

Supporting Information

Assessing the effects of bioturbation on metal bioavailability in contaminated sediments by diffusive gradients in thin films (DGT)

Elvio D. Amato^{†‡}, Stuart L. Simpson^{†*}, Timothy M. Remaili^{†‡}, David A. Spadaro[†], Chad V. Jarolimek[†], and Dianne F. Jolley^{‡*}

[†] Centre for Environmental Contaminants Research, CSIRO Land and Water, Locked Bag 2007, Kirrawee, NSW 2232, Australia

[‡] School of Chemistry, University of Wollongong, NSW 2522, Australia

* To whom correspondence may be addressed (djolley@uow.edu.au and stuart.simpson@csiro.au)

Phone: +61 2 442213516

Number of pages: 14

Summary of figures and tables:

Page S2 - Table 1. Total recoverable metal and metalloid concentrations in sediments

Page S3 - Figure S1. Relationships between DGT-metal fluxes measured in the sediment (between 0 and -3 cm depth) and OLW (within 1cm above the SWI).

Page S4 - Figure S2. DGT_{OLW}-metal concentrations measured within 1 cm above the SWI in the clean (S1) and the contaminated sediments (S2, S3) in presence of (i) no animals (zero bioturbation), (ii) bivalves (low bioturbation) and (iii) amphipods + bivalves (high bioturbation).

Page S5 - Figure S3. Relationships between *V. australiensis* bioaccumulation and DGT-metal fluxes (measured in different compartments of the sediment and overlying water) and dilute-acid extractable metal (AEM) concentrations (measured in the top 1 cm of the sediment).

Page S6 - Figure S4. Relationships between metal bioaccumulation in tissues of *T. deltoidalis* exposed to low and high bioturbation conditions and DGT-metal fluxes measured in different compartments of the sediment and overlying water.

Page S7 - Figure S5. Metal concentrations in *T. deltoidalis* and *V. australiensis*.

Page S8 - Figure S6. Comparison between relationships obtained using DGT_{OLW}-metal and dissolved metal concentrations with bioaccumulation in *T. deltoidalis* exposed to low (bivalves only, circles) and high (bivalves and amphipods, squares) bioturbation conditions.

Page S9 - Figure S7. Comparison between relationships obtained using DGT_{OLW}-metal and dissolved metal concentrations with bioaccumulation in *V. australiensis*.

Page S10 - Figure S8. Relationships between DGT_{OLW}-metal concentrations and dissolved metal concentrations measured in sediments overlying waters (OLW).

Page S11 - Figure S9. Vertical profiles of DGT-metal fluxes measured in pore and overlying waters in sediment S1 exposed to (i) zero (no organisms), (ii) low (bivalves) and (iii) high (amphipods and bivalves) bioturbation during first and second deployment.

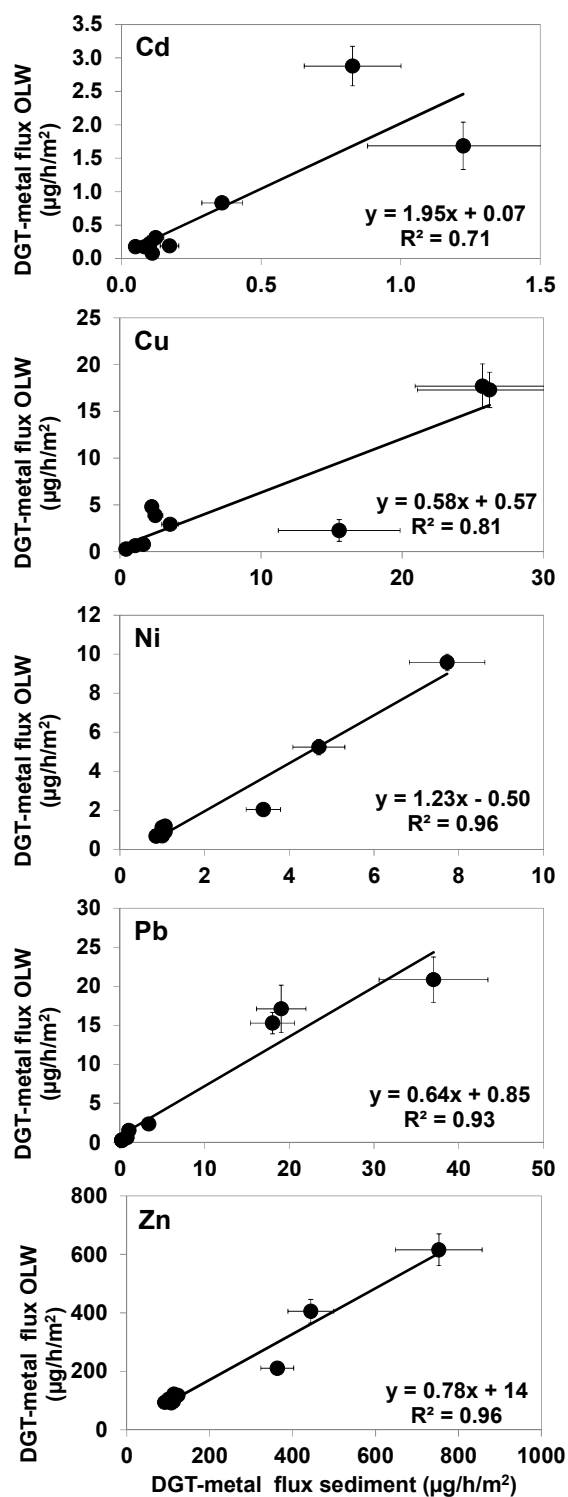
Page S12 - Figure S10. Vertical profiles of DGT-metal fluxes measured in pore and overlying waters in sediment S2 exposed to (i) zero (no organisms), (ii) low (bivalves) and (iii) high (amphipods and bivalves) bioturbation during first and second deployment.

Page S13 - Figure S11. Vertical profiles of DGT-metal fluxes measured in pore and overlying waters in sediment S3 exposed to (i) zero (no organisms), (ii) low (bivalves) and (iii) high (amphipods and bivalves) bioturbation during first and second deployment.

Table 1. Total recoverable metal and metalloid concentrations in sediments

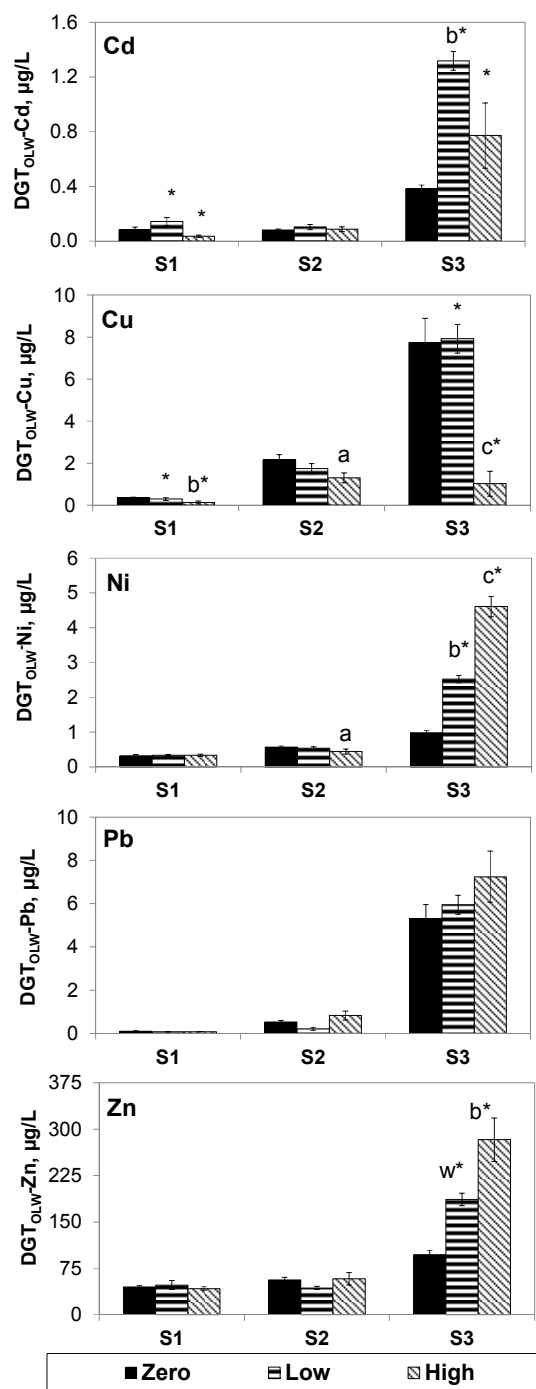
Sediment	Total Recoverable Metals (TRM), mg/kg									
	Al	As	Cd	Cr	Cu	Fe (%)	Mn	Ni	Pb	Zn
S1	0.3 ± 0.04	2.1 ± 0.3	0.3 ± 0.1	5.0 ± 1	16 ± 2	0.8 ± 0.1	44 ± 16	2.3 ± 0.3	10 ± 1	37 ± 4
S2	1.1 ± 0.1	40 ± 2*	2 ± 0.2*	68 ± 1	550 ± 10*	4.0 ± 0.5	240 ± 90	19 ± 0.1	460 ± 6*	900 ± 3*
S3	1.0 ± 0.04	50 ± 1*	8 ± 0.1*	390 ± 10*	1200 ± 30*	0.5 ± 0.02	67 ± 2	36 ± 0.7*	1400 ± 20*	2800 ± 30*

Concentrations are averages with standard deviation (n=3). Asterisks* indicate concentrations exceeding SQG threshold values (ANZECC/ARMCANZ 2000)



2

3 Figure S1. Relationships between DGT-metal fluxes measured in the sediment (between 0 and -3 cm
 4 depth) and OLW (within 1cm above the SWI). Data points are averages of 12 (3 depths × 3 replicates
 5 × 2 deployments) and 24 (4 depths × 3 replicates × 2 deployments) measurements for DGT_{OLW} and
 6 DGT_{SEDIMENT}, respectively, with standard error.



7
8 Figure S2. DGT_{OLW}-metal concentrations measured within 1 cm above the SWI in the clean (S1) and
9 the contaminated sediments (S2, S3) in presence of (i) no animals (zero bioturbation), (ii) bivalves
10 (low bioturbation) and (iii) amphipods + bivalves (high bioturbation). Mean values are the average of
11 six replicates reported with standard error. Significant differences between treatments exposed to
12 biota and undisturbed sediments are indicated with the letters *a* ($p < 0.05$), *b* ($p < 0.01$) and *c* ($p < 0.001$),
13 and differences between treatments exposed to biota are indicated by an asterisk. The letter *w*
14 indicates weak differences ($0.05 < p < 0.06$).

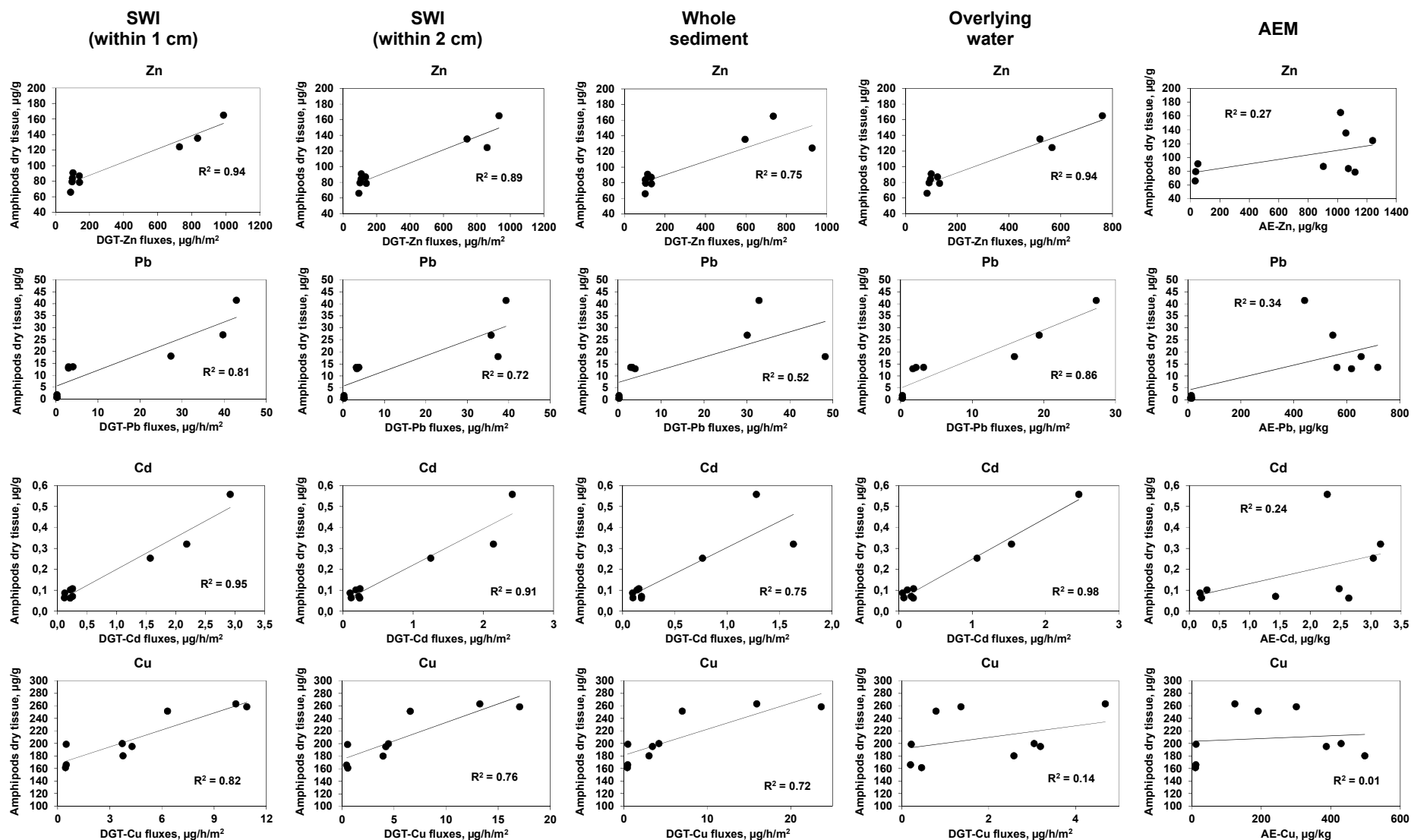


Figure S3. Relationships between *V. australiensis* bioaccumulation and DGT-metal fluxes (measured in different compartments of the sediment and overlying water) and dilute-acid extractable metal (AEM) concentrations (measured in the top 1 cm of the sediment). Data points are mean values with standard error ($n=3$ for tissue concentrations and AEM concentrations, $n=12$ for DGT fluxes at the SWI (within 1cm) and overlying water, and $n=24$ for DGT fluxes at SWI (within 2cm) and whole sediment).

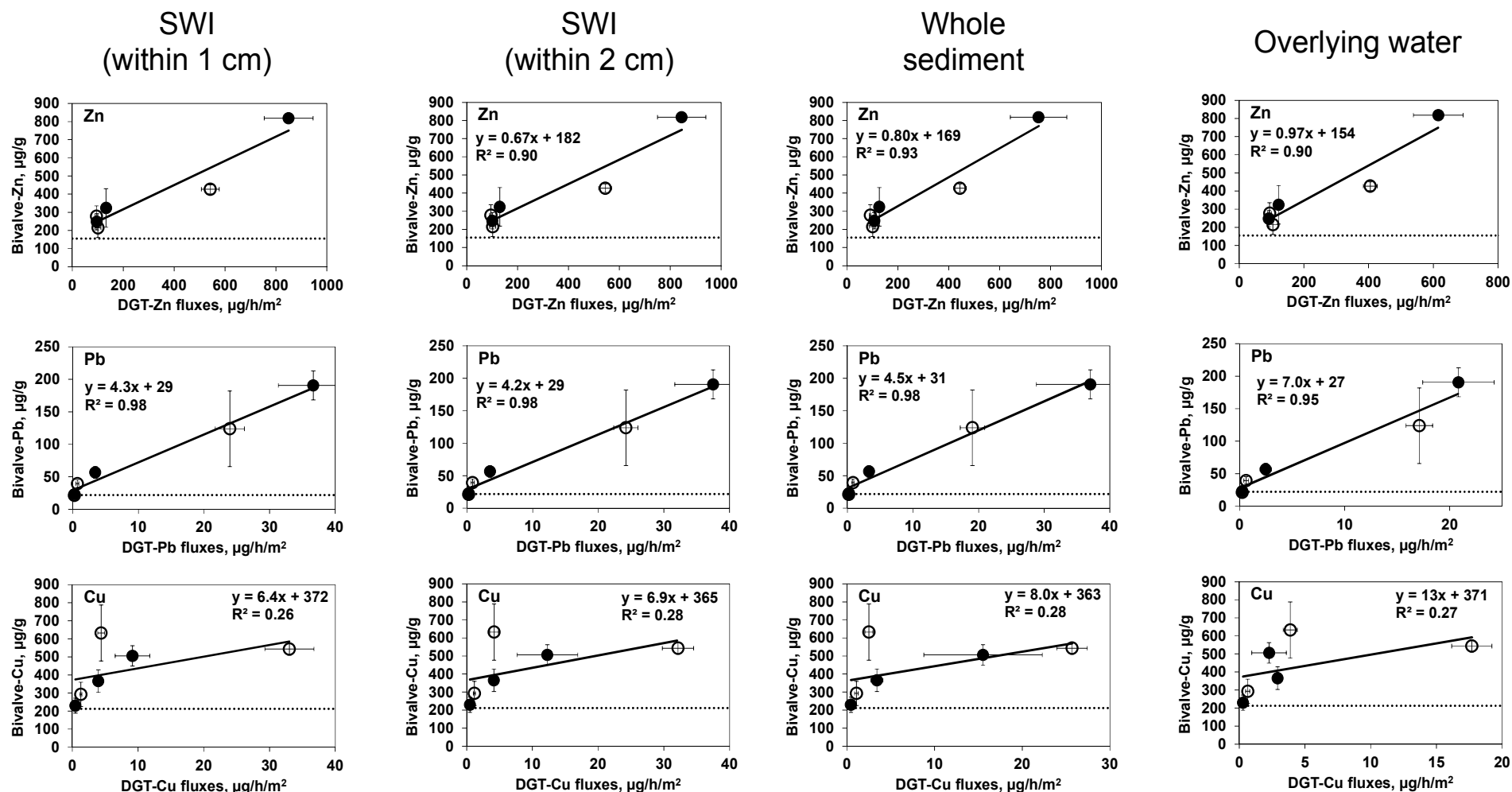
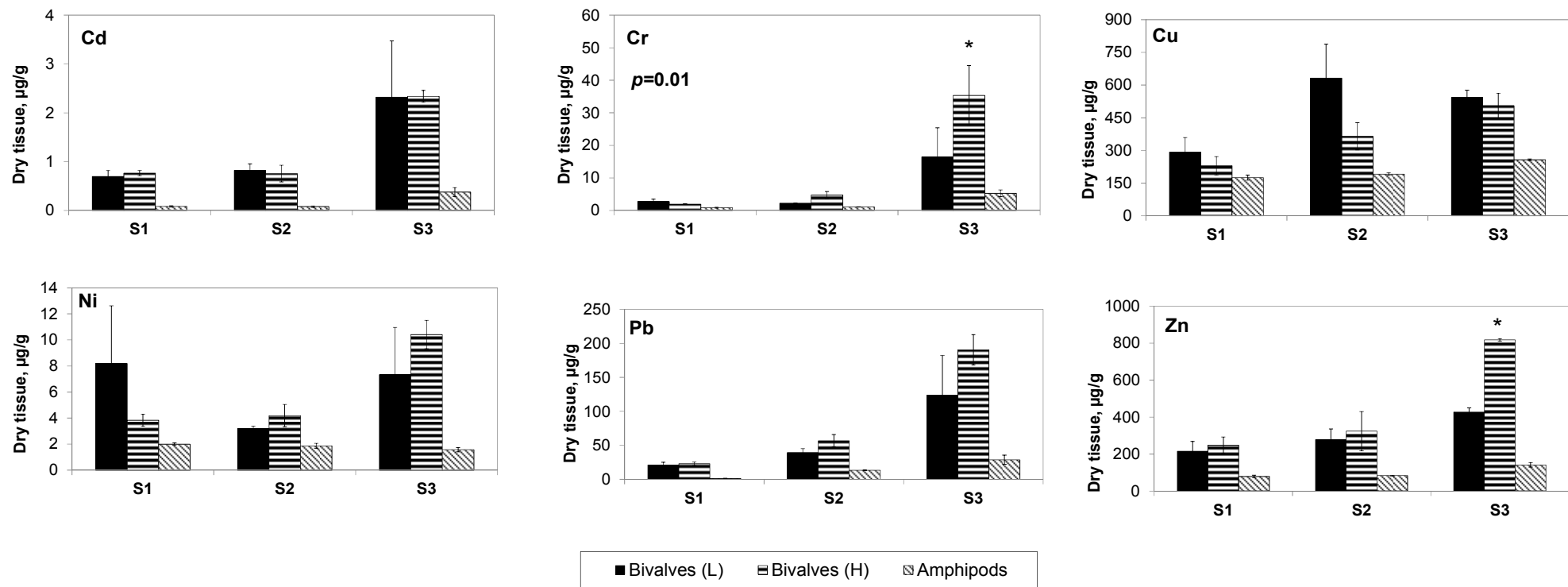


Figure S4. Relationships between metal bioaccumulation in tissues of *T. deltoidalis* exposed to low (bivalves only, empty circles) and high (bivalves and amphipods, full circles) bioturbation conditions and DGT-metal fluxes measured in different compartments of the sediment and overlying water. DGT-metal fluxes are average values of first (day 8) and second (day 23) deployment. Data points are mean values with standard error ($n=3$ for tissue concentrations, $n=12$ for DGT fluxes at the SWI (within 1cm) and overlying water, and $n=24$ for DGT fluxes at SWI (within 2cm) and whole sediment). The dotted line indicates average metal concentrations measured in non-exposed bivalves.

25



26

27 **Figure S5.** Metal concentrations in *T. deltoidalis* (bivalve) and *V. australiensis* (amphipod). Data are mean values of three replicates with standard error. Asterisk* indicates
28 statistical differences between *T. deltoidalis* exposed to low (L - dark fill) and high (H - dark horizontal fill) bioturbation.

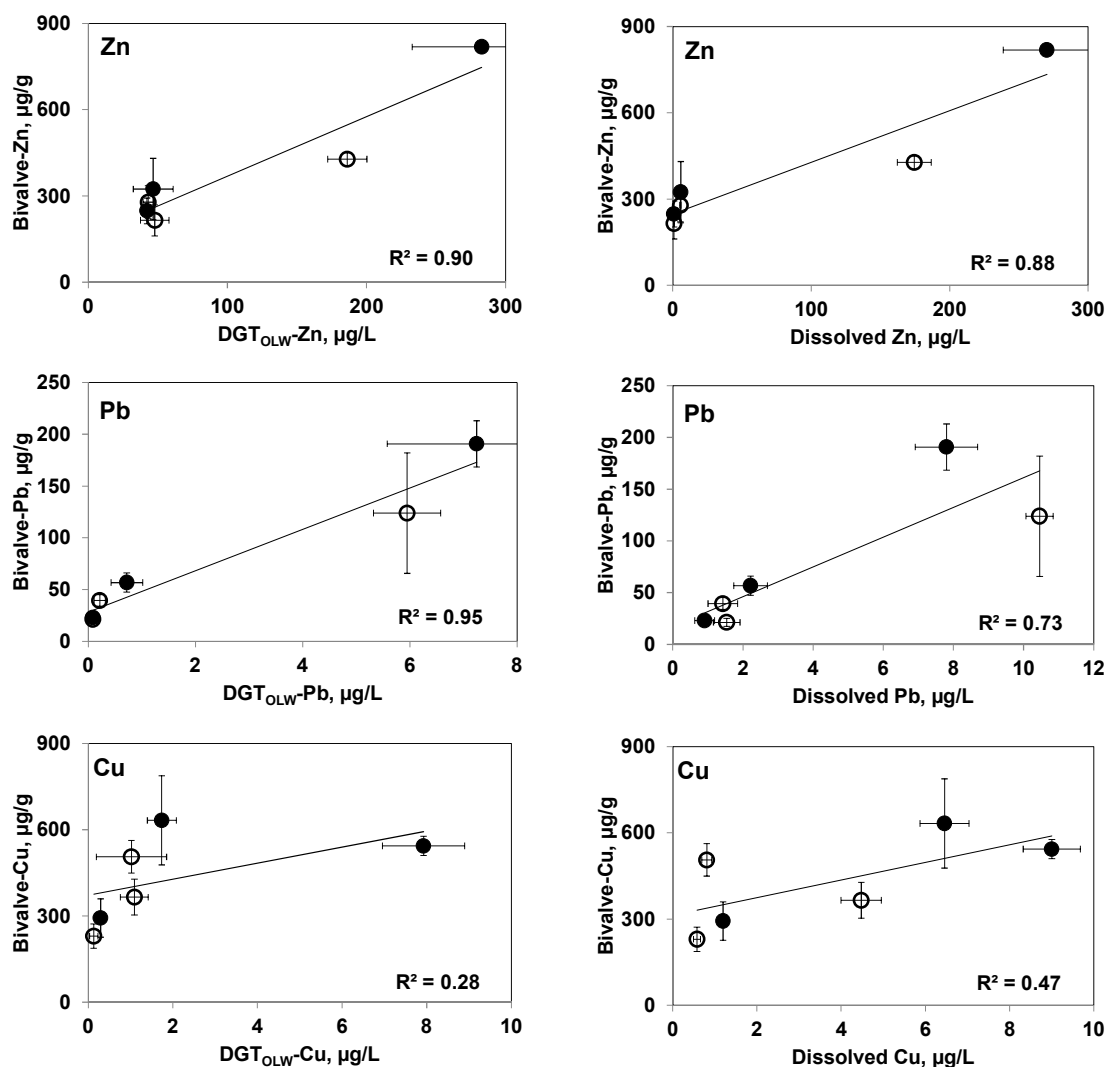


Figure S6. Comparison between relationships obtained using DGT_{OLW}-metal and dissolved metal concentrations with bioaccumulation in *T. deltoidalis* exposed to low (bivalves only, empty circles) and high (bivalves and amphipods, full circles) bioturbation conditions. DGT-metal fluxes are average values of first (day 8) and second (day 23) deployment. Dissolved metal concentrations are average values of overlying water subsamples collected throughout the test. Data points are mean values reported with standard error (n=6 and 15 for DGT-metal fluxes and dissolved metal concentrations, respectively). The dotted line indicates average metal concentrations measured in non-exposed bivalves.

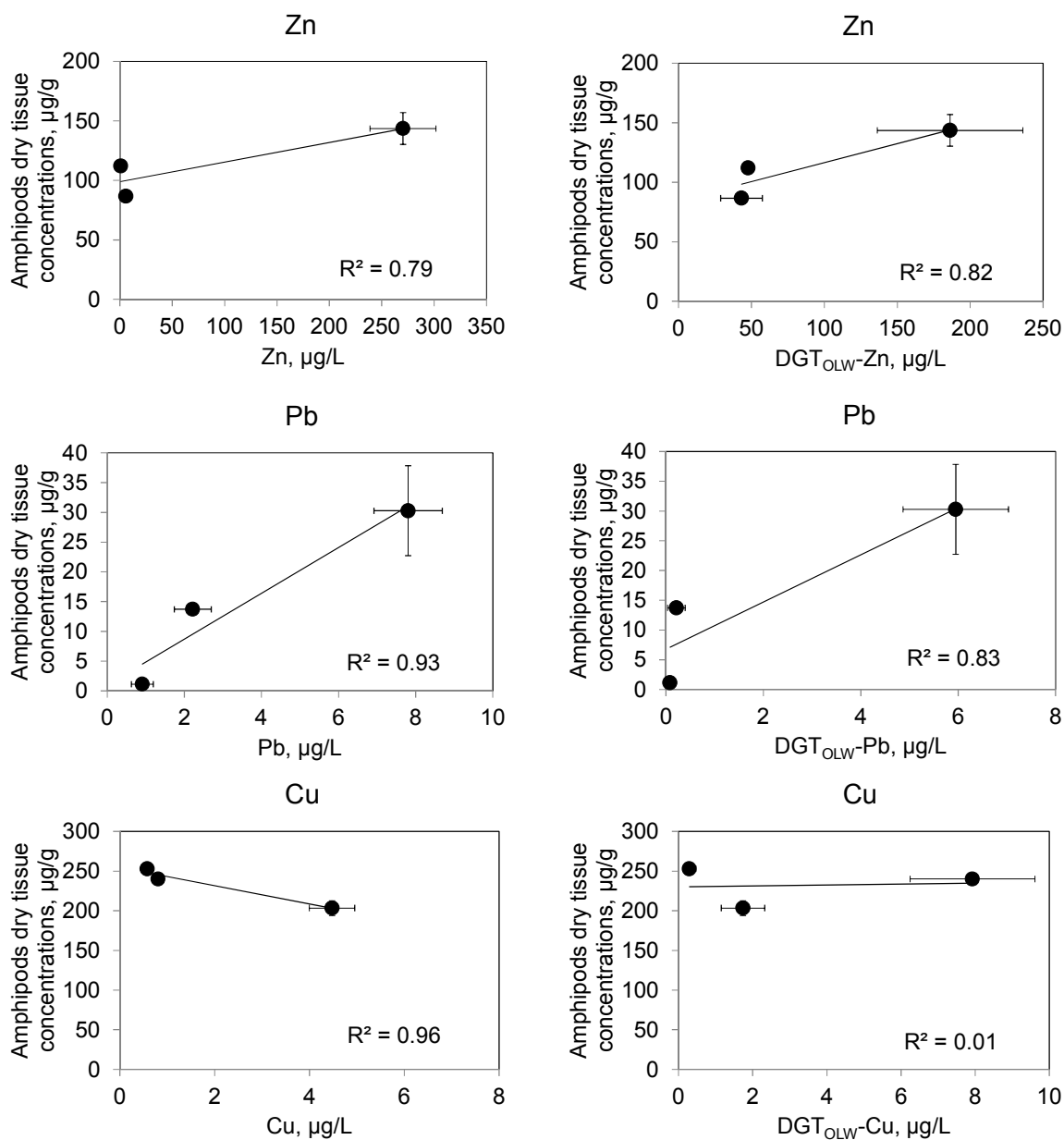


Figure S7. Comparison between relationships obtained using DGT_{OLW}-metal and dissolved metal concentrations with bioaccumulation in *V. australiensis*. DGT-metal fluxes are average values of first (day 8) and second (day 23) deployment. Dissolved metal concentrations are average values of overlying water subsamples collected throughout the test. Data points are mean values reported with standard error (n=6 and 15 for DGT-metal fluxes and dissolved metal concentrations, respectively).

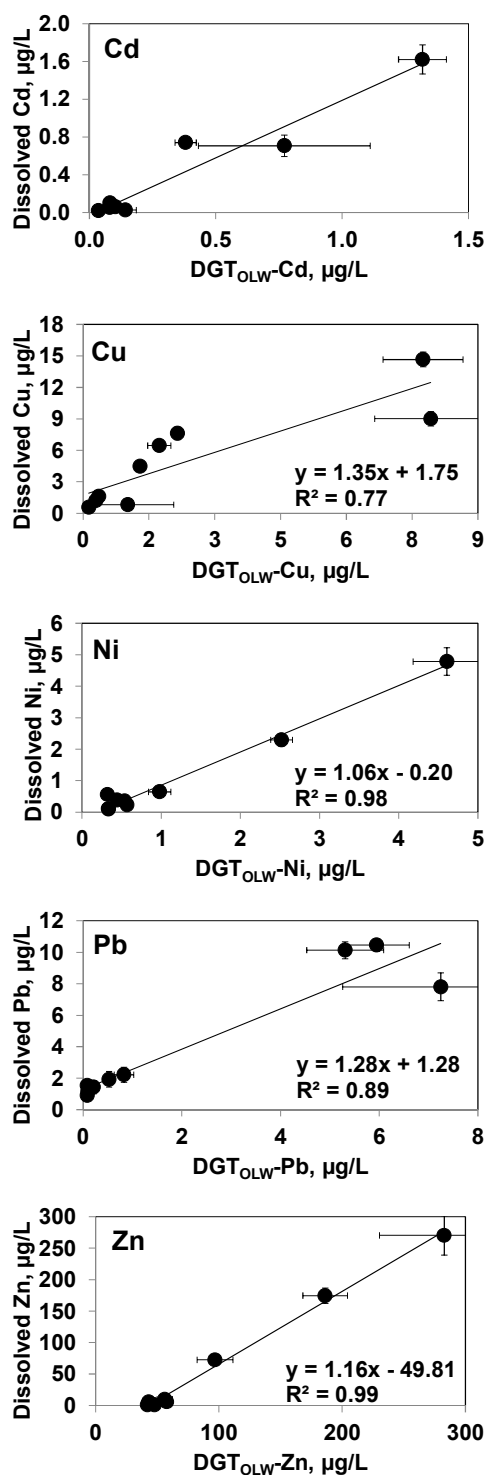


Figure S8. Relationships between DGT_{OLW}-metal concentrations and dissolved metal concentrations measured in sediments overlying waters (OLW). DGT_{OLW}-metal concentrations are mean values with standard error of 3 replicate measurements performed on day 8 and 23 (n=6). Dissolved metal concentrations are the average concentration measured in OLW aliquots sampled throughout the test reported with standard error (n=15).

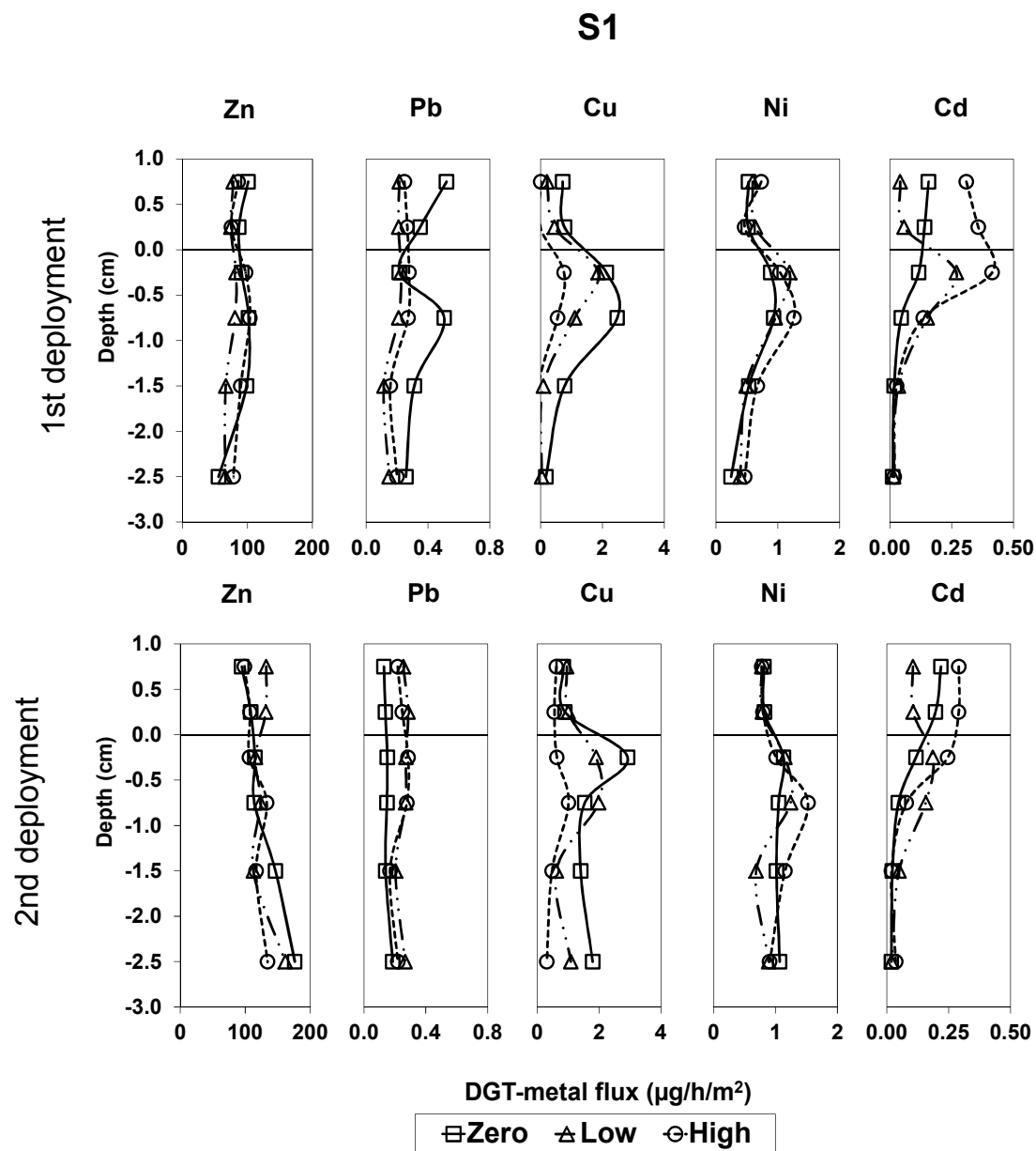


Figure S9. Vertical profiles of DGT-metal fluxes measured in pore and overlying waters in sediment S1 exposed to (i) zero (no organisms), (ii) low (bivalves) and (iii) high (amphipods and bivalves) bioturbation. Data points are averages of three replicates, and associated standard errors were on average <20%.

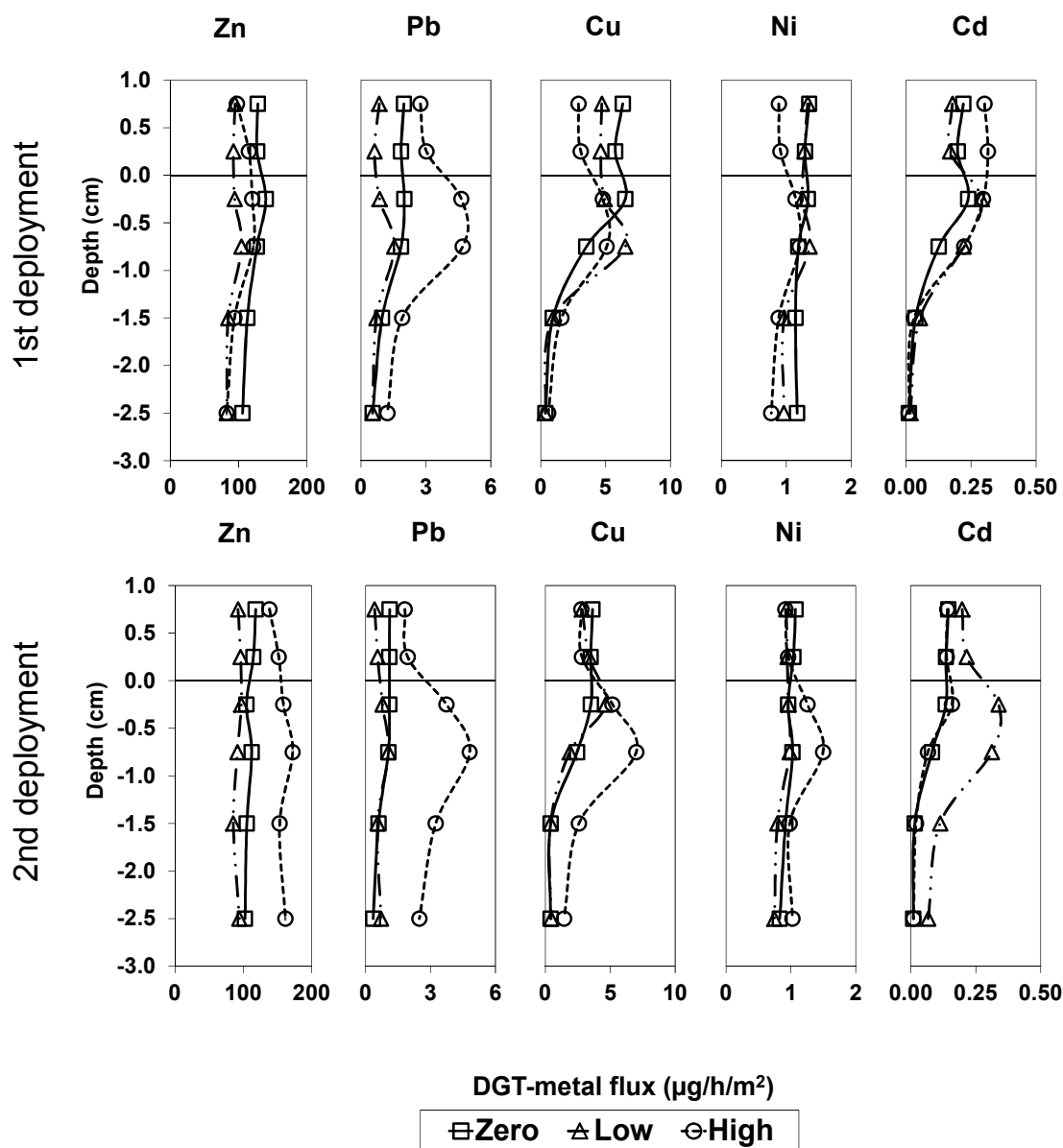


Figure S10. Vertical profiles of DGT-metal fluxes measured in pore and overlying waters in sediment S2 exposed to (i) zero (no organisms), (ii) low (bivalves) and (iii) high (amphipods and bivalves) bioturbation. Data points are averages of three replicates, and associated standard errors were on average <20%.

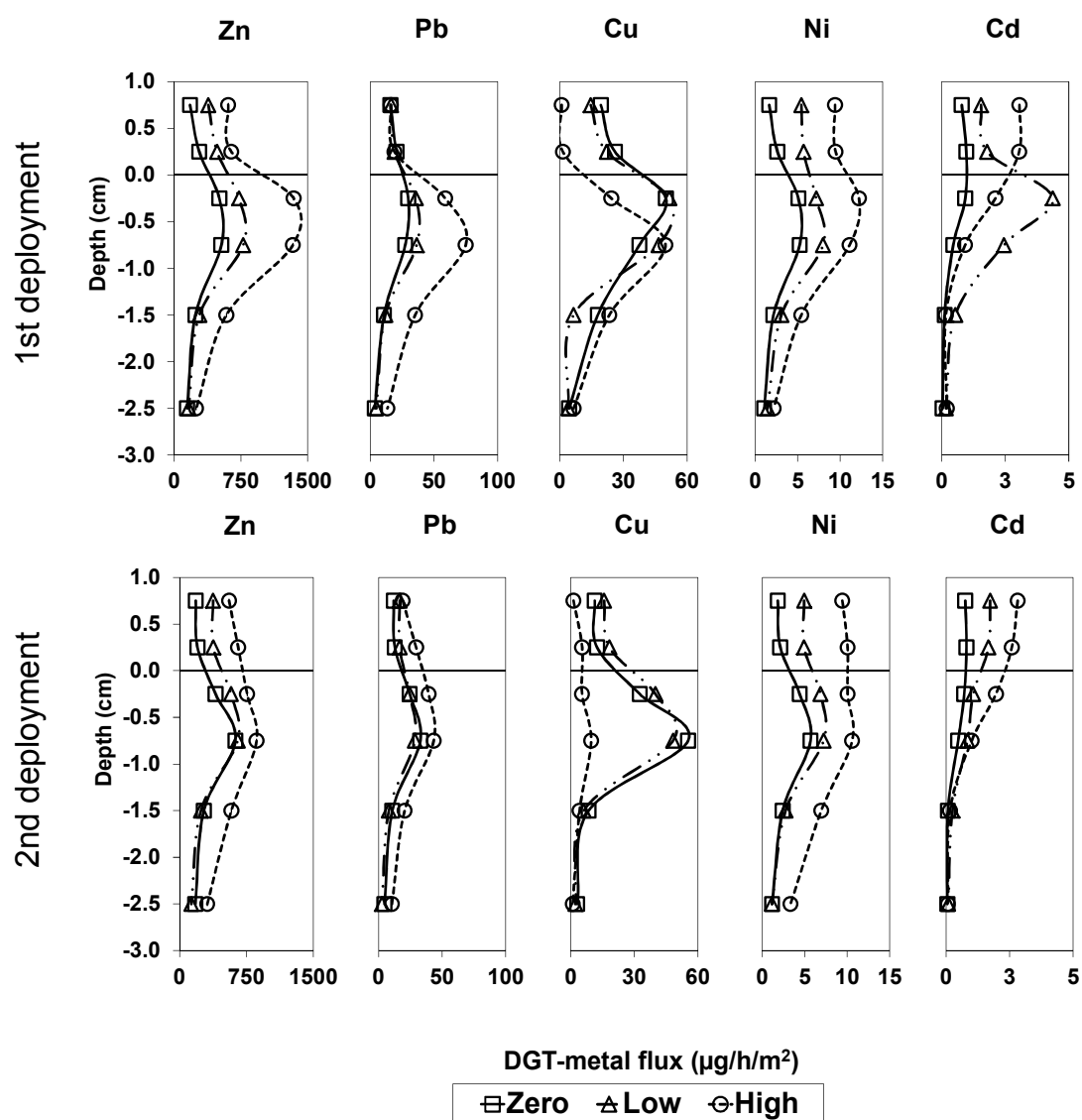


Figure S11. Vertical profiles of DGT-metal fluxes measured in pore and overlying waters in sediment S3 exposed to (i) zero (no organisms), (ii) low (bivalves) and (iii) high (amphipods and bivalves) bioturbation. Data points are averages of three replicates, and associated standard errors were on average <20%.