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BASELINE SURVEY OF SURFACE SEDIMENT TOTAL MERCURY CONCENTRATIONS,

NARRAMISSIC RIVER, MAINE

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SUMMARY

In November 2015 we conducted a baseline survey of mercury concentrations in the sediments of the Narramissic River near Orland, Maine. Surface sediment samples collected in triplicate from 12 sites along the river and in the adjacent marshes at Duck Cove and Wight's Brook were analyzed for total mercury, total organic carbon and grain size.

Elevated mercury concentrations, up to ten times greater than regional background (28 – 38 ng/g dw), were found in the surface sediments of the Narramissic River. Mercury concentrations were greatest directly upstream of the Orland Dam (305 ng/g dw), and decreased to a low of 69 ng/g dw near the Upper Falls Road Bridge, 4 km to the northeast. At these concentrations sediment toxicity to benthic organisms is not expected, but is possible at the site closest to the Orland Dam. Mercury concentrations in the marsh sediments were in the lower range of concentrations found in the river, averaging 66 to 115 ng/g dw. However, mercury concentrations in the marsh sediments were influenced by the relatively high total organic carbon content of the marsh soils.

In the river, the decline in mercury concentrations with distance from the dam was not altered by variations in total organic carbon or sediment grain size. This finding supports the hypothesis that the mercury in the surface sediments of the Narramissic River came from contaminated sediments present in the Orland River that moved over or through the Orland Dam, carried by storm surges or tidal flows, and into the Narramissic River.

Given this finding, the decision to remove or alter the Orland Dam must consider the possibility that changes to the dam will allow tidal water to carry additional contaminated sediment into the Narramissic.

INTRODUCTION

The Narramissic River was dammed just below the town of Orland, Maine in the early 1900s, creating an upstream freshwater embayment extending from Orland to the vicinity of Alamoosic Lake, located approximately four kilometers upstream to the northeast. By design, the Orland Dam, a head of tide dam, formed the upper tidal extent of the Orland River.

Today the Narramissic River is a bucolic reservoir. Shoreside houses line the river near Orland Village and further upstream scattered houses are interspersed with long stretches of undeveloped riparian habitat. Freshwater grassy-shrub marshes are located at Duck Cove and Wight's Brook, which enter the Narramissic from the north. A cattail marsh dominates the southeast shore upstream from Wight's Brook.

Within the last 50 years mercury discharges from the former HoltraChem chlor-alkali plant in Orrington, Maine significantly contaminated the lower Penobscot River, from Brewer, Maine, south to Fort Point (PRMS 2013, Rudd et al. 2013). Mercury contaminated sediment has accumulated throughout this area, including in the Orland River directly downstream of the Orland Dam. Removal of the Orland Dam and the restoration of tidal flows in the Narramissic River may introduce contaminated sediment from the Orland River into the Narramissic River.

At the beginning of this study we hypothesized that mercury from the HoltraChem discharges had already contaminated the river upstream of the Orland Dam. Historically, the dam had fallen into periods of disrepair, and had been overtopped by storm surge and extreme spring tides (Chelminski and Giumarro 2013). During periods of disrepair, or overtopping, tidal water carrying contaminated sediment from the Orland River may have moved upstream of the dam and entered the Narramissic River.

This study analyzed surface sediment samples to answer whether there has been recent movement of mercury contaminated sediment from the Orland River to areas upstream of the Orland Dam. A companion study analyzing sediment cores would provide critical information on the historical movement of mercury upstream of the dam, and the total volume of mercury stored in the sediment.

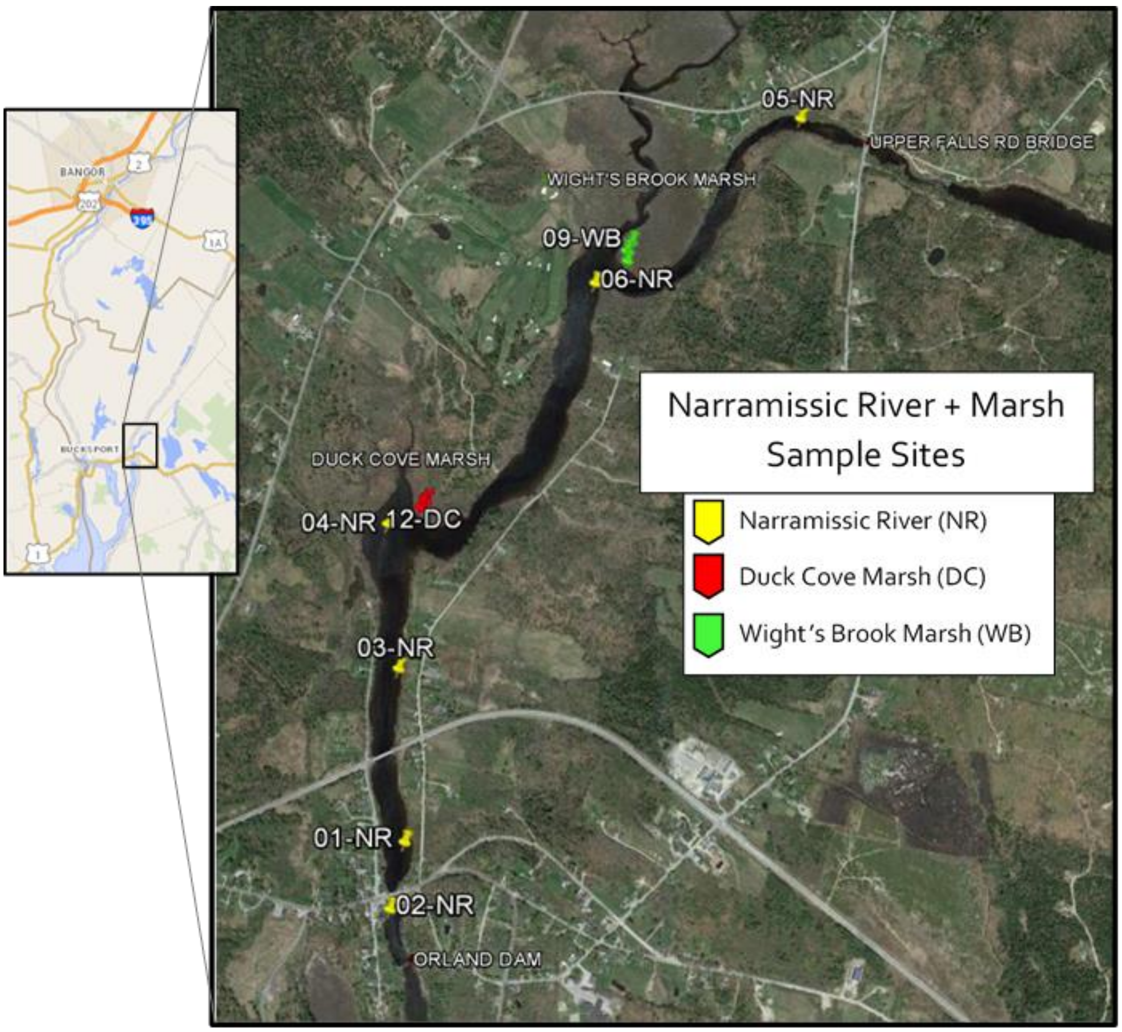

Figure 1. Sediment sample sites within the Narramissic River and adjacent marshes.

METHODS

Sediment samples were collected from the Narramissic River and adjacent marshes November 10-18, 2015. Six sampling sites were located in sediment deposits on either side of the main river channel between the Orland Dam and the Upper Falls Road Bridge. Little to no current was evident during the November sampling period except in the immediate vicinity of the dam and near the bridge at the northern end of the sample area. Sediment samples were also collected from the marshes on the north shore of the river, at Duck Cove and at the mouth of Wight's Brook. (Figure 1) Each marsh was sampled at three sites along a transect running from the outer limit of vegetation at the water's edge, up onto the marsh platform and inland to the edge of woody vegetation. (If the marshes are restored to tidal action, the habitat characteristics of these sites will change and the collection sites will be located at the outer edge of the intertidal vegetation, the berm on the outer edge of the marsh platform, and the interior of the marsh platform.)

Figure 2. Core head with sediment sample pulled from river.

At each site, three replicate samples were collected at equidistant points along a 3-meter transect. Each sample was a composite of three cores collected within a 0.5 meter square plot. Samples were collected using a Universal Core Head (WaterMark™) with polycarbonate barrels, 67 mm I.D, cut to a length of 30 cm (Figure 2). The core head was dropped vertically through the water until it touched bottom, then driven into the sediment using a 10 kg weight to an average depth of 10 – 15 cm, except at one site where sediment characteristics limited the total core depth to 3 – 5 cm. Only the top three centimeters (0 – 3 cm) of each surface core was collected, using an incremental core extruder with funnel (Aquatic Research Instruments), and transferred to a stainless steel bowl for mixing. After homogenization, each composite sample was divided between two types of sample jars, 120 ml HDPE jars for mercury analysis and 240 ml glass jars for sedimentology, and placed in a cooler with ice for transport from the field. Sedimentology samples were refrigerated at 4^{o}C prior to analysis and transported on wet ice to the laboratory; samples for mercury analysis were frozen at -20^{o}C and shipped on dry ice to the laboratory.

Sedimentology analyses were conducted by Katahdin Analytical Services in Scarborough, Maine. Total organic carbon (TOC) was determined using EPA method 9060, samples were analyzed within the prescribed hold time of 28 days. Sediment grain size was determined using the ASTM D422 sieve method. Standard chain of custody and QA/QC standards were met for each type of analysis.

Total mercury analyses were performed by Flett Research Ltd. In Winnipeg, MB, Canada, using EPA method 7473 for a DMA-80 Total Mercury Analyzer. Samples were freeze-dried and ground prior to analysis to ensure complete homogenization. QA/QC standards were met for laboratory duplicates, sample spikes, and blanks.

Statistical analyses were done using R. Variables were log-transformed to meet normality assumptions for ANOVA and regression models.

FINDINGS

A total of 36 composite sediment samples (0 – 3 cm depth) were collected from 12 sites along the Narramissic River and adjacent marshes. Appendix 1 summarizes the total mercury concentrations and sedimentology for each of the 12 sample sites.

1. **Mercury concentrations in surface sediment of the Narramissic River are up to ten times greater than background mercury concentrations found in the sediment of nearby coastal rivers.**

Low sediment mercury concentrations are expected in Maine's coastal rivers unless there is a local source of mercury contamination. Bodaly (2013) reported both historical mercury concentrations in sediment and present day background concentrations of mercury in two coastal rivers which were outside of the direct aquatic influence of the HoltraChem discharges on the Penobscot. Pre-industrial levels of mercury in sediment were reported at 20 ng/g dw based on deep cores 55 to 75 cm below the surface of the sediment, sampled from upper Fort Point Cove at the mouth of the Penobscot River.

Currently, regional background mercury concentrations in the surface sediments of coastal rivers with no known source of mercury contamination were only slightly higher than pre-industrial levels. In the lower Narraguagus River near Milbridge, Maine, the mean total mercury concentration in surface sediments was 28 ± 2 ng/g dw (mean ± SD) and in the St. George River just west of Penobscot Bay the mean mercury concentration was 38 ± 13 ng/g dw. These local background concentrations are in the range for background concentrations for total mercury in sediment reported nationally by NOAA (National Oceanic and Atmospheric Administration) of 2 – 51 ng/g dw (Buchman 2008).

Mercury concentrations in the surface sediments of the lower Narramissic River exceeded regional background levels. Among the surface sediment samples collected at six sites along the length of the Narramissic total mercury concentrations ranged from 69 ± 12 ng/g dw (mean ± SD) to 305 ng/g dw. The highest mean concentrations in the river were roughly 10 times greater than regional background concentrations. Total mercury concentrations in marsh sediments ranged from 66 ± 11 ng/g dw to 94 ± 0.6 ng/g dw.

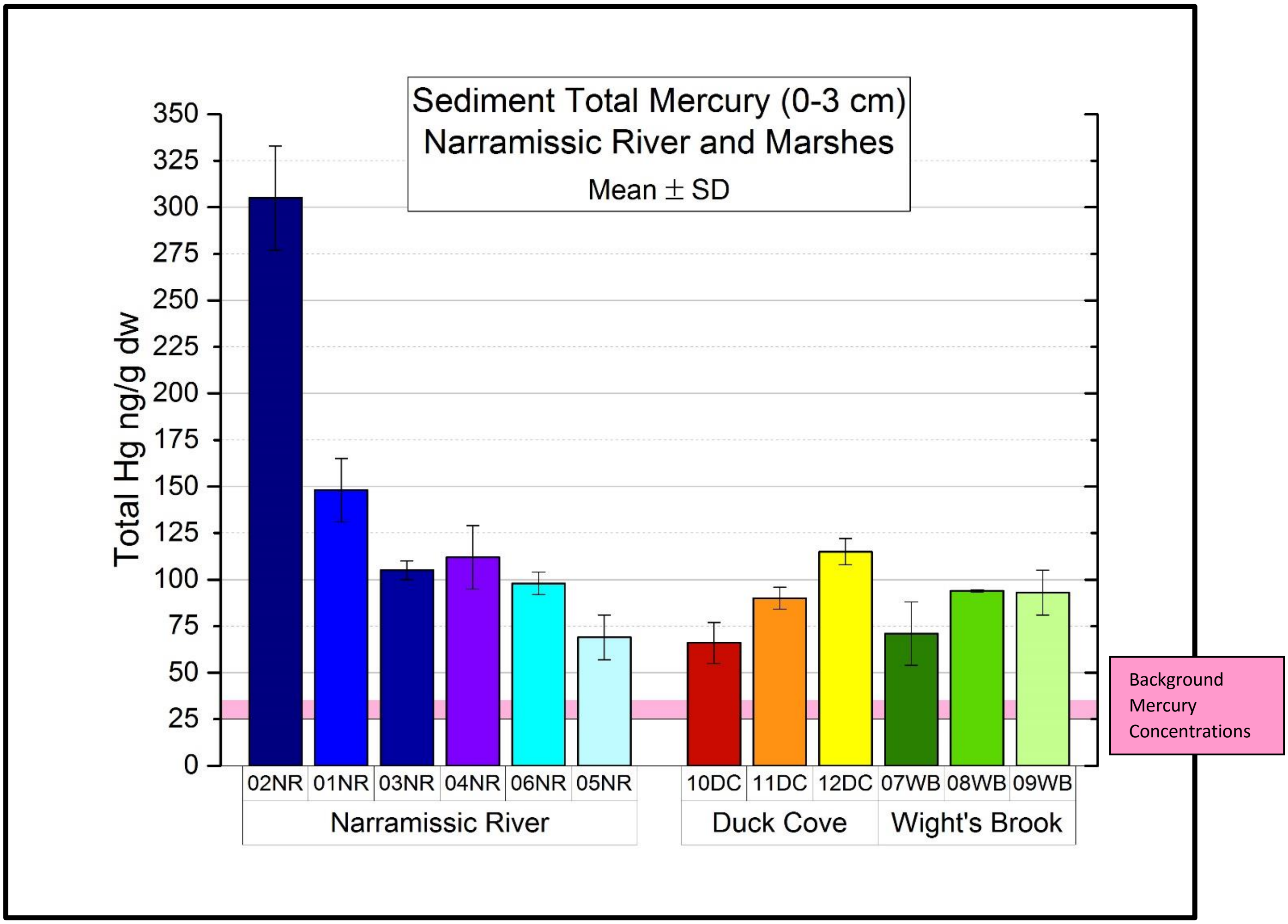

Figure 3. Total mercury concentrations in the surface sediments (0-3cm) of the Narramissic River and adjacent marshes. River sites are ordered from south (near the Orland Dam) to north (near Upper Falls Road Bridge), and marsh sites are ordered from the water's edge inland to the marsh platform. The pink band between 28 and 38 ng/g dw represents background sediment mercury concentrations found in coastal Maine rivers. Note the significant decline in mercury concentrations with distance from the Orland Dam.

2. **In the Narramissic River the significant decline in sediment mercury concentrations with distance upstream from the Orland Dam supports the hypothesis that contaminated sediment from the Orland River moved over or through the dam on incoming tides and is the source of mercury contamination.**

In the Narramissic River total mercury concentrations in the surface sediments were significantly greater at the sites closest to the Orland Dam, 305 ± 28 ngHg/g dw (mean ± SD; 02NR), and decreased with distance upstream to a low of 69 ngHg/g dw at the site near the Upper Falls Road Bridge (ANOVA, $P < 0.05$; Tukey HSD, $\alpha = 0.05$; Figure 3). This declining mercury gradient with distance from the dam is consistent with the mercury source being the contaminated sediment downstream of the Orland Dam. As a head tide dam, the Orland Dam, when intact, blocks the upstream movement of tidal water. However, tidal water can flow over or through the dam during extreme tides, storm surges or when the dam is damaged and in need of repair (Chelminski and Giumarro 2013).

In coastal rivers such as the Orland River, surface sediment is re-suspended and relocated daily by tidal currents (Wang 2002) and can be carried upstream on an incoming tide. The Penobscot River illustrates this process by which tidal waters carry contaminated sediment upstream. Contaminated sediment is found both upstream and downstream of the mercury source at the former HoltraChem plant site in Orrington (Rudd et al. 2013).

3. **Normalizing the river sediment mercury concentrations to total organic carbon (TOC) or sediment grain size (% fines) did not change the observed gradient in mercury concentrations with distance from the dam.**

The physical characteristics of sediment are known to influence mercury concentrations and the methylation of inorganic mercury into a more bioavailable and toxic form. Mercury preferentially binds to organic carbon and variation in organic carbon concentrations can explain over 90% of the variation in particulate mercury concentrations (Schuster et al 2008). Similarly, fine-grained sediments have a greater surface area available for binding mercury and other trace metals (Chakraborty et al 2015, Yu et al. 2012). In general, mercury retention is greater in fine-grained sediments with high total organic carbon (TOC) and elevated organic carbon can increase methylation in conjunction with other factors (Mitchell and Gilmour 2008). Both sediment grain size and TOC were examined to reduce bias when comparing mercury concentrations among sites. Note, however, that sediment mercury concentrations on a dry weight (dw) basis, not normalized to sediment organic carbon, are used to predict sediment toxicity (Macdonald et al. 2000).

Total organic carbon concentrations (TOC) in the surface sediments of the Narramissic River were more variable at the two downstream sites closest to the dam than at the riverine sites further upstream. Sites 02NR and 01NR (Figure 1) had mean TOC concentrations of 587 and 307 mg/g dw, respectively. TOC was fairly constant at the four upstream sites, ranging from 467 to 493 mg/g dw. Marsh sediments, influenced by the high organic carbon input from decaying marsh vegetation, had notably greater TOC concentrations than found in the river, up to 1,900 mg/g dw. Within each marsh TOC generally increased from the water's edge to the marsh platform (Appendix 1).

The percentage of fine grains (<75 µm in diameter) in the riverine sediment varied by roughly 15 %. Grain size was smallest near Duck Cove in the center of the sample area, with 53 % fines, while coarser sediment was collected near the Orland Dam, with only 38 % fines, and near the Upper Falls Road Bridge, with 45 % fines. This slight variation in grain size may relate to relatively faster currents noted at either end of the study area. Fine grained sediments tend to settle to the bottom in calm water. Marsh sediments, dominated by organic material, were coarser than sediments from the river, with percent fines ranging as low as 18 to 26 % on the marsh platform.

There was no substantive change in the gradient of sediment mercury levels following normalization to total organic carbon (TOC, Figure 4), confirming that the elevated mercury concentrations near the dam were not solely the result of increased TOC in that area. Note in Figure 4 the relative increase in mercury when normalized to organic carbon at the site 01NR, where TOC was at the lowest concentration found in this study. Similarly, total mercury normalized to % fines (sediment grain size < 75 µm) showed the same declining gradient with distance upstream from the dam.

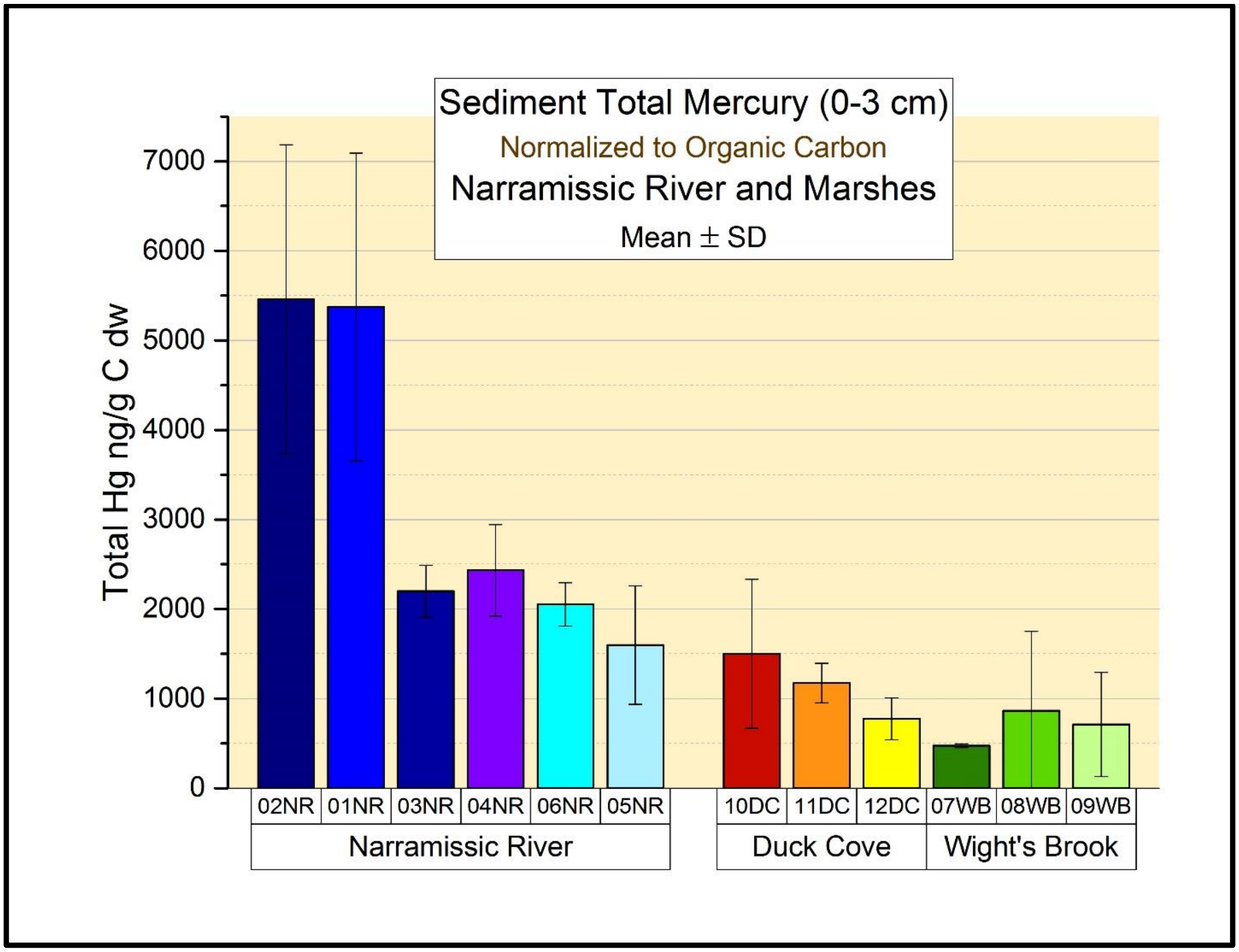

Figure 4. Total mercury concentrations normalized to total organic carbon in surface sediments of the Narramissic River and adjacent marshes. River sites are ordered from south to north, and marsh sites are ordered from the water's edge inland to the marsh platform. Note that the gradient of mercury from south to north is retained after carbon normalization and that the concentration of mercury in most marsh sediments is reduced relative to the concentrations found in the river.

4. **The relative concentrations of mercury in marsh sediments were altered by normalization to TOC reflecting the high total organic carbon concentrations in the marsh sediments.**

Total mercury concentrations in the two marshes were similar to those found in the upper river, but strongly influenced by the relatively elevated TOC concentrations present in the marsh sediment. At Duck Cove Marsh the significant trend of increasing total mercury concentrations from the water's edge to the interior marsh platform (Figure 3) was reversed when mercury concentrations were normalized to TOC in the marsh sediment (Figure 4). At Wight's Brook Marsh the relative pattern of mercury concentrations in the sediment was not changed by TOC normalization, but given the extremely high TOC concentrations found at that marsh the mercury concentrations normalized to organic carbon declined to the lowest levels found at any sample site.

Given these findings, caution must be used when comparing sediment mercury concentrations in the river and the adjacent marshes. The relatively elevated mercury concentrations in the marsh sediments is at least partially driven by the high organic carbon content of the sediment and does not indicate an alternate or secondary mercury source.

5. **The concentrations of mercury in the surface sediments of the Narramissic River are up to six times lower than found in the highly contaminated Orland River.**

The thick, fine sediments of the Orland River are highly contaminated with mercury discharged from the former HoltraChem plant in Orrington. Elevated mercury concentrations are found both at the surface, and at depth (Yeager 2013). Surface mercury concentrations (0 - 3 cm) in the Orland River range from 375 ng/g dw, in the cove west of Fish Point, to over 1,800 ng/g dw at site OR2B, 900 m downstream from the Orland Dam.

Further south in the Orland River, some deeper layers of sediment contain even greater concentrations of mercury, up to 4,600 ng/g dw at 30 cm in depth. Since particles accumulate on the surface of the sediment over time, mercury concentrations at depth indicate deposition from historical mercury discharges. This buried sediment is generally not bioavailable if it is not disturbed. However, sediment at depth can be re-suspended by erosion or course changes in slough channels, large scale flooding, or during major storms, especially if storms occur at low tide (Walker and Hammack 2000; Kitheka et al. 2005; Rao et al. 2011). Figure 5 illustrates the concentration of mercury in the surface sediments in the Orland River, downstream of the dam, and in the Narramissic River upstream of the Orland Dam.

6. **Based on NOAA screening values for mercury in sediment, toxicity to sediment dwelling organisms is not expected at most sites sampled in the Narramissic River. However, at site 02NR closest to the Orland Dam, toxicity to sediment dwelling organisms remains a possibility.**

Elevated total mercury concentrations in surface sediments (0-3 cm) indicate recent mercury contamination, and the potential for mercury to enter the food web in the Narramissic River. Mercury exists in several chemical forms. Of greatest concern to the food web is methyl mercury (CH_3Hg^+), which is converted from inorganic mercury by bacteria present in the top few centimeters of the sediment. Methyl mercury can enter the aquatic food web via a number of pathways, including directly from the sediment to benthic organisms (Desy et al. 2000; Pennuto et al. 2005; Ridal et al. 2010), and directly from the water column to phytoplankton and zooplankton (Pickhardt et al. 2005; Stewart et al. 2008), after which it can be biomagnified as it moves up the food web, reaching potentially harmful levels in higher trophic level aquatic and terrestrial organisms (Watras et al. 1998; Kim and Rochford 2008; Syaripuddin et al. 2014).

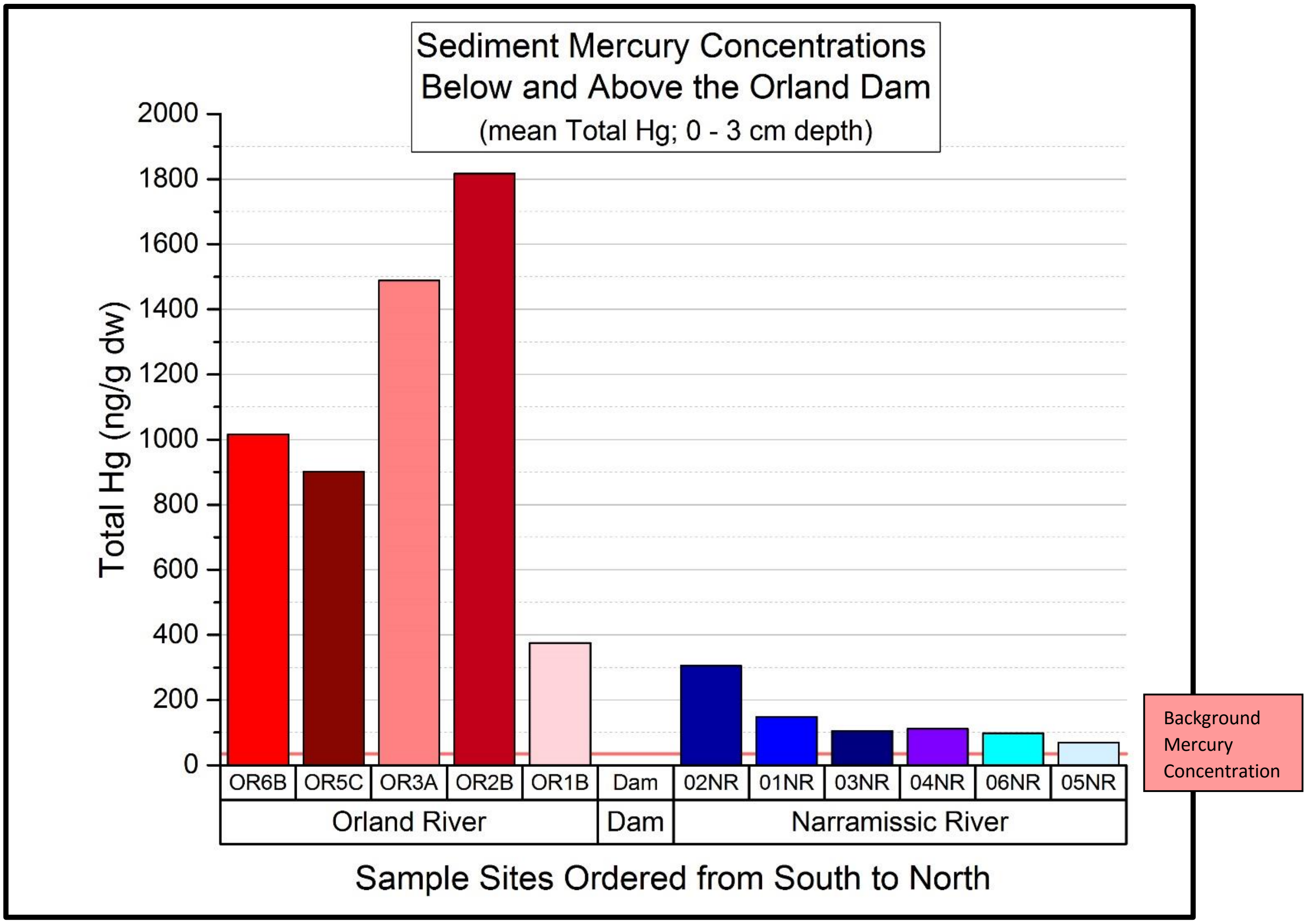

Figure 5. Total mercury concentrations in surface sediments in the Orland River below the Orland Dam and in the Narramissic River above the dam. Note that site OR1B is in the cove west of Fish Point, outside of the main channel of the Orland River. The pink band between 28 and 38 ng/g dw represents background sediment mercury concentrations found in coastal Maine rivers.

Methyl mercury concentrations are often directly linked to the amount of total mercury in the sediment, as was found in the Penobscot River (Bodaly et al. 2013).

It is important to look at sediment mercury concentrations in relation to toxicity to benthic (sediment dwelling) organisms and to the fish, birds and other upper trophic level organisms that forage on benthic organisms. There are no official criteria, or clean-up levels, for mercury concentrations in sediment. However, NOAA has issued preliminary screening values which in the absence of site-specific toxicity tests may be used to estimate potential impacts associated with specific concentrations, as outlined below.

The Threshold Effect Concentration (TEC) for total mercury was developed from a meta-analysis of the TELs (Threshold Effect Level) with the Lowest and Minimal Effect Levels and is the concentration below which toxicity to benthic organisms is rarely expected. Similarly, the Probable Effect Concentration (PEC) was developed from a meta-analysis of the PELs (Probable Effect Level), the Severe Effect Levels and the Toxic Effect Thresholds and represents the level above which toxicity to benthic organisms is expected to occur frequently. TECs and PECs combine the results from both freshwater and marine sediments. The individual TELs and PELs for mercury in freshwater and marine sediments are also given in Table 1 (Buchman 2008, MacDonald et al. 2000, MacDonald et al. 1996).

Table 1. NOAA screening values for total mercury in sediment.

NOAA Screening Value	Total Mercury ng/g dw	Description
TEC - Threshold Effect Concentration	180	Below which toxicity rarely expected
PEC - Probable Effect Concentration	1,060	Above which toxicity frequently expected
FRESHWATER		
TEL - Threshold Effect Level	174	
PEL - Probable Effect Level	486	
MARINE		
TEL - Threshold Effect Level	130	
PEL - Probable Effect Level	700	

The potential for contamination of fish or birds, upper trophic level animals that consume benthic organisms, is not addressed by these screening values. The sediment dwelling organisms assayed for the screening values included amphipods, mayflies, midges, oligochaetes, and daphnids. Given that mercury is biomagnified in the food web, the usefulness of these screening values is limited to benthic toxicity.

In the Narramissic River, the mercury concentrations in the top three centimeters of sediment are below the TEC except at the site closest to the Orland Dam, 02NR, which had a mean mercury concentration of 305 ng/g dw. As the mercury concentration at that site is above the TEC, but below the PEC, toxicity to benthic organisms is not expected, but remains a possibility.

In contrast, in the Orland River downstream of the dam, mercury concentrations exceeded the PEC at two sites and exceeded the PEL for marine sediment at all sites except at OR1B (Figure 5). Site OR1B is located in the cove west of Fish Point, outside of the main channel of the Orland River. If tidal flows are restored to the Narramissic it is likely that some portion of the toxic sediments currently in the Orland River will be carried by incoming tides into the Narramissic. More information is needed on the range of the tidal influence predicted for the Narramissic River before the geographic extent and the change in mercury concentrations can be estimated.

7. **The findings reported here describe mercury concentrations in the surface sediments of the river but provide no information on the accumulation of mercury in the sediment below 3 cm, or on the time period in which the mercury was deposited.**

Information on mercury in the surface sediments of the Narramissic is valuable for defining recent mercury inputs, but leaves other questions unanswered. The current data give no information on the time period during which mercury was deposited in the surface sediments.

Nor do they indicate the concentration of mercury at depth, which could be greater than found at the surface, if dam breaches and the tidal movement of contaminated sediment upstream occurred during past periods of peak mercury discharge.

As there are no data on the sedimentation rates in the Narramissic River it is not possible to determine the time period during which mercury was deposited in the surface sediments. Rough estimates can be made using sedimentation rates reported for other areas of the Penobscot system. Yeager (2013) and Santschi (2013) reported the average sedimentation rate in the tidal Mendall Marsh, near Franklin, as 0.61 cm/year, with a range of 0.11 to 1.85 cm/year. If it is assumed that the sedimentation rate in the Narramissic River is similar to the average rate found in Mendall Marsh, mercury concentrations in the top three centimeters of sediment result from depositions occurring within the last five years. However, given the range of sedimentation rates reported for Mendall Marsh, the mercury could have been deposited between 2 and 27 years ago.

Deep core samples, in which a column of sediment up to 90 cm long is collected and analyzed in sequential layers, would provide additional answers. Core data would determine the concentration of mercury at different depths and, using radionuclide analyses, the time period during which the mercury was deposited. The core data would show whether there has been a fairly constant mercury input over the last 50 years or whether pulses of mercury moved upstream of the dam during discrete time periods, such as the period of deterioration prior to the 1985 repairs of the Orland Dam. Further, the total amount of mercury stored in the sediments of the Narramissic could be calculated using the core data. The possible removal of the Orland Dam would undoubtedly result in the mixing and resuspension of sediment in the Narramissic River below 3 cm depth, making this information important to the decision on whether to remove or alter the dam at this time.

CONCLUSIONS

- Mercury concentrations found in the surface sediments of the Narramissic River exceed regional background concentrations but are lower than concentrations associated with benthic toxicity.
- The decline in mercury concentrations with distance from the Orland Dam suggests that the mercury source is contaminated sediment from the Orland River originating from the HoltraChem plant in Orrington.
- Further research is needed to determine the extent to which removal or alteration of the Orland Dam will increase the tidal flow of water and the associated movement of contaminated sediment from the Orland River into the Narramissic River.

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APPENDIX 1. Summary statistics for each of the 12 sample sites surveyed in this study. The Narramissic River Sites are ordered from south (near the Orland Dam) to north (near the Upper Falls Road Bridge). The marsh sites are ordered from the water's edge inland to the marsh platform.

Sample Area	Site ID	Total Hg ng/g dw mean ± SD (min-max)	TOC mg/g dw mean ± SD (min-max)	% Fines (grain size <75µm) mean ± SD (min-max)	Water Depth (m)	Latitude	Longitude
Narramissic River	02NR	305 ±28 (274-328)	587±133 (440-700)	38±9 (27.5-45.3)	0.50	44 34.273	68 44.637
	01NR	148 ± 17 (131-165)	307 ±150 (220-480)	43±8 (35.2-50.2)	2.25	44 34.381	68 44.607
	03NR	105 ± 5 (101-111)	483±72 (400-530)	52±10 (43.9-62.9)	2.25	44 34.668	68 44.631
	04NR	112 ± 17 (97-130)	467±47 (430-520)	53±4 (49.1-56.0)	4.10	44 34.668	68 44.631
	06NR	98 ± 6 (93-105)	480±46 (430-520)	49±7 (41.5-55.8)	1.00	44 35.342	68 44.170
	05NR	69 ± 12 (57-81)	493±233 (240-700)	45±5 (40-51)	1.25	44 35.645	68 43.654
Duck Cove Marsh	10DC	66 ± 11 (54-75)	616±506 (300-1,200)	35±6 (29-40)	0.50	44 34.944	68 44.589
	11DC	90 ± 6 (83-92)	790±184 (660-1,000)	32±13 (18-44)	0.10	44 34.948	68 44.585
	12DC	115 ± 7 (109-123)	1,667±737 (1,100-2,500)	26±5 (20-30)	0.05	44 34.955	68 44.575
Wight's Brook Marsh	07WB	71 ± 17 (59-90)	1,500±435 (1,200-2,000)	40±8 (33-48)	0.50	44 35.382	68 44.091
	08WB	94 ± 0.6 (93-94)	1,933±1,242 (500-2,700)	35±27 (14-65)	0.05	44 35.401	68 44.807
	09WB	93 ± 12 (81-104)	1,887±1,120 (760-3,000)	18±12 (4-25)	0.10	44 35.417	68 44.079