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Stream Mercury Export in Response to Contemporary Timber Harvesting Methods (Pacific Coastal Mountains, Oregon, USA)

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Abstract

Land-use activities can alter hydrological and biogeochemical processes that can affect the fate, transformation, and transport of mercury (Hg). Previous studies in boreal forests have shown that forestry operations can have profound but variable effects on Hg export and methylmercury (MeHg) formation. The Pacific Northwest is an important timber producing region that receives large atmospheric Hg loads, but the impact of forest harvesting on Hg mobilization has not been directly studied and was the focus of our investigation. Stream discharge was measured continuously, and Hg and MeHg concentrations were measured monthly for 1.5 years following logging in three paired harvested and unharvested (control) catchments. There was no significant difference in particulate-bound Hg concentrations or loads in the harvested and unharvested catchments which may have resulted from forestry practices aimed at minimizing erosion. However, the harvested catchments had significantly higher discharge (32%), filtered Hg concentrations (28%), filtered Hg loads (80%), and dissolved organic carbon (DOC) loads (40%) compared to forested catchments. MeHg concentrations were low (mostly $<0.05 \text{ ng L}^{-1}$) in

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ASSOCIATED CONTENT

Supporting Information

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Text describing the site conditions, field measurements, blank and replicate analysis, inter-lab comparison, and statistical analysis, plus 6 figures showing a map of downstream sample locations, the results of THg-F interlab comparison, soil and sediment organic matter and THg concentrations, the exceedance probability curve, and the results of the downstream synoptic sampling (PDF)

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harvested, unharvested, and downstream samples due to well-drained/unsaturated soil conditions and steep slopes with high energy eroding stream channels that were not conducive to the development of anoxic conditions that support methylation. These results have important implications for the role forestry operations have in affecting catchment retention and export of Hg pollution.

Graphical Abstract

1. INTRODUCTION

Atmospheric long-range transport and subsequent deposition of mercury (Hg) have resulted in the global distribution of Hg contamination.^{1–3} In watersheds, deposited Hg is influenced by a wide range of hydrological and biogeochemical processes occurring within the landscape.⁴ Of particular concern are processes that promote the mobilization of Hg to area waterways and its transformation to the more toxic and bioaccumulative form, methylmercury (MeHg), by anaerobic bacteria.⁵

Most of the deposited Hg within forested watersheds is retained by vegetation and soils, with only a relatively small proportion mobilized in runoff⁶ or re-emitted.⁷ As a result, watershed yields (the ratio of Hg in runoff to the amount deposited) in forested landscapes are typically low and range from <0.5% to 10%.^{8,9} Because of Hg retention in catchments, soil Hg concentrations have increased over time.^{10,11} Much of the Hg retained in forested areas is associated with organic matter (OM),^{12–14} and the Hg concentrations in forested streamwater are typically highly correlated with the stream's dissolved organic carbon (DOC)^{15–17} concentrations. As such, the timing and extent of Hg sequestration versus mobilization in forested catchments is affected by hydrologic processes governing flow paths as well as biogeochemical processes influencing the formation of particulate carbon and DOC.¹⁸ Landscape alterations, such as forestry operations, affect both catchment hydrology and carbon processing,^{19–21} which can impact the cycling of Hg.^{22,23}

Timber harvest can have a profound but variable effect on Hg cycling. For example, minor or no changes in postharvest stream total-Hg (THg) concentrations have been reported in some studies;^{24–27} whereas others have demonstrated increases of up to 55%.^{28–30}

Responses in MeHg concentrations associated with forestry operations have also been similarly variable, with different studies showing decreases, no changes, or increases in MeHg concentrations.^{24–29,31,32} In the studies where increases in MeHg were observed, the relative magnitude of response was larger (up to 250% increases) than that observed for THg.^{24,28} Despite the variability in THg and MeHg concentrations due to timber harvest, calculated THg and MeHg stream loads typically increase after timber harvest (even when there are no changes in concentrations) by up to 450%, largely because of forestry-associated increases in stream discharge.^{25–29} Studies in which both MeHg concentrations and loads increase indicate a likely increase in methylation rates, which can be spurred by some combination of enhanced processing of labile carbon from the degradation of logging residue and soil organic matter²⁴ or enhanced water saturation of soils following clear-cutting.^{31,32} The impact of forestry operations on MeHg accumulation in downstream aquatic biota (zooplankton and fish) has also been observed in several studies.^{33–36} In addition to increases in aqueous fluxes, surface-air Hg fluxes have been shown to be higher in clear-cut areas compared to forests.³⁷ Thus, the global extent of timber harvest activities may be a land use that has a profound influence on global Hg cycling. Among the most important areas for timber production are the temperate forests along the Pacific coast of North America. There are more than 155,000 km² of harvestable forest land in the Pacific Northwest, which accounts for nearly 25% of the region's total land area.

The Pacific coastal region receives some of the highest loads of wet-deposited THg in North America, which is largely driven by the high amounts of precipitation.³⁸ Dry deposition of Hg also occurs and accounts for about a third of the total atmospheric deposition flux.⁹ This area is defined by mountainous topography with forestry operations often occurring on relatively steep slopes during snow-free periods. These conditions are very different from the existing literature on the impact of forestry operations on Hg mobilization to streams, which focuses entirely on northern boreal forests, mostly in Fennoscandia.^{22,23} In these studies, logging operations typically occurred in relatively flat areas and during periods of snow cover to lessen erosion and other soil impacts.^{26,27,29,39} Currently, there is a lack of information regarding the impacts of forestry operations in other major timber producing regions such as the Pacific Coastal region which have different topographical, vegetative, and climatic characteristics that can influence Hg cycling.⁴⁰ Additionally, different forest harvesting practices can have a large influence on the resulting environmental impacts.

Forestry practices have changed over time in response to environmental regulations, and they have shifted from old growth harvesting to logging younger trees in intensively managed forest lands. Assessing the effects of timber harvest on stream Hg dynamics in younger-harvest, regenerated forests that are logged using contemporary techniques will facilitate a better understanding of these processes that are representative of current conditions and scalable to the potential regional effects.

Best Management Practices (BMPs) in forestry are aimed at reducing the environmental impacts of harvest operations and can represent a wide range of measures such as leaving nonharvested buffer zones along streams, installing water diversion bars/ditches to reduce the impacts of road construction, harvesting during seasons where impacts would be reduced, and decreasing postharvest erosion by leaving woody debris on-site.^{41,42} While

previous research has been conducted on reducing the impacts of sediment loading and thermal impacts on streams from harvested catchments, there remains limited investigation into how contemporary forestry practices can impact Hg cycling and watershed transport.^{22,23}

In this study, we sought to evaluate how forestry operations in mountainous regions of the Pacific Northwest influenced Hg cycling. We specifically address two primary objectives: 1) test whether forestry operations increase THg and MeHg concentrations and loads in small headwater streams and 2) assess whether MeHg concentrations increase downstream from the headwater forestry operations as the stream/river conditions become more conducive to methylation. We address these objectives in the Trask River watershed in Oregon, USA, as part of a controlled research project conducted in eight paired watersheds.

2. METHODS

2.1. Study Location.

Our study was conducted in four headwater catchments of the east fork of the south fork of the Trask River which flows into the Pacific Ocean at Tillamook Bay, OR, USA (Figure 1). Additional information on the catchment characteristics is provided in the SI.

We sampled eight headwater streams within the Trask River Watershed Study area, as well as from several locations within the Trask River, to measure downstream conditions (Figure S1 in the Supporting Information–SI). The stream catchments were clustered within four subwatersheds, three of which were subjected to timber harvest (Pothole, Upper Main, and Gus Creeks) and one that served as a reference for cumulative effects without any harvest activity (Rock Creek). Within the clusters that had forest harvest, one headwater catchment was not harvested and served as a within-cluster paired control. Both of the streams within the Rock Creek reference catchment were unharvested.

The harvested and reference headwater catchments are characterized by steep slopes, with most stream section gradients ranging from 4 to 20%. The high gradient stream channels were mostly distinguished by step pools and cascade transport, and the stream beds were dominated by cobbles and boulders.

Forest harvest was conducted in spring and summer of 2012 at three of the catchments. No riparian buffer was left in Gus Creek harvest sites, whereas Upper Main and Pothole Creeks had buffer strips of remaining trees along the lower section of streams (4 and 8 m wide, respectively). The upslope areas of these three watersheds were clear-cut harvested using a combination of ground and cable logging. Existing gravel roads for harvest and hauling were upgraded prior to forest harvest, and the Gus Creek catchment had a new road constructed prior to harvest. Logging debris/slash which consists of branches, foliage tops, and other unmerchantable wood was left on-site following the logging operations in all of the catchments for the duration of the study period (2012–2014).

2.2. Field Measurements.

Flumes for measuring discharge were constructed on seven of the headwater streams, which included both harvested and unharvested streams in Gus, Main, and Pothole Creeks. Of the two reference Rock Creek sites, only one of them had a flume. Data loggers continuously recorded water depth and stream temperature. Grab samples for water quality analysis were taken during 15 sampling events occurring approximately monthly between October 2012 and December 2013 ($n = 135$ with duplicates). The first sampling event occurred approximately one month following the completion of the forest harvesting. Grab samples were collected into new 1 L PETG bottles by dipping the bottle into the water immediately upstream of the flumes following USGS Method 5.6.4.B.

Concurrent precipitation measurements were collected from six locations (all three of the paired forested and harvested catchments) using open wide mouth 500 mL PETG bottles that were fastened to stakes 1-m above the surface and deployed for ~24 h during a precipitation event on February 7, 2015 ($n = 6$). The three locations in the harvested areas measured open canopy precipitation, whereas the forested sites measured throughfall. Only THg concentrations were measured in precipitation in this study; however, the Mercury Deposition Network (MDN) OR01 site about 50 km away measured MeHg and showed average concentrations of $0.23 \pm 0.02 \text{ ng L}^{-1}$, which is about 3% of the THg concentrations.

Downstream synoptic sampling at 22 locations along the course of the Trask River, from the headwaters to the estuary at the Pacific Ocean (approximately 20 miles downstream), occurred during baseflow conditions between July 23 and 29, 2014. For this effort, we divided the Trask River into three general zones based on the river channel gradient: the steep gradient headwater/erosion zone; the midsection transport zone; and the low gradient downstream depositional zone. Additional details of the downstream synoptic sampling are provided in the SI text and Figure S1.

After collection, water samples were stored on ice in a dark cooler during transport back to the USGS Contaminant Ecology Research Laboratory (CERL) in Corvallis, OR (~2 h from the study area). Upon return, the samples were refrigerated prior to filtration which typically occurred the day following sample collection. On two occasions filtration occurred 2 days after collection (November 2013 and July 2014 sampling events). Sample filtration and sample preservation procedures were based on USGS Method 5.6.4.B and used $0.7 \mu\text{m}$ precombusted quartz fiber filters (550 °C for 4 h) for all filtered and particulate constituents.

Water samples were collected for the following parameters: particulate THg (THg-P), particulate-MeHg (MeHg-P), filtered THg (THg-F), filtered MeHg (MeHg-F), filtered sulfate, total suspended solids (TSS), dissolved organic carbon (DOC), and specific ultraviolet absorbance at 254 nm (SUVA_{254}). Filtered water samples for THg and MeHg analysis were stored in acid cleaned amber glass bottles that were filled and stored with dilute (0.1%) HCl. Filters for particulate analysis were stored in new plastic Petri dishes and frozen until analysis. Sulfate and DOC were collected into 40 mL amber glass vials, and TSS was collected onto 25 mm, $0.7 \mu\text{m}$ Whatman glass fiber filters. Duplicates and field blanks were collected at a frequency of one per field run for all water sample parameters.

The top ~2 cm of soil were collected from 113 locations throughout each of the forested ($n = 58$) and harvested ($n = 55$) catchments. If present, the overlying duff layer was removed, and a new plastic trowel was used to collect the soil sample, which was stored in double sealable plastic bags for transport to USGS CERL. At the laboratory, soils samples were sieved to <2 mm and frozen until analysis. Sediments were collected at 7 of the 8 flume locations (the exception being one of the Rock Creek reference sites). Surface sediment samples were collected using a new plastic hand scoop. Sediment samples were stored in new glass jars and stored frozen.

2.3. Laboratory Analysis.

Filtered THg and MeHg samples were analyzed at the US EPA Manchester Environmental Laboratory (EPA MEL) and/or the USGS Mercury Research Laboratory (MRL). Analysis of THg-F followed USEPA 1631E (Modified) using a Tekran 2600 with cold vapor atomic fluorescence spectroscopy (CVAFS), and MeHg-F followed US EPA 1630 (modified using a Brooks Rand MERX instrument (both methods modified for filter size/type and filtration time). Analysis of THg-P and MeHg-P followed USGS Techniques and Methods 5 A-8 and 5 A-7, respectively. Sulfate (US EPA Method 300.0, Dionex CS-1500–500 ion chromatograph), TSS (USGS Method I-3765), DOC (EPA Method 415.3, Shimadzu TOC-V), and SUVA (absorbance scan 200–800 nm, Varian-Cary 300) were analyzed at the USGS CERL. Field blanks and duplicate data are presented in the SI. An interlab comparison between the USGS and EPA laboratories of THg and MeHg samples was conducted and showed good agreement (full details of the interlab comparison are summarized in the SI text and Figure S2).

Sediment and soil samples were analyzed for THg by direct thermal combustion using a Nippon Model MA-3000 Mercury Analyzer following EPA method 7473 (QA/QC data available in the SI text). All sediment samples were also analyzed for loss-on-ignition (LOI) to estimate the organic matter content by weighing the sample before and after heating in a muffle furnace at 400 °C for 16 h.

2.4. Statistical Methods.

Uncensored data that were below the method reporting limit (THg-F $n = 25$ of 134 samples were <0.2 ng L⁻¹; THg-P $n = 2$ of 133 <0.02 ng L⁻¹) were used in the statistical analysis and loading model calculation. Annual THg and MeHg loads were calculated using LOADEST software,⁴³ with details of the analysis provided in the SI. The paired harvested and forested stream data from Upper Main, Gus, and Pothole Creeks were used to determine if there were significant differences on aqueous and particulate Hg concentrations and loads from harvested and reference catchments. Statgraphics Centurion software was used for the statistical analysis of the data. Stream THg and MeHg concentrations were compared between the paired clear-cut and forested catchments using mixed-effects generalized linear models (GLM). Details of the statistical analysis are included in the SI.

3. RESULTS AND DISCUSSION

3.1. Mercury Concentrations in Precipitation, Soil, and Sediment.

The THg-F concentrations in throughfall precipitation from the forested catchments were 2-fold higher than the concentrations in the open canopy harvested catchments (Figure 2A; t test $p = 0.03$, $df = 4$). A similar degree of enrichment under coniferous canopies has been observed in several other studies and highlights the important role vegetation plays in the accumulation and wash off of dry deposited Hg from foliage.⁴⁴ Our comparison of THg-F is an underestimation of the total difference in open canopy and throughfall precipitation concentrations since filtering the sample excluded wash off of dry deposited THg-P ($>0.7 \mu m$).

There was no significant difference in soil THg concentrations between the forested ($0.12 \pm 0.01 \mu g g^{-1}$) and harvested catchments ($0.12 \pm 0.01 \mu g g^{-1}$; GLM $p = 0.86$, $df = 99$). The soil THg concentrations in our study area are on the lower end of concentrations measured in Scandinavia (0.15 to $0.37 \mu g g^{-1}$) where other studies examining the effects of forestry operations on Hg mobilization have been conducted.^{45–47} Soil THg concentrations were positively correlated with soil organic matter ($R^2 = 0.35$, $p < 0.001$; Figure S3 in the SI), thus the relatively high soil Hg concentrations in our study area (as well as those from the other Hg forestry studies in Scandinavia) are likely related to the high organic matter content (mean %LOI: $35 \pm 1.9\%$) of the soils effectively sequestering atmospherically deposited Hg.^{12,23} The similarity in soil Hg contents between forested and harvested catchments likely exists because measurements were obtained within the first few months to a year following harvest. However, over the ensuing decades, the soil THg concentrations in the harvested catchments may decrease as Hg is mobilized to the air and water.⁴⁷ A significant effect of the forest harvesting on the soil organic matter content was observed (%LOI harvested: $28 \pm 2.5\%$; forested: $35 \pm 2.2\%$; t test on natural log transformed data, p -value = 0.02 , $df = 111$). Reductions in soil organic matter following harvesting could result from increased soil moisture, temperature, and/or mixing the forest floor with mineral soil, all of which lead to increased microbial organic matter decomposition.^{48–50}

Similar to soils, sediment THg concentrations were correlated with sediment %LOI ($R^2 = 0.81$, $p = 0.01$; Figure S4 in the SI) and were similar between the streams in the forested ($29.4 \pm 2.3 ng g^{-1}$) and the harvested catchments ($34.0 \pm 1.8 ng g^{-1}$; t test $p = 0.20$, $df = 5$). The organic matter content of the stream sediments between the forested and harvested catchments were similar, both having mean LOI values of 12% (t test $p = 0.99$, $df = 5$).

Identifying the unique impact from forest harvesting on Hg mobilization in streams is facilitated by having similar soil and sediment THg concentrations between the forested and harvested catchments in this study.

3.2. Forest Harvesting Impacts on Filtered Mercury, DOC, and Sulfate.

The collection of field samples was highly representative of the full range of annual hydrological conditions encountered in the study's streams (Figure S5 in the SI).

All of the THg water concentrations measured in forested and harvested headwater catchment streams were low (overall mean $\pm$ standard error (SE) THg-F: $0.40 \pm 0.03 \text{ ng L}^{-1}$; THg-P: $0.20 \pm 0.02 \text{ ng L}^{-1}$ $n = 119$) compared to streams measured in other parts of the US (mean $\pm$ SE THg-W: $3.0 \pm 0.33 \text{ ng L}^{-1}$, $n = 250$)⁵¹ and roughly an order of magnitude lower compared to levels found in Scandinavian forestry studies.^{24,26–28,30,39}

In both the forested and harvested catchments, the majority ($70 \pm 17\%$) of the THg in stream samples was in the filter passing phase and was significantly correlated with DOC concentrations ($R^2 = 0.39$ and 0.54 , $p < 0.001$; Figure 3A). The slopes between THg-F and DOC did not differ between harvested and forested catchments (interaction effect p-value: 0.95). However, the THg-F concentrations were significantly higher in the harvested catchments ($0.37 \pm 0.02 \text{ ng L}^{-1}$) compared to the forested catchments ($0.29 \pm 0.01 \text{ ng L}^{-1}$; $p = 0.004$, $df = 89$; Figure 2B and 3A), representing a 28% increase in THg-F concentration from the harvested catchments in comparison to unharvested catchments. Analysis of just the harvested catchments showed that the highest LSM THg-F concentrations were from the stream with no riparian buffer (Gus Creek: $0.41 \pm 0.03 \text{ ng L}^{-1}$); however, they were not significantly different from the clear-cut catchments with riparian buffers (Pothole and Main: $0.35 \pm 0.02 \text{ ng L}^{-1}$).

Whereas THg-F concentrations were higher in harvested catchments than in paired forested catchments (Gus, Main, and Pothole Creeks), we did not find a difference in THg-F concentrations between those harvested catchments and the two separate, unharvested reference catchments (Rock Creek 1 and 3; GLM $p = 0.53$, $df = 73$, harvested: $0.38 \pm 0.02 \text{ ng L}^{-1}$; Rock Creek: $0.40 \pm 0.03 \text{ ng L}^{-1}$). This indicates that despite the 28% differences in THg-F concentrations associated with timber harvest between paired catchments, the magnitude of the response was still within the natural range of stream THg concentrations for this region.

The DOC concentrations between the harvested and forested catchments were similar (GLM forested: $0.63 \pm 0.04 \text{ mg L}^{-1}$; harvested $0.58 \pm 0.04 \text{ mg L}^{-1}$; $p = 0.38$ $df = 89$), suggesting that the DOC mobilized from the harvested catchments was more Hg enriched. SUVA₂₅₄ was also correlated with THg-F and also did not differ between forested and harvested catchments (GLM $p = 0.18$, $df = 89$). The ratio of THg-F to DOC was 25% higher in the harvested catchments than the forested catchments (GLM $p < 0.001$, $df = 89$). Sources of Hg-enriched DOC from the harvested catchments could include Hg mobilized from increased degradation of soil organic matter, changes in root zone chemistry/root exudates in harvested areas, and/or changes in hydrological flow paths that mobilize new pools of Hg in the soil horizon due to changes in soil moisture associated with decreased evapotranspiration.²³ Also, the lower amount of THg-F per amount of carbon in the forested sites may result from a dilution effect⁵² due to the significantly higher soil organic carbon content in the forested catchments compared to the harvested ones. The important role that carbon mobilization plays in promoting Hg transport in response to forest harvesting is highlighted by the lack of an increase in non-DOC bound constituents, such as sulfate, which were lower in the harvested catchment (forested $4.3 \pm 0.2 \text{ mg L}^{-1}$; harvested $2.1 \pm 0.2 \text{ mg L}^{-1}$, $p < 0.001$, $df = 42$).

The ratio of mean soil Hg concentrations from each catchment (in ng kg^{-1}) to the stream THg-F (ng m^{-3}) concentrations can be used to calculate a distribution coefficient ($\log K_d$) which represents the preference for Hg to be bound to the soil matrix versus mobile in runoff. The $\log K_d$ values for the forested sites (2.60 ± 0.03) were significantly higher than the values from the harvested catchment (2.46 ± 0.03 ; t test $p < 0.001$, $\text{df} = 88$) suggesting that the soils of the harvested sites have lower sorption capacity and a greater ability to mobilize Hg. The $\log K_d$ value is related to the soil organic carbon density and the DOC concentration in the aqueous phase, both of which have binding sites for Hg. While such calculations are typically comparing intact soil-porewater Hg relationships,^{52,53} the use of dissolved phase stream concentrations in our study instead of porewater can be used to highlight the effect that forest harvesting has on mobilizing Hg from the catchment.

Filtered MeHg concentrations were routinely low in all streams, with 97% of the measurements being below the 0.05 ng L^{-1} method reporting limit. While some studies have shown that logging can increase MeHg concentrations in streams;^{24,28} the conditions in those studies were very different from our catchments. For example, the factors that can effect MeHg production such as the THg and organic carbon concentrations were higher in the Scandinavian studies on forestry.²⁴ In addition, and perhaps most importantly, the physical attributes of the Trask catchments were not favorable for the conditions associated with Hg methylation. Specifically, the catchments all had steep slopes with high energy eroding stream channels with minimal fine-sediment that would not be conducive to anoxic conditions developing. In addition, the water table was not near the soil surface, resulting in well-drained, unsaturated soil conditions, which would also limit the development of anoxic conditions necessary for MeHg production. These catchment conditions are distinct from those where previous studies on the impacts of forestry on Hg cycling have been conducted which are typically low gradient areas containing peatlands and/or wetland areas.^{24,28} Our study area is representative of commonly encountered conditions where logging activity occurs in western North America, which is often associated with mountainous topography.

3.3. Forest Harvesting Impacts on Particulate Mercury Concentrations.

There were no significant differences between forested and harvested catchments in THg-P concentrations (GLM $0.13 \pm 0.02 \text{ ng L}^{-1}$; $p = 0.78$, $\text{df} = 77$; Figure 2C), TSS concentrations (GLM $1.1 \pm 0.3 \text{ mg L}^{-1}$; $p = 0.93$, $\text{df} = 77$), nor Hg concentration of the suspended particles (GLM $0.12 \pm 0.02 \text{ } \mu\text{g g}^{-1}$; $p = 0.87$, $\text{df} = 77$), which were the same as the soil THg concentrations in the catchments. However, there was a significant difference in the slopes of THg-P and TSS between forested and harvested catchments ($p < 0.007$; Figure 3B), indicating that that relationship between THg-P and TSS differed between harvested and forested treatments. Specifically, there was a significant positive relationship between TSS and THg-P in forested sites, which is commonly observed in other stream and river systems,¹⁵ whereas no relationship existed between the two variables in the harvested catchments. This was largely driven by a handful of high TSS measurements that were not associated with high THg-P concentrations, which may have occurred from the mobilization of lower Hg content mineral soil and/or dead plant material in the harvested catchments. Particulate MeHg concentrations in both forested and harvested catchments were low, with all concentrations $< 0.04 \text{ ng L}^{-1}$.

The lack of increase in stream particulate THg and MeHg or TSS concentrations in clear-cut catchments is in contrast to what has been observed in other studies, where logging has been shown to increase catchment erosion and suspended sediment concentrations.^{41,54} The absence of a response of these parameters in our study may result from the forestry BMPs that were used to reduce erosion, such as limiting new road construction, effective use of road prism drainage techniques, using cable logging where the felled trees are moved across the site suspended by a cable, and the practice of leaving a thick layer of slash materials covering the soil after harvesting.^{42,55} These BMPs are widely utilized as part of forestry operations in the US and elsewhere; however, it is also not uncommon for slash to be removed or burned on-site. The removal of woody biomass, including stump harvesting, has gained interest in recent years for use as a renewable fuel or other products.^{56,57} As such, the results from our study are believed to be representative of commonly applied contemporary forestry techniques; however, forestry operations not utilizing these BMPs may have larger influence on Hg mobilization and other water quality end points.

3.4. Downstream Total and Methylmercury Concentrations: Headwaters to Estuary Assessment.

Synoptic sampling from the headwater catchments to the estuary 45 km downstream showed that THg-F, THg-P, and MeHg-P concentrations all significantly decreased with distance downstream (Figure S6 in the SI; THg-F $R^2 = 0.27$, $p = 0.02$; THg-P $R^2 = 0.28$, $p = 0.01$; MeHg-P $R^2 = 0.54$, $p < 0.001$). The MeHg-F concentrations in all samples were less than the 0.05 ng L⁻¹ reporting limit. The %MeHg in streams is often 5%;⁵¹ therefore given the low THg-F concentrations in this watershed (mean $\pm$ SE: 1.0 $\pm$ 0.1 ng L⁻¹), it is not surprising that the MeHg-F concentrations were routinely below the method reporting limit. Other research conducted on streams near our study site (Lookout Creek, OR) has also shown that MeHg-F and MeHg-P concentrations were routinely below the reporting limit.⁵⁸

Discharge increases from the small headwater streams (3 L s⁻¹) to the Trask River downstream by over 3 orders of magnitude (>3000 L s⁻¹). The decreasing THg-F, THg-P, and MeHg-P concentrations with increasing discharge downstream suggests that the upstream portions of the watershed where forestry operations are occurring may act as a source for these Hg species, which becomes diluted with distance downstream.

3.5. Annual Mercury Loads in Headwater Streams and Influential Variables.

The annual mean discharge from the harvested catchments was 32% higher than from the paired forested catchments when normalized to catchment area (LSM harvested: 1.04 $\pm$ 0.03 m³/m²; forested: 0.79 $\pm$ 0.03 m³/m²; $p < 0.001$; df = 4379; Figure 4). The difference in discharge between harvested and forested catchments can be larger during the periods of elevated discharge associated with heavy rainfall and/or snowmelt. For example, comparing just the days when the highest 5% of discharge was measured showed a 2-fold difference in discharge between the harvested and forested catchments (LSM harvested: 0.015 $\pm$ 0.002 m³/m²; forested: 0.007 $\pm$ 0.001 m³/m²; $p < 0.001$, df = 209). Our results showing higher discharge in harvested catchments does not take into consideration potential differences in hydrology that existed between these catchments prior to harvesting.

Increased stream discharge from clear-cut areas typically occurs through a reduction in vegetation interception and transpiration, as well as modified snow accumulation and melt rates. Some forestry operations can also result in reduced soil infiltration due to compaction from driving equipment on the site and/or yarding; however, this affect was likely minimal in our study area since felled logs were mostly moved via suspended cables and ground yarding only occurred in some lower gradient areas.

Annual THg-F loads (normalized by catchment area) were 80% higher in the harvested catchments compared to the forested catchments (LSM harvested: $0.52 \pm 0.03 \mu\text{g Hg m}^{-2}$; forested: $0.29 \pm 0.02 \mu\text{g Hg m}^{-2}$; $p < 0.001$, $\text{df} = 143$; Figure 5A). Although THg-F concentrations were higher in the harvested catchments, the higher loads were also influenced by higher stream discharge in the harvested catchments compared to the forested catchments. The differences in loads between the forested and harvested streams showed large seasonality, with the largest differences between the catchments occurring during the fall when the THg-F loads from the harvested catchments were 4-fold higher than the forested catchments. Fall is a period of heavy rainfall for this region, and this period of higher discharge appears to facilitate the larger loads from the harvested catchments compared to the forested ones. Conversely, during the dry summer months when the streams were dominated by low/base flow conditions the difference between forested and harvested catchment loads was small (Figure 5A).

The annual DOC loads were 47% higher in the harvested catchments compared to the forested catchments (harvested: $0.96 \pm 0.08 \text{ g m}^{-2}$; forested: $0.65 \pm 0.6 \text{ g m}^{-2}$; $p < 0.002$, $\text{df} = 143$; Figure 5C). This higher export of DOC from the harvested catchments was likely caused by increased organic matter mineralization in response to harvesting and appears to be an important driver resulting in the elevated THg-F loads from the harvested catchments. Without an increase in DOC mobilization, the higher discharge in the harvested catchments could have resulted in a dilution of the DOC concentrations. However, the similar DOC concentrations among streams combined with the increased discharge from the harvested catchments resulted in the larger loads.

Annual THg-P loads were not significantly higher from harvested catchments than forested catchments (harvested: $0.19 \pm 0.02 \mu\text{g Hg m}^{-2}$; forested: $0.17 \pm 0.01 \mu\text{g Hg m}^{-2}$; $p = 0.27$, $\text{df} = 143$; Figure 5B). During the summer months, the THg-P loads between the forested and harvested catchments were very similar but diverged during the periods of higher discharge in the fall and winter. These results suggest that the impact of logging on THg-P mobilization mainly occurs during periods of elevated discharge from rainfall and snowmelt when erosion and particle entrainment are higher.

Annual wet + dry deposition of THg to the study area is estimated to be $33 \mu\text{g Hg m}^{-2}$, with 67% occurring as wet deposition.⁹ The watershed THg yields (whole water runoff load/whole water wet deposition load) for the forested and harvested catchments were 4 and 6%, respectively. If including dry deposition, these yields (whole water runoff load/wet + dry deposition load) are a bit lower at 2 and 4% for forested and harvested catchments. These runoff yields are of similar magnitude as observed in forested catchments in other studies (1–10%^{8,9}). As such, while forest harvesting results in significantly higher THg loads, the

clear-cut catchments are still a large net-sink for atmospheric Hg—just a slightly less efficient one compared to the forested catchment.

The most dramatic impacts of forest harvesting on a catchment's hydrological and soil organic matter characteristics occur in the first few years following harvesting (e.g., 0–4 years), with the impacts decreasing as the forest is reestablished. In the longer term (5+ years), subtler impacts to soil and hydrological processes can also occur, though this is not well studied with regard to Hg dynamics.^{50,59,60} An exception is from a Norwegian study that found that in the first few years following harvesting Hg methylation increased as logging residue/slash were degraded; however, in older clear-cuts (4–8 years) THg and MeHg concentrations were indistinguishable from unharvested reference areas.²⁴

The precipitation, soil, and stream THg concentrations in our study area were all lower than the concentrations observed in previous forestry studies conducted in Scandinavia,^{45,46,61,62} which has been more impacted by historical and contemporary regional Hg emissions.⁶³ In addition, the impact of forestry operations on THg and MeHg concentrations and loads was less in our study than observed in several of the Scandinavian Hg forestry studies.^{24,28,30,32} The varying degree of impact on Hg cycling between studies is likely due to differences in both the geographic settings as well as forestry practices.

An important factor determining whether there is an increase in MeHg production appears to be tied to the degree to which forestry operations result in an increase in saturated soil and/or ponding water which can occur from increased infiltration raising the water table to the surface. Such hydrological changes would also impact surface and near-surface runoff dynamics and dissolved and particulate THg and MeHg mobilization. As such, the steep topography resulting in high water flow rates, and the deep depth to the water table likely contributed to the low MeHg concentrations in our study area. The potential for overland flow and particulate Hg mobilization was also reduced because the water table remained well below the surface.

In addition, forestry practices that occur during and following tree cutting, such as the amount of soil disturbance from machinery, road construction, postharvest site preparation techniques, and stump harvest/removal among others, are all expected to have a large influence on catchment THg and MeHg dynamics. The limited new road construction and soil disturbance from machinery combined with the thick slash layer covering the surface may have contributed to the relatively small increase in Hg concentrations from our harvested catchments compared to some other Hg forestry studies.^{24,27,28,30,32,39}

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Figure 1.

Map showing the study area with its eight paired catchments. The forested catchments included those from Rock Creek (RK1 and RK3), Pothole 3 (PH3), Gus Creek 1 (GS1), and Upper Main 1 (UM1). The harvested catchments included Pothole 4 (PH4), Gus Creek 3 (GS3), and Upper Main 2 (UM2).

Figure 2.

Graph A: Comparison of mean $\pm$ SE filtered total mercury (THg-F) concentrations measured in precipitation during a rain event on February 7, 2015 from the three paired forested and harvested catchments. Graph B: Least squares mean (LSM) $\pm$ SE THg-F concentrations (derived from general linear model estimates) from the three paired forested and harvested catchment streams. The LSM values are associated with the covariate DOC mean concentration of 0.60 mg L^{-1} . Note: the LSM THg-F concentrations from the forested catchment streams contained 10 values that were below the 0.2 ng L^{-1} reporting limit, and the harvested catchment streams contained 1 value below this level. The uncensored values below the reporting limit were included in the analysis that generated the LSM concentrations. Graph C: Results of GLM analysis showing LSM $\pm$ SE THg-P concentrations from the three paired forested and harvested catchment streams. The LSM values are associated with the covariate TSS mean concentration of 1.13 mg L^{-1} .

Figure 3.

Panel A shows that regression of filtered total mercury (THg-F) versus dissolved organic carbon (DOC) was significantly different (determined by GLM analysis) in the paired forested and harvested catchments. Panel B shows the regression of particulate total mercury (THg-P) versus total suspended solids (TSS) for all paired forested and harvested catchments. There was a significant positive relationship between THg-P and TSS for the forested catchments but not for the harvested catchments. Note: in both graphs, the axis values are plotted on a natural log scale.

Figure 4. Exceedance probability curves for discharge from the paired forested and harvested catchments. Each line represents the mean daily discharge of the three streams normalized for catchment unit, and the shaded gray area around the line represents the standard error of the mean.

Figure 5.

Mean $\pm$ standard error (SE) monthly loads for filtered total mercury (THg-F; graph A), particulate total mercury (THg-P; graph B), and dissolved organic carbon (DOC; graph C) for forested and harvested streams normalized for catchment areas. LOADEST software was used to calculate monthly loads over a 2-year time period for each of the six paired catchments. The two years of data were combined for each stream, and then the three

forested and three harvested streams loads were averaged. The SE values represent the variability between the three catchments (Gus, Pothole, and Main Creeks).