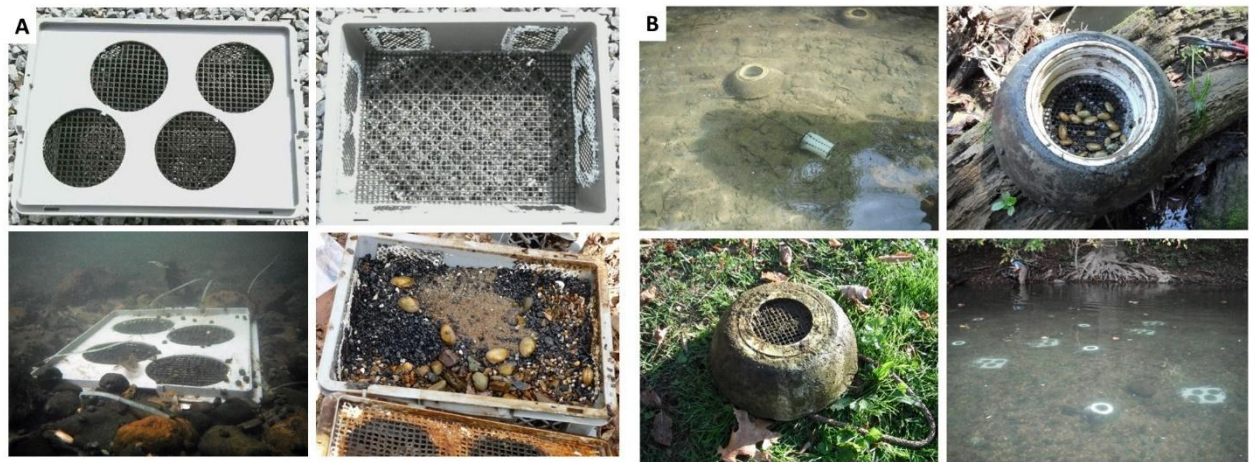
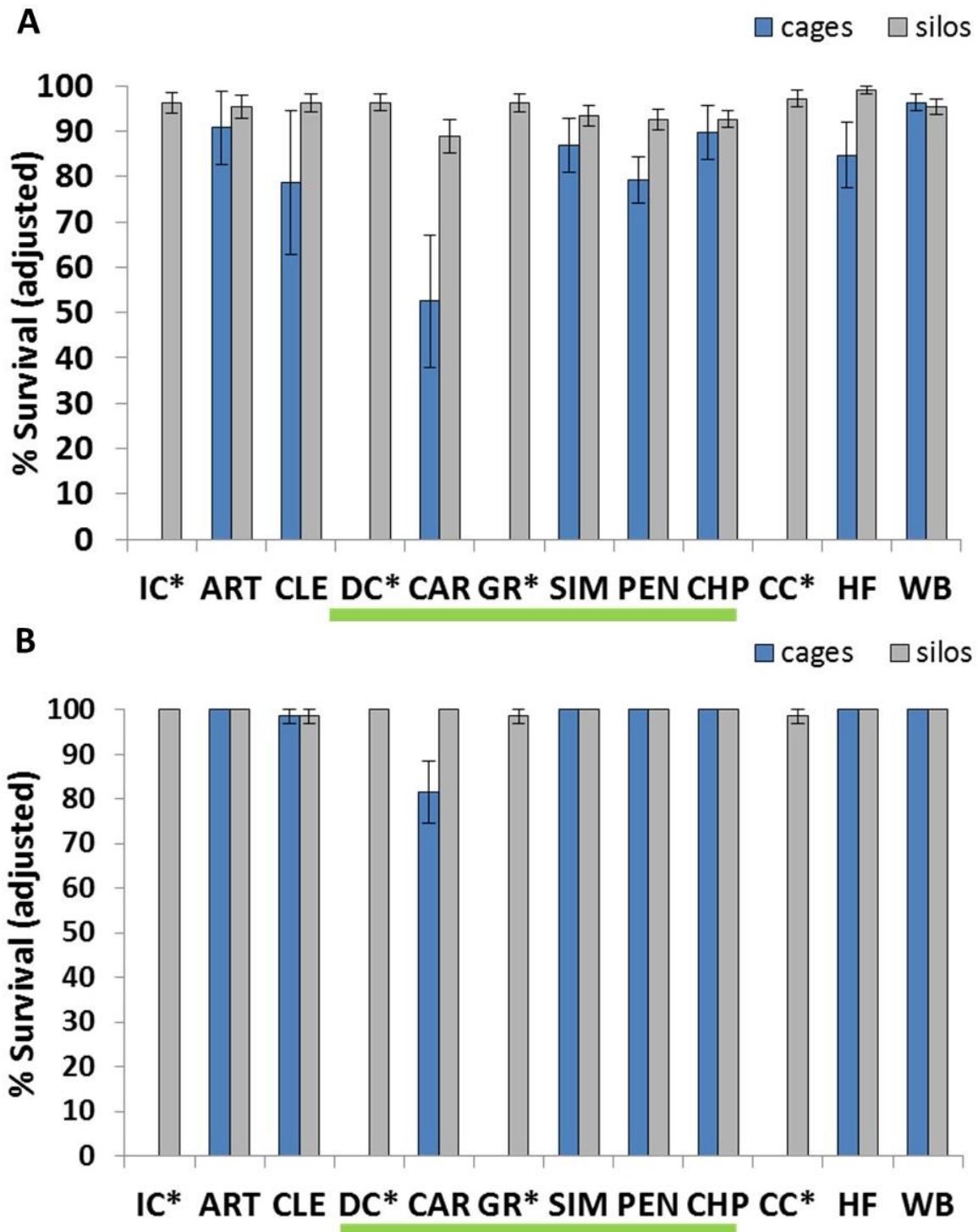


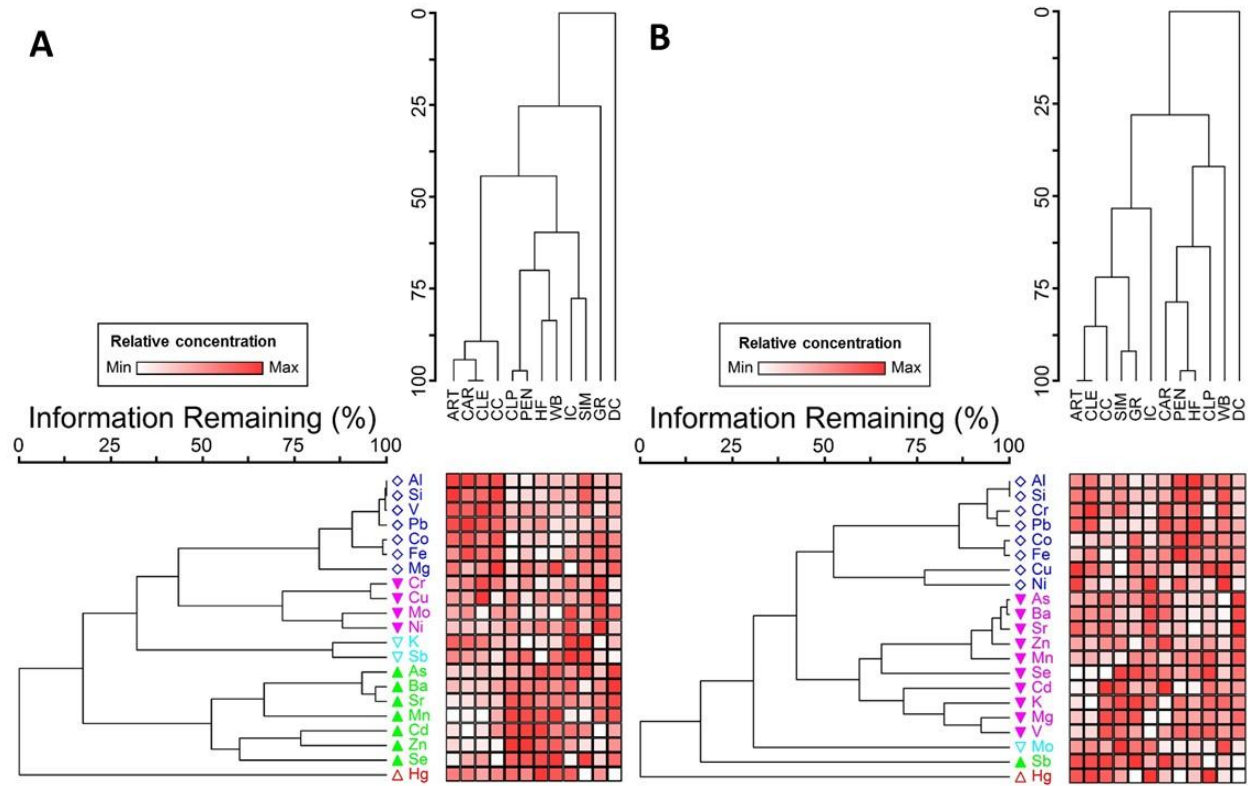
Supplemental Figures and Tables



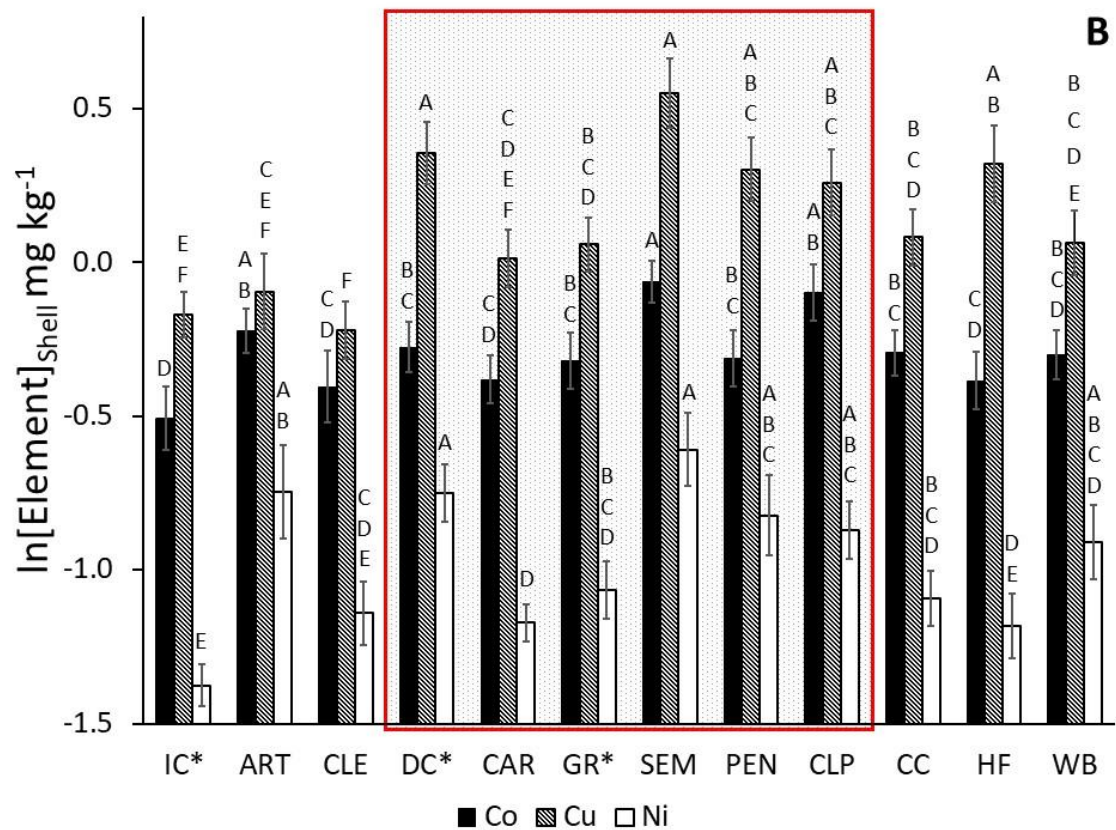
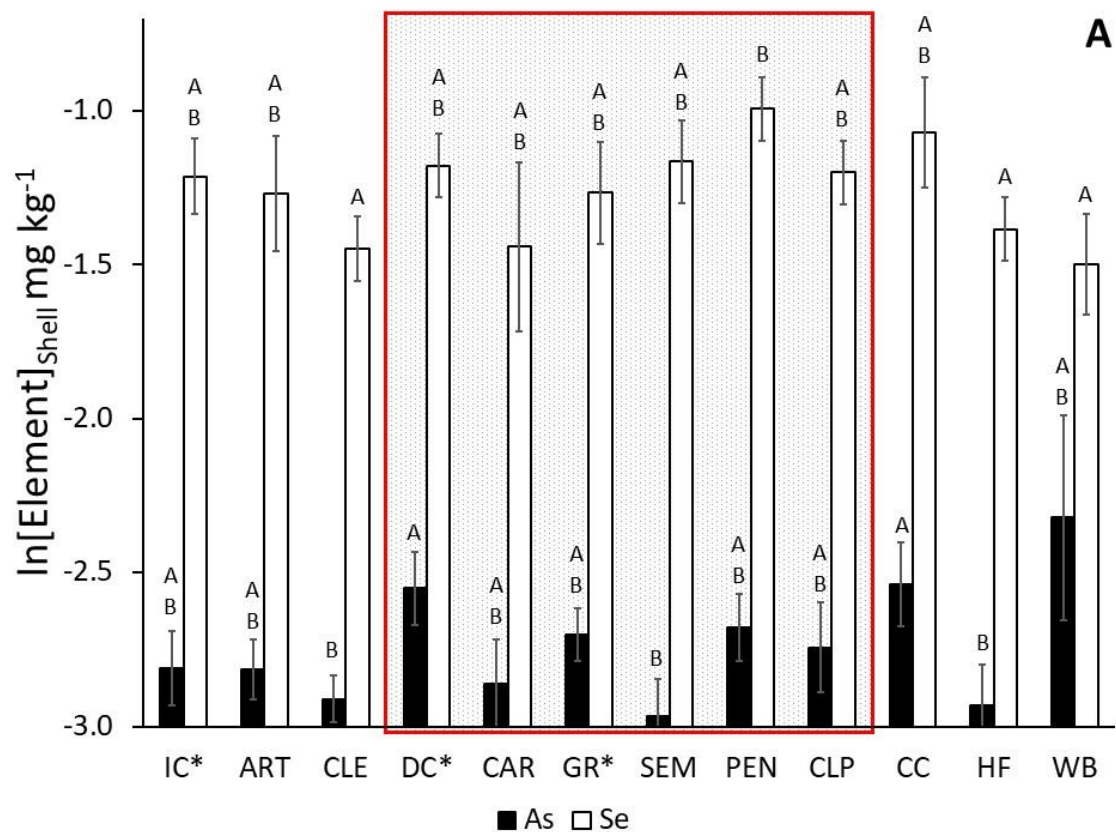
Supplemental Figure S1. Mussel deployment methods. **(A)** Cages were constructed from polyethylene industrial containers with mesh sides and base with plastic mesh (6.34 mm) affixed to the inside of the cages on the top, sides, and bottom to contain mussels and allow water to flow through the cages. Cages were deployed in areas with fine and coarse gravel and river substrate was used to fill the cages as they were partially buried to ensure the tops of the cages were flush with the sediment-water interface. **(B)** Concrete mussel silos were constructed to house mussels in the water column; cylindrical chambers had mesh tops and bottoms to allow water to circulate through the chamber.

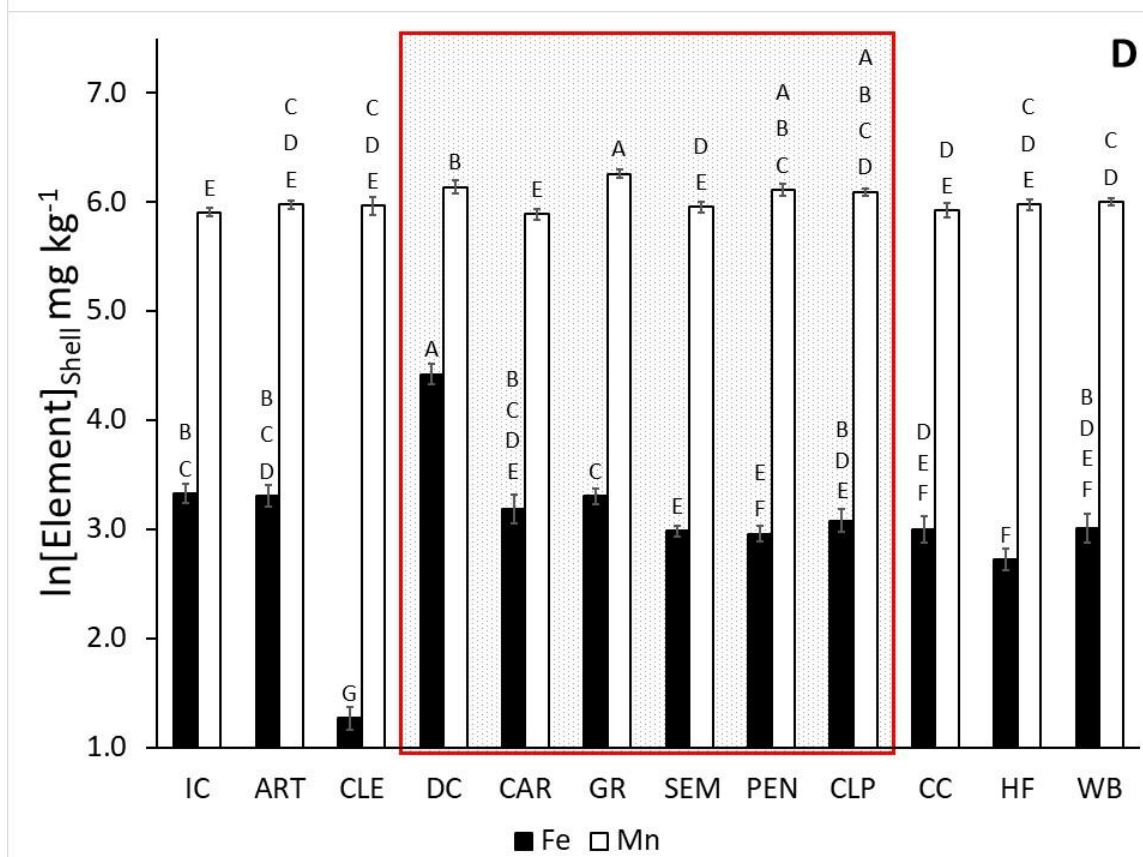
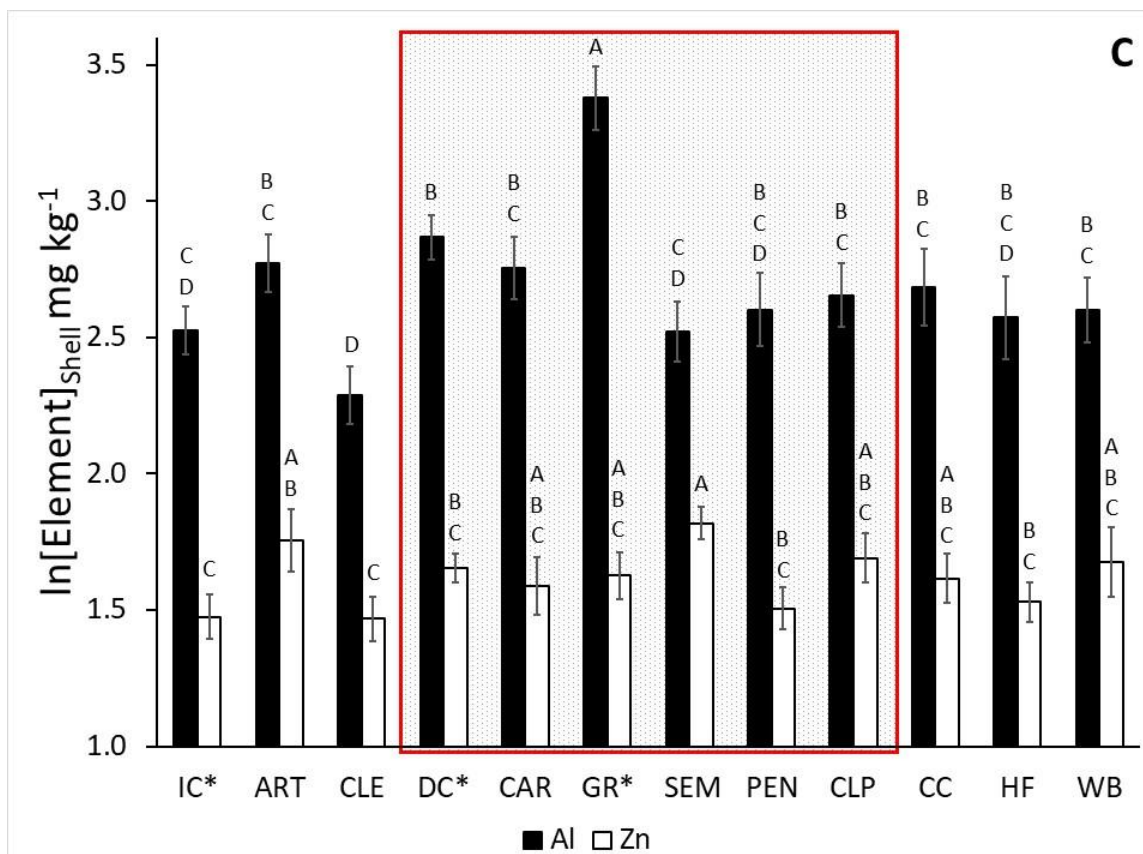


Supplemental Figure S2. Adjusted survival (percent of mussels recovered alive out of the mussels recovered) for (A) *Villosa iris* (2012) and (B) *Lampsilis fasciola* (2013). Tributary sites are denoted by asterisks. Green bar indicates sites within the 'Zone of Decline' (see Figure 1). For sites where survival in each sample was 100%, the standard error (SE) was 0 (no SE bar shown).

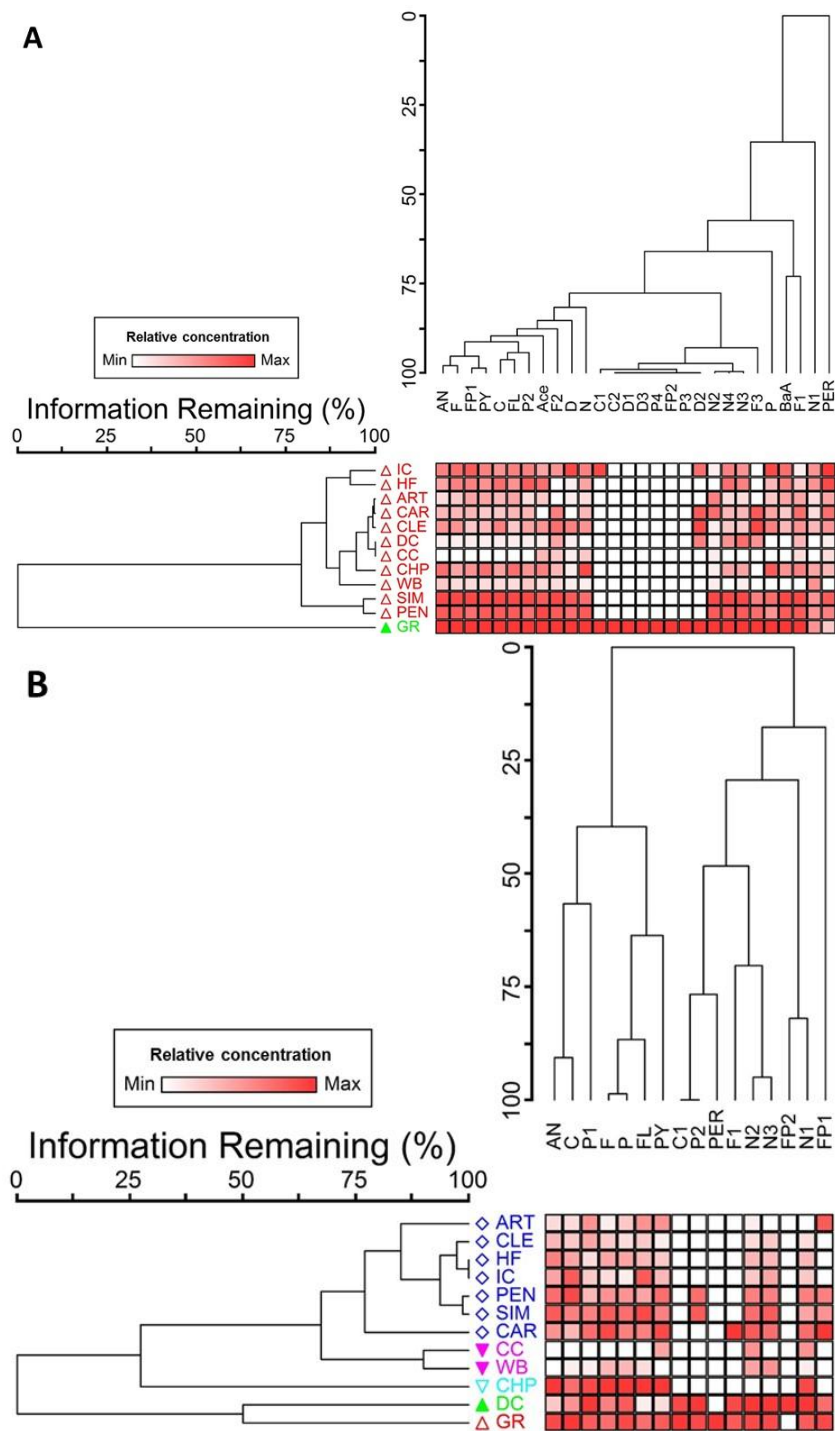


Supplemental Figure S3. Two-way dendrogram from cluster analysis of trace element concentrations from soft tissues of (A) juvenile *Villosa iris* deployed in 2012 and (B) juvenile *Lampsilis fasciola* deployed in 2013. Relative metal concentrations are indexed by the color gradient as darker red cells indicate higher concentrations of a given metal in the tissues at a particular site. The 'Information Remaining' metric is the percent of the total original variation in multivariate distances among all 21 metals that remains after each joining (node) of clusters or sites.





Supplemental Figure S4. Natural log transformed mean concentrations (originally mg kg⁻¹) of trace elements in the shells of *Villosa iris* and *Lampsilis fasciola* deployed in silos at sites within the Clinch River System. **(A)**=As, Se; **(B)**=Co, Cu, Ni; **(C)**=Al, Zn; **(D)**=Fe, Mn. Error bars are standard errors. Means within the same series with the same letters are not significantly different from one another. The shaded area inside the red box represents sites considered to be within the zone of decline.



Supplemental Figure S5. Two-way dendrogram from cluster analysis of PAH concentrations from soft tissues of (A) juvenile *Villosa iris* deployed in 2012 and (B) juvenile *Lampsilis fasciola* deployed in 2013. Relative PAH concentrations are indexed by the color gradient as darker red cells indicate higher concentrations of a given metal in the tissues at a particular site. The ‘Information Remaining’ metric is the percent of the total original variation in multivariate distances among all 21 metals that remains after each joining (node) of clusters or sites.

Supplemental Table S1. Mean trace element concentrations in the soft tissues of deployed mussels, pooled for both species/years of deployment (*Villosa iris* in 2012 and *Lampsilis fasciola* in 2013) and deployment method (silo or cage). Concentrations with the same superscript letter are not significantly ($\alpha = 0.05$) from one another.

	Al	As	Ba	Cd	Co	Cr	Cu	Fe	Hg	K	Mg
IC	393 ^a	7.38 ^a	262 ^a	0.804 ^a	1.02 ^a	2.98 ^a	49.4 ^a	1705 ^a	0.165 ^{abc}	3170 ^a	2021 ^a
ART	997 ^a	7.73 ^a	247 ^a	0.754 ^a	1.41 ^a	4.36 ^a	80.7 ^a	2132 ^a	0.138 ^{abc}	3022 ^a	2381 ^a
CLE	1075 ^a	8.26 ^a	238 ^a	0.821 ^a	1.52 ^a	5.76 ^a	139. ^a	2437 ^a	0.138 ^{abc}	2869 ^a	2391 ^a
DC	960 ^a	13.6 ^a	469 ^a	0.614 ^a	1.68 ^a	3.98 ^a	56.5 ^a	2796 ^a	0.068 ^a	3283 ^a	2797 ^a
CAR	1092 ^a	7.38 ^a	287 ^a	0.539 ^a	1.58 ^a	4.95 ^a	87.1 ^a	2496 ^a	0.139 ^{abc}	3043 ^a	2330 ^a
GR	407 ^a	6.84 ^a	187 ^a	0.541 ^a	1.50 ^a	6.07 ^a	132 ^a	2431 ^a	0.097 ^{ab}	2868 ^a	2320 ^a
SIM	668 ^a	8.19 ^a	208 ^a	0.625 ^a	1.26 ^a	3.59 ^a	57.3 ^a	1823 ^a	0.108 ^{abc}	3207 ^a	2373 ^a
PEN	765 ^a	8.04 ^a	263 ^a	0.914 ^a	1.44 ^a	4.86 ^a	60.9 ^a	2260 ^a	0.123 ^{abc}	3091 ^a	2486 ^a
CLP	518 ^a	7.33 ^a	248 ^a	0.651 ^a	1.16 ^a	2.98 ^a	57.1 ^a	1747 ^a	0.170 ^{bc}	3000 ^a	2323 ^a
CC	1206 ^a	7.57 ^a	247 ^a	0.592 ^a	1.67 ^a	4.55 ^a	53.4 ^a	2247 ^a	0.147 ^{abc}	3024 ^a	2700 ^a
HF	528 ^a	9.12 ^a	216 ^a	0.938 ^a	1.19 ^a	3.15 ^a	40.6 ^a	1598 ^a	0.228 ^c	2817 ^a	2259 ^a
WB	492 ^a	7.35 ^a	334 ^a	0.532 ^a	1.10 ^a	3.48 ^a	44.5 ^a	1673 ^a	0.121 ^{abc}	3073 ^a	2389 ^a

	Mn	Mo	Ni	Pb	Sb	Se	Si	Sr	V	Zn
IC	1782 ^a	0.661 ^{ab}	6.05 ^a	1.64 ^a	0.689 ^a	8.68 ^a	543 ^a	468 ^{ab}	3.57 ^a	318 ^a
ART	1901 ^a	0.433 ^a	5.58 ^a	2.79 ^a	0.551 ^a	9.16 ^a	1037 ^a	468 ^a	5.29 ^a	322 ^a
CLE	1873 ^a	0.511 ^{ab}	3.32 ^a	3.15 ^a	0.533 ^a	11.1 ^a	1064 ^a	471 ^a	5.90 ^a	319 ^a
DC	3073 ^c	0.545 ^{ab}	3.91 ^a	2.45 ^a	0.568 ^a	13.9 ^a	918 ^a	780 ^b	6.31 ^a	372 ^a
CAR	2384 ^{ab}	1.07 ^{ab}	3.88 ^a	3.42 ^a	0.591 ^a	11.5 ^a	979 ^a	431 ^a	6.23 ^a	335 ^a
GR	2201 ^{ab}	0.595 ^{ab}	6.42 ^a	1.82 ^a	0.530 ^a	9.65 ^a	534 ^a	430 ^a	3.91 ^a	304 ^a
SIM	1853 ^a	1.83 ^b	2.82 ^a	1.94 ^a	0.602 ^a	12.3 ^a	727 ^a	452 ^a	4.83 ^a	322 ^a
PEN	2527 ^{bc}	0.499 ^{ab}	6.06 ^a	2.40 ^a	0.558 ^a	12.8 ^a	815 ^a	548 ^{ab}	5.31 ^a	346 ^a
CLP	2467 ^{bc}	0.547 ^{ab}	3.29 ^a	2.04 ^a	0.549 ^a	11.9 ^a	530 ^a	516 ^{ab}	4.60 ^a	352 ^a
CC	2027 ^{ab}	0.597 ^{ab}	2.70 ^a	3.03 ^a	0.549 ^a	7.65 ^a	1222 ^a	495 ^{ab}	6.31 ^a	318 ^a
HF	2271 ^{ab}	0.443 ^{ab}	3.36 ^a	2.19 ^a	0.508 ^a	12.4 ^a	552 ^a	441 ^a	4.35 ^a	328 ^a
WB	2193 ^{ab}	1.78 ^{ab}	8.05 ^a	1.84 ^a	0.508 ^a	11.1 ^a	621 ^a	457 ^a	4.34 ^a	381 ^a

Supplemental Table S2. Spearman's rho correlation coefficients for correlations between site-specific bed and particulate sediment and juvenile mussel trace element soft tissue concentrations. *Villosa iris* were deployed in 2012 and *Lampsilis fasciola* were deployed in 2013. Bold and Italics with * = significant ($p \leq 0.05$); Bold and Italics only = marginal significance ($0.05 < p < 0.10$). BDL = more than half of samples were below detection limit; therefore, the element was excluded from our analyses.

Element	2012 <i>Bed</i>	2013 <i>Bed</i>	2013 <i>Particulate</i>
Al	-0.09	<i>0.53</i>	0.12
As	0.34	0.02	<i>0.74*</i>
Ba	-0.22	-0.12	0.28
Be	BDL	BDL	BDL
Cd	BDL	BDL	BDL
Co	-0.27	0.27	0.43
Cr	-0.22	<i>0.54</i>	0.05
Cu	0.01	0.14	-0.01
Fe	-0.29	-0.01	0.19
Hg	BDL	BDL	BDL
K	-0.27	0.27	0.30
Mg	-0.14	-0.12	-0.22
Mn	0.22	-0.11	0.34
Mo	BDL	BDL	BDL
Ni	<i>-0.52</i>	0.20	<i>-0.58</i>
Pb	-0.04	0.38	0.10
Sb	BDL	BDL	BDL
Se	BDL	BDL	BDL
Si	-0.01	0.47	0.27
Sr	-0.05	-0.31	0.14
V	0.15	0.16	0.11
Zn	0.14	-0.09	-0.22

Supplemental Table S3. Spearman's rho correlation coefficients for correlations between site-specific bed and particulate sediment and juvenile mussel PAH tissue concentrations. *Villosa iris* were deployed in 2012 and *Lampsilis fasciola* were deployed in 2013. Bold and Italics with * = significant ($p \leq 0.05$); Bold and Italics only = marginal significance ($0.05 < p < 0.10$). BDL = more than half of samples were below detection limit; therefore, the compound was excluded from our analyses.

PAH	2012 <i>Bed</i>	2013 <i>Bed</i>	2013 <i>Particulate</i>
AN	0.35	0.41	0.41
Ace	0.10	BDL	BDL
BaA	0.39	BDL	BDL
C	0.24	0.42	0.21
D	0.04	BDL	BDL
F	0.19	<i>0.67*</i>	<i>0.83*</i>
F1	<i>0.57</i>	BDL	BDL
F2	0.42	BDL	BDL
F3	0.35	BDL	BDL
FL	0.38	0.45	0.31
FP1	0.22	<i>0.53</i>	<i>0.63*</i>
N	0.25	BDL	<i>0.75*</i>
N1	0.13	<i>0.69*</i>	BDL
N2	0.39	0.48	0.49
N3	<i>0.65*</i>	0.62	<i>0.56</i>
N4	<i>0.51</i>	BDL	BDL
P	0.11	<i>0.80*</i>	<i>0.88*</i>
P1	BDL	<i>0.74*</i>	<i>0.81*</i>
P2	0.21	BDL	BDL
PER	-0.17	BDL	BDL
PY	0.21	0.45	<i>0.62*</i>
Sum34	0.23	<i>0.72*</i>	<i>0.79*</i>
Sum42	0.23	<i>0.73*</i>	<i>0.79*</i>

Supplemental Table S4. Spearman's rho correlation coefficients for correlations between site-specific surface water concentrations of trace elements (by sampling event) and juvenile mussel soft tissue concentrations. Bold and Italics with * = significant ($p \leq 0.05$); Bold and Italics only = marginal significance ($0.05 < p < 0.10$). BDL = more than half of samples were below detection limit; therefore, the metal was excluded from our analyses. Sampling events were as follows: Q1 = August 21-23, 2012 (mainstem only); Q2 = October 24-25, 2012 (tributaries) and November 15-16, 2012 (mainstem); Q4 = July 18-19, 2013 (mainstem) and August 5-6, 2013 (tributaries); Q5 = September 18-20, 2013 (mainstem only). Juvenile mussels deployed from June 5-6, 2012 to October 24-25, 2012 and July 31-August 1, 2013 to November 5-7, 2013.

Element	<u>2012</u>		<u>2013</u>	
	Q1	Q2	Q4	Q5
Al	0.45	-0.24	0.42	<i>0.72*</i>
Cr	BDL	BDL	-0.41	-0.35
Cu	BDL	BDL	-0.22	BDL
Fe	0.43	BDL	0.06	<i>0.78*</i>
K	0.19	-0.17	0.47	<i>0.79*</i>
Mg	<i>-0.77*</i>	0.16	<i>0.58</i>	<i>0.73*</i>
Mn	0	<i>0.68*</i>	0.14	<i>0.86*</i>
Ni	-0.35	-0.24	-0.09	0.09
Zn	0.32	BDL	0.26	0.14

Supplemental Table S5. Spearman's rho correlation coefficients for correlations between site-specific surface water concentrations of PAHs (by passive diffusion sampler deployment period) and deployed mussel PAH tissue concentrations. Bold and Italics with * = significant ($p \leq 0.05$); Bold and Italics only = marginal significance ($0.05 < p < 0.10$). BDL = more than half of samples were below detection limit; therefore, the PAH was excluded from our analyses. Seasonal deployments of PSDs were as follows: 2012 summer = July 18 – August 21; 2012 fall = September 27 – October 24; 2013 spring = May 7 – June 5; 2013 summer = July 30 – August 26; 2013 fall = October 8 – November 5. Juvenile mussels deployed from June 5/6, 2012 to October 24/25, 2012 and from July 31/August 1, 2013 to November 5 – 7, 2013.

PAH	2012 Summer	2012 Fall	2013 Spring	2013 Summer	2013 Fall
Ace	<i>0.51</i>	<i>0.97*</i>	BDL	BDL	BDL
AN	BDL	<i>0.73*</i>	<i>0.58</i>	<i>0.94*</i>	0.45
BaA	<i>0.64*</i>	<i>0.99*</i>	BDL	BDL	BDL
C	BDL	<i>0.96*</i>	0.05	0.20	0.48
D	BDL	<i>0.80*</i>	BDL	BDL	BDL
F	BDL	<i>0.95*</i>	0.10	<i>0.71*</i>	0.25
F1	BDL	<i>0.66*</i>	BDL	BDL	BDL
F2	BDL	<i>0.90*</i>	BDL	BDL	BDL
F3	BDL	BDL	BDL	BDL	BDL
FL	<i>0.74*</i>	BDL	-0.02	<i>0.70*</i>	0.29
FP1	BDL	<i>1.00*</i>	0.29	0.35	0.22
N	<i>0.97*</i>	BDL	BDL	BDL	BDL
N1	0.38	BDL	0.05	0.30	0.48
N2	BDL	BDL	0.18	0.42	<i>0.64*</i>
N3	BDL	<i>0.92*</i>	<i>0.73*</i>	<i>0.71*</i>	<i>0.74*</i>
N4	<i>0.54</i>	<i>0.71*</i>	BDL	BDL	BDL
P	BDL	<i>0.89*</i>	0.35	0.43	0.17
P1	BDL	BDL	0.51	<i>0.55</i>	0.34
P2	BDL	<i>0.81*</i>	BDL	BDL	BDL
PER	BDL	0.22	BDL	BDL	BDL
PY	BDL	<i>0.96*</i>	-0.11	0.28	0.39
Sum34	<i>0.69*</i>	<i>0.98*</i>	0.48	0.45	<i>0.55</i>
Sum42	<i>0.64*</i>	<i>0.98*</i>	0.48	0.45	<i>0.57</i>

Supplemental Table S6. Spearman's rho correlation coefficients for correlations between site-specific June and October 2013 pore water metal concentrations and 2013 juvenile mussel trace element soft tissue concentrations. Bold and Italics with * = significant ($p \leq 0.05$); Bold and Italics only = marginal significance ($0.05 < p < 0.10$). BDL = more than half of samples were below detection limit; therefore, the metal was excluded from analyses.

Metal	<i>June 2013</i>	<i>October 2013</i>
Al	-0.30	0.20
As	BDL	0.38
Ba	0.11	0.03
Be	BDL	BDL
Cd	BDL	BDL
Co	0.45	0.15
Cr	<i>BDL</i>	0.33
Cu	0.01	-0.42
Fe	0.15	0.18
Hg	BDL	BDL
K	BDL	0.34
Mg	<i>0.68*</i>	<i>0.66*</i>
Mn	0.05	-0.08
Mo	0.24	-0.04
Ni	-0.27	-0.09
Pb	-0.43	0.14
Sb	BDL	BDL
Se	BDL	BDL
Si	BDL	BDL
Sr	0.01	-0.01
V	-0.43	0.29
Zn	-0.36	<i>0.50</i>

Supplemental Table S7. Spearman's rho correlation coefficients for correlations between site-specific June and October 2013 pore water PAH concentrations and 2013 juvenile mussel PAH tissue concentrations. Bold and Italics with * = significant ($p \leq 0.05$); Bold and Italics only = marginal significance ($0.05 < p < 0.10$). BDL = more than half of samples were below detection limit; therefore, the metal was excluded from analyses.

PAH	<i>June 2013</i>	<i>October 2013</i>
AN	0.06	BDL
C	-0.15	BDL
F	0.26	BDL
FL	0.49	BDL
FP1	0.09	BDL
N1	0.27	0.38
N2	0.09	BDL
N3	0.48	BDL
P	BDL	0.45
P1	<i>0.75*</i>	0.21
PY	0.38	BDL
Sum34	0.27	<i>0.69*</i>
Sum42	0.27	<i>0.69*</i>

Supplemental Table S8. Results from correlation analyses of site-specific mean total mussel density vs. metal concentrations measured in deployed mussel tissues in 2012 and 2013. r_s is the Spearman rho correlation coefficient. Bold and Italics with * = significant ($p \leq 0.05$); Bold and Italics only = marginal significance ($0.05 < p < 0.10$). Mussel densities were based on quantitative quadrat surveys conducted intermittently over the years 2005-2014.

Metal	<u>2012 Juveniles</u>		<u>2013 Juveniles</u>	
	r_s	p-value	r_s	p-value
Al	-0.64	0.09	0.50	0.21
As	0.07	0.87	-0.69	0.06
Ba	-0.31	0.46	-0.38	0.35
Cd	0.17	0.69	-0.35	0.40
Co	-0.62	0.10	0.14	0.74
Cr	-0.50	0.21	0.43	0.29
Cu	-0.52	0.18	0.57	0.14
Fe	-0.60	0.12	0.21	0.61
Hg	0.12	0.78	0.14	0.74
K	-0.79*	0.02	0.12	0.78
Mg	-0.24	0.57	0.00	1.00
Mn	0.26	0.53	0.19	0.65
Mo	-0.60	0.12	-0.05	0.91
Ni	-0.69	0.06	0.31	0.46
Pb	-0.62	0.10	0.45	0.26
Sb	-0.86*	0.01	-0.19	0.65
Se	-0.33	0.42	-0.45	0.26
Si	-0.62	0.10	0.62	0.10
Sr	-0.55	0.16	-0.24	0.57
V	-0.52	0.18	0.10	0.82
Zn	0.24	0.57	-0.40	0.32