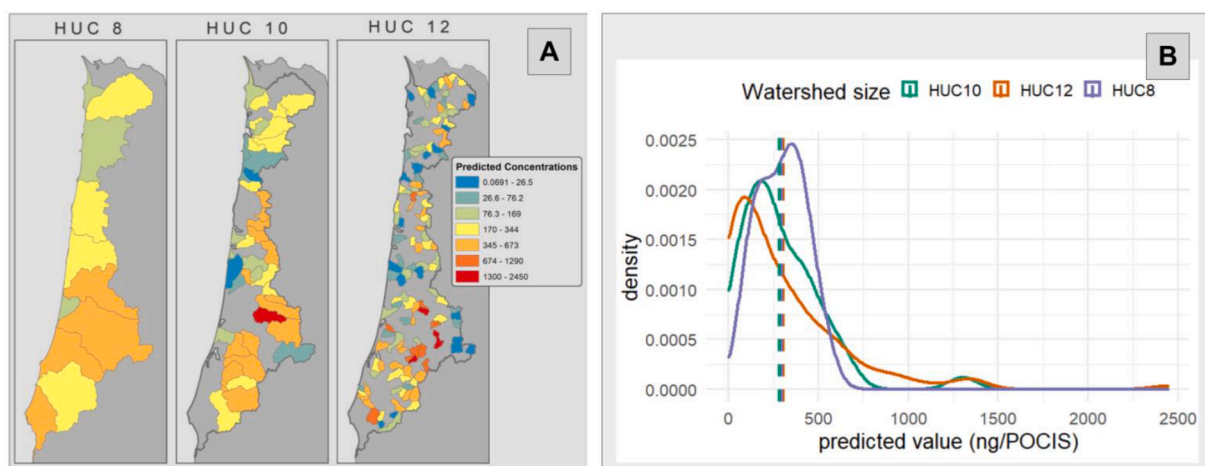

Fig. 3. Model predicted concentrations across HUC 8, 10, and 12 scales in the Coast Range (A), and compared in a density distribution plot (B).
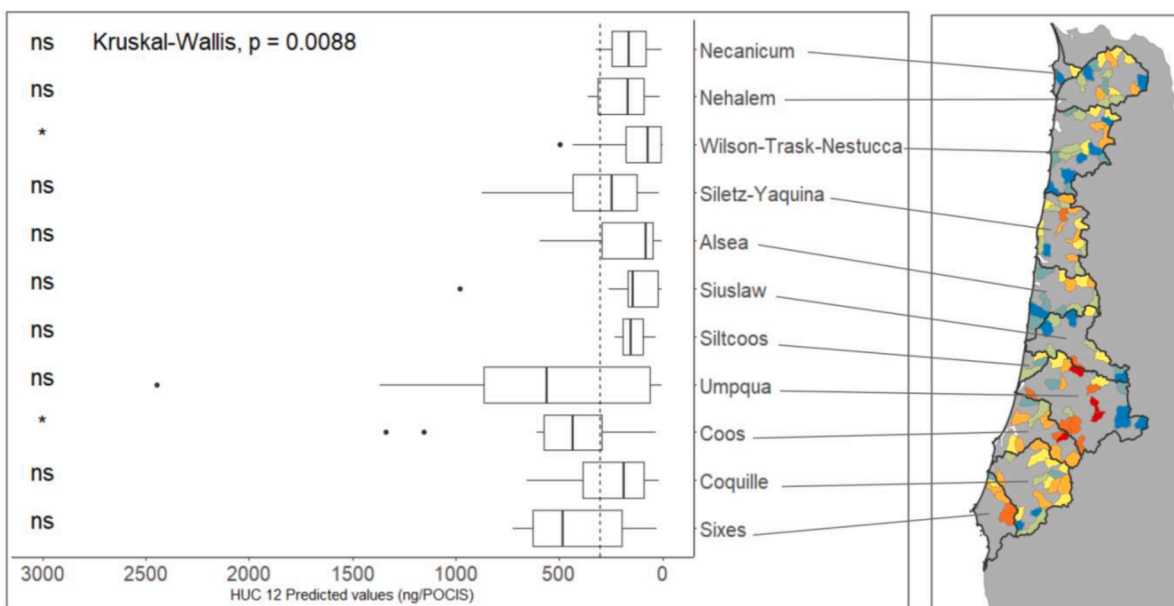
Fig. 4. Predicted concentration values within HUC 12 catchments grouped by HUC 8 watersheds with multiple pairwise tests against the base mean. Abbreviations: ns = not significant, * = p ≤ 0.05.
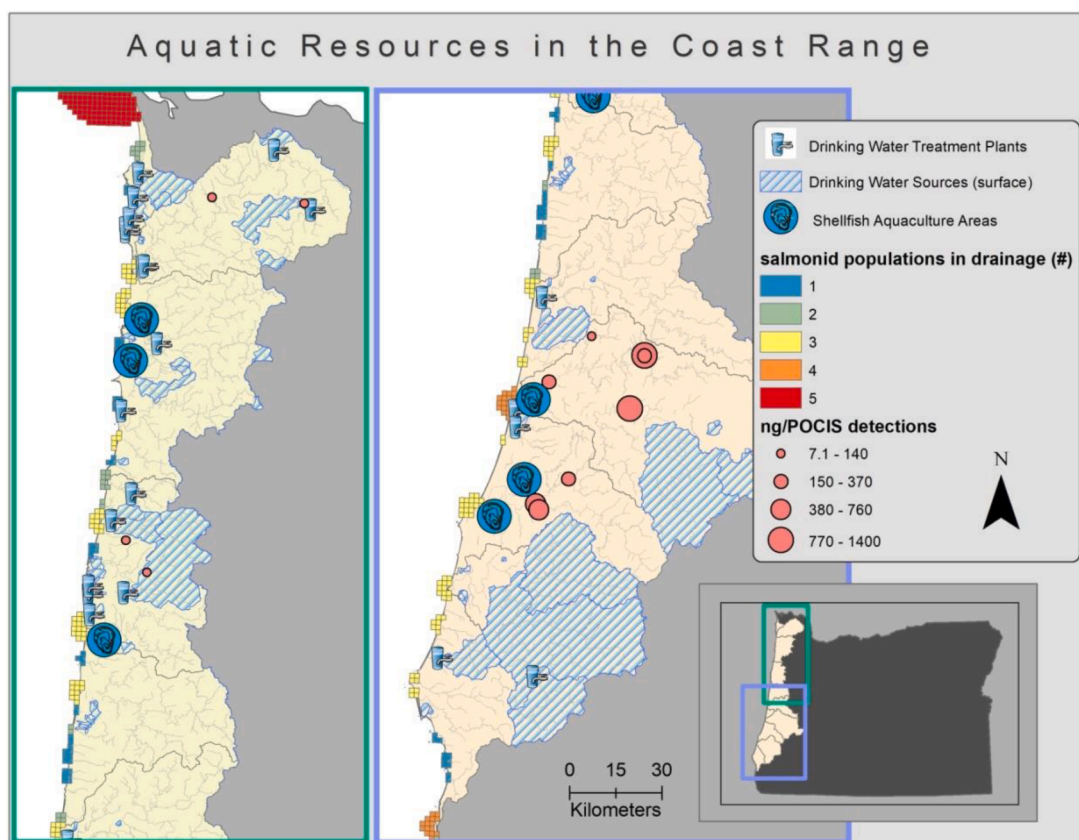

Fig. 5. A subset of aquatic resources in the Coast Range, and the various scales they occupy. Total herbicide accumulation detected in POCIS samplers (ng/POCIS) is overlaid at sampling locations. Data sources: Oregon Department of Fish and Wildlife, Oregon Department of Environmental Quality, Oregon Department of Agriculture.

context, but herbicide movement during other times of year as well as during the same time frame across years may not be well characterized by these data.

Our results suggest fundamental connections between landscape patterns of watershed management/characteristics and downstream pesticide exposure can be predicted based on relatively simple indicators, but the applicability of these indicators (slope, herbicide use, and clearcuts) in different regions remains elusive. For example, our

model may not be useful beyond the southern portion of the Coast Range region (past Cape Blanco to the south), where biogeographical, management and climatic differences in the landscape makeup likely impact the ability of this regional specific model in predicting movement of pesticides in watersheds. Similarly, in eastern portions of the state, federal and state forestry herbicide use regulations diverge from coastal provisions (US Bureau of Land Management, 2010) (OAR 629-642-0400), which coupled with differing in biogeographical features (Franklin and Dyrness, 1973) between regions further constrain model applicability. However, data collection in these areas and other seasons could be utilized to build similar predictive models.

Factors described above may limit the applicability to other regions of the specific model developed in this study. However, the model serves to demonstrate the efficacy and application of publicly available land management data to predict water quality conditions to view cumulative effects of management activities in watersheds. Region-specific models may be developed using similar publicly available data to raise awareness and support policy development to address environmental contaminants. By basing this analysis on simplified but clearly interpretable variable/response relationships provided by multiple linear regression, this research promotes a unique way to engage with publicly available data in illustrating the connection between management intensity and watershed health.

5. Conclusions

In this investigation we found that a physical watershed variable (steep slopes) coupled with notified forestland management activities (herbicide use and clearcut harvest) successfully predicted measured herbicide presence ($R^2 = 0.8914$) during the spring spray period (March to May). These results highlight connections between spatial landscape patterns of environmental factors, anthropogenic land-uses, and offsite herbicide movement in coastal watersheds in Oregon. When applied to unsampled watersheds in the same region, predicted concentrations from our model exhibited similar spatial patterns as measured concentrations, wherein south coast watershed displayed higher on average concentrations compared to mid and north coast watersheds. Across three watershed sizes (scales) we found that the greatest ranges in predicted values were seen in smaller catchments (HUC 12), followed by

medium and large catchments (HUCs 10 & 8), but the average concentrations did not differ among scales. The final model provides insight into patterns of herbicide use and movement in coastal watershed in Oregon, but its application is constrained by the sampling window from which the data were derived, small sample size, and the region-specific context. Furthermore, herbicide detections overlap with important aquatic resources, highlighting the need for further research to determine effects of transported herbicides on these resources. This research demonstrates the importance of approaching interpretation of non-point sources of pollution at appropriate landscape scales and contexts.

CRediT authorship contribution statement

Kaegan M. Scully-Engelmeyer: Conceptualization, Methodology, Investigation, Formal analysis, Visualization, Writing – original draft. **Elise F. Granek:** Conceptualization, Supervision, Funding acquisition, Writing – review & editing.

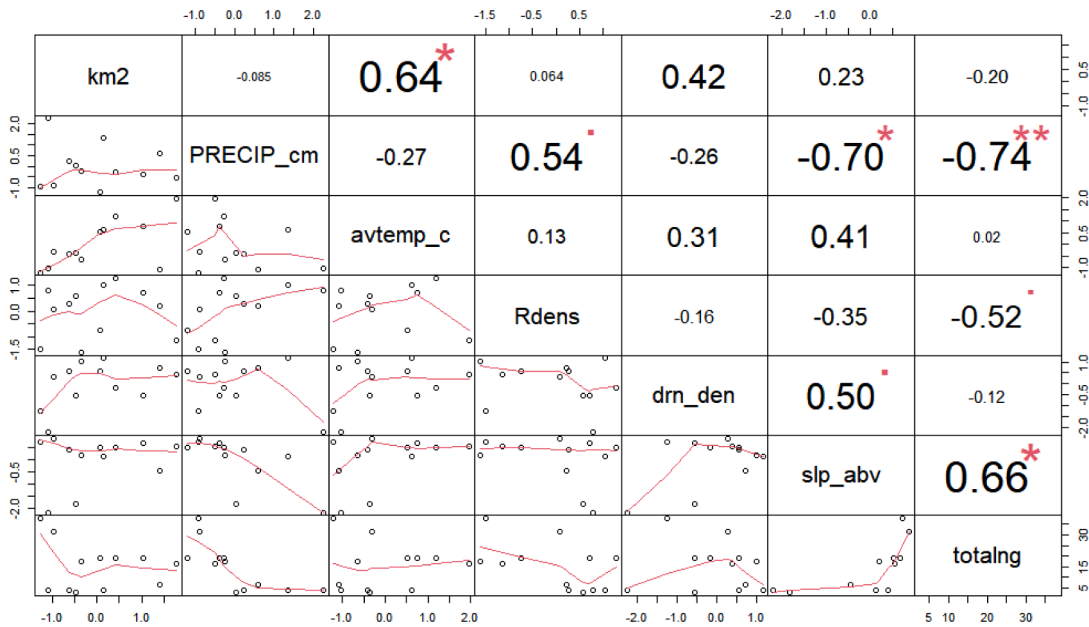## Declaration of Competing Interest

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.
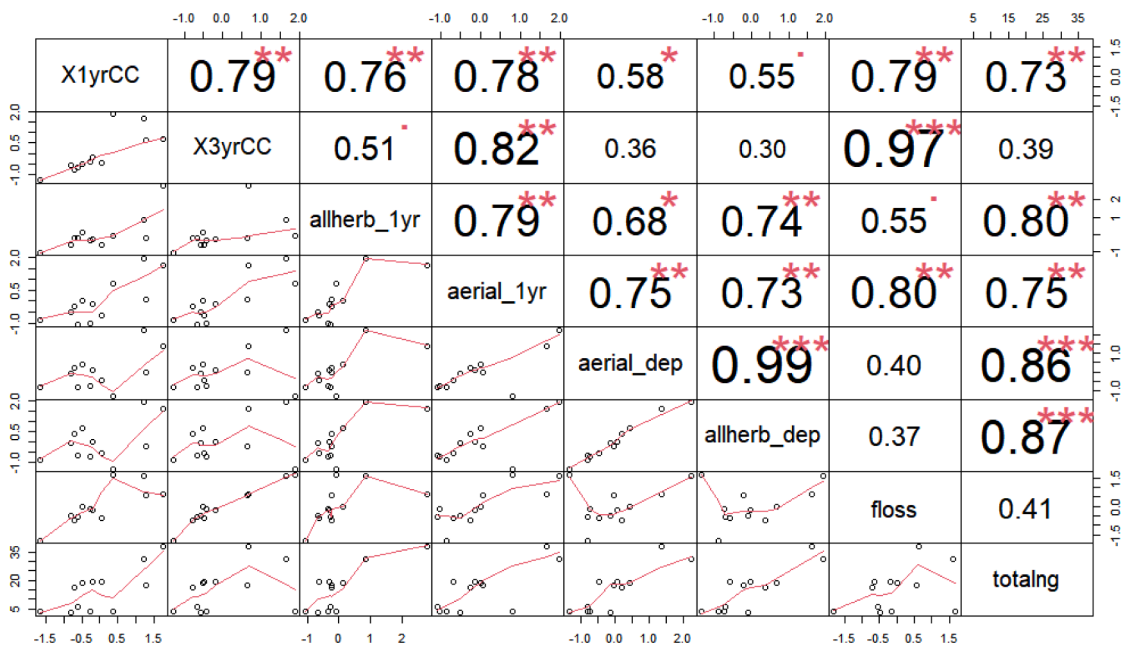
Data availability

Data will be made available on request.

Appendix A

A & B: Correlation matrices of physical (A) and management (B) watershed variables with total herbicide accumulation (totalng). Variable abbreviations are provided in Table 2 of the document.

A

B

Appendix B. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.ecolind.2022.109195.

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