memorandum

Division of Health and Environment

Date: 2/10/2021

To: John DeArment, Clark Fork Coalition

cc: Elena Evans, Missoula Valley Water Quality District

From: Jamie Holmes, Jeff Morris, and Heather Forth, Abt Associates

Subject: Comments on the Smurfit-Stone/Frenchtown Mill Draft Baseline Ecological Risk Assessment

In November 2020, the U.S. Environmental Protection Agency (EPA) and SRC Inc. published the Draft Baseline Ecological Risk Assessment (BERA) for Operable Units (OUs) 2 and 3 of the Smurfit-Stone/Frenchtown Mill Site in Missoula County, Montana (U.S. EPA and SRC, 2020). On behalf of the Clark Fork Coalition (in cooperation with the Missoula Valley Water Quality District), Abt Associates (Abt) has reviewed the draft BERA. This memorandum provides our comments on the draft assessment.

Our focus in these comments is on the apparent weakness in the assessment of potential risk from dioxins and dioxin-like compounds. We have not commented on whether EPA has conducted the risk assessment appropriately according to the U.S. EPA (1997, 1998) guidelines.

Section 1 below provides a general comment on the data used to assess risk at this site. Section 2 provides more specific comments. References are provided in the last section.

1. General Comment

EPA has attempted to assess ecological risk using a paucity of data and many modeling substitutions and assumptions to address data gaps. Many of the sediment and tissue samples are composite samples, and those composites at times are further aggregated to create a single estimate of dose or exposure over hundreds of acres of waste ponds. Some data gaps were not addressed at all, which led to methods that strain credulity, such as estimating risk to bats based on contaminant concentrations in earthworms. Other assessments were done with convoluted calculations and assumptions, such as comparing dioxin data from composite rainbow trout fillets and carcasses to tissue residue benchmarks (TRBs) from 30-year-old studies of dioxins injected into fish eggs.

The lack of sufficient data and the attempts to estimate risk based on assumptions and models instead of actual data do not allow for a reasonable conclusion that this site poses little or no risk to biota. However, as will be discussed in the comments below, the existing data show that the site is a source of dioxins and dioxin-like compounds, there is a potential pathway for the compounds to reach the Clark Fork River, and the concentrations of those compounds in rainbow trout are higher downstream of the site than they are upstream. Concluding that the site is not a source of contaminants that pose a risk to biota in the Clark Fork River is potentially premature.

2. Specific Comments

2.1 Prioritization of Persistent Organic Pollutants

BERA Section 2.1, p. 3: *"Most of the pulp was used to produce unbleached linerboard, but a small fraction (about 6 percent) of the total pulp produced from 1960-1999 was used to create white linerboard or sold as bleached pulp."*

EPA and site contractors discount the amount of bleached pulp (a source of dioxins) produced at this site. If this "small fraction" of overall production was 6%, approximately 1,600,000 tons of bleached pulp were produced. This is a substantial quantity of a product that is a known source of dioxins and furans.

Chlorinated dioxins and furans, including 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD) and 2,3,7,8-tetrachlorodibenzofuran (TCDF), are persistent organic pollutants (POPs) that do not readily degrade in the environment and are highly toxic. In the late 1980s, EPA and the paper industry conducted a study incorporating sampling data from all 104 bleaching kraft paper mills in the United States (U.S. EPA, 1990), including the Frenchtown mill. In 1988, when the mill was sampled for the 104-Mill Study, TCDD was found in pulp, sludge, and effluent; and TCDF was found in pulp and sludge at the mill. The effluent discharged to the Clark Fork River and the sludges were placed in unlined waste ponds. In the problem formulation, EPA could have prioritized dioxins and furans as likely drivers of ecological risk, with more sensitive sampling methods and a different sampling design focused on assessing these contaminants.

2.2 Spatial Extent of the BERA

BERA Section 2.1, p. 4: *"OU3 includes locations in the CFR where hazardous substances from Site activities have come to be located, if any... For the purposes of this assessment, sampling conducted to date within the CFR adjacent to the Site and downstream of the Site is considered to represent potential areas where Site-related contamination may have deposited."*

The 104-Mill Study report (U.S. EPA, 1990) confirmed that dioxins were in the effluent in 1988. The mill released dioxins and furans into the Clark Fork River. These POPs may have come to be located in sediment deposition areas well downstream of the site, and they may still be biologically available at concentrations that harm biota. EPA has not assessed potential ecological risk in the Clark Fork River beyond the immediate vicinity of the site. POPs released when the mill was operating may have come to be located in depositional areas well downstream of the site.

At other contaminated sites, EPA requires the responsible parties to identify potential ecological risk downstream of the original release site. For example, the Clark Fork River Superfund complex upstream of Missoula extends from the original source areas to Milltown Dam, where contaminants from Butte and Anaconda came to be located, in a sediment deposition area over 100 miles downstream. Most of the contamination behind the dam was likely released decades earlier, but was still biologically available and required remediation. At the Frenchtown Mill site, EPA has not assessed downstream depositional areas where dioxin/furan releases from the mill may have come to be located.

2.3 Sources of Coplanar Polychlorinated Biphenyls

Section 2.4.2, p. 9: "PCBs were only detected in OU2 soils at specific locations, the high-density pulp tank (HDPT) and the transformer storage building (TSB) foundation areas. Aroclor 1260 was detected in the HDPT area, and Aroclors 1254 and 1260 were detected in the TSB area."

The conceptual model for polychlorinated biphenyls (PCBs) focused on Aroclors, such as those in oil leaking from transformers. Coplanar (dioxin-like) PCBs can also be generated from incomplete combustion in the presence of chlorine, similar to dioxins (Lemieux et al., 1999). These dioxin-like congeners have a similar mode of toxicity as dioxins and furans, and therefore are often included in a calculation of TCDD toxicity equivalence (TEQ). In addition, the detection limits for analyses of PCB congeners are orders of magnitude lower than the detection limits for PCBs in Aroclors.

The only samples of any media from the site that included PCB congeners were from the 2017 OU2 supplemental soil sampling. This study included PCB congeners from nine composite samples, where each composite covered from 10 to 20 acres. Coplanar PCBs, including PCB-126 (the most toxic), were detected in all nine samples. In four of the samples, PCB congeners comprised more than 50% of the TCDD TEQ, and in two samples, PCBs comprised nearly 80% of the TCDD TEQ. The data suggest that the site is a potential source of PCBs other than just Aroclors, and these coplanar PCBs should have been evaluated in other locations and other media.

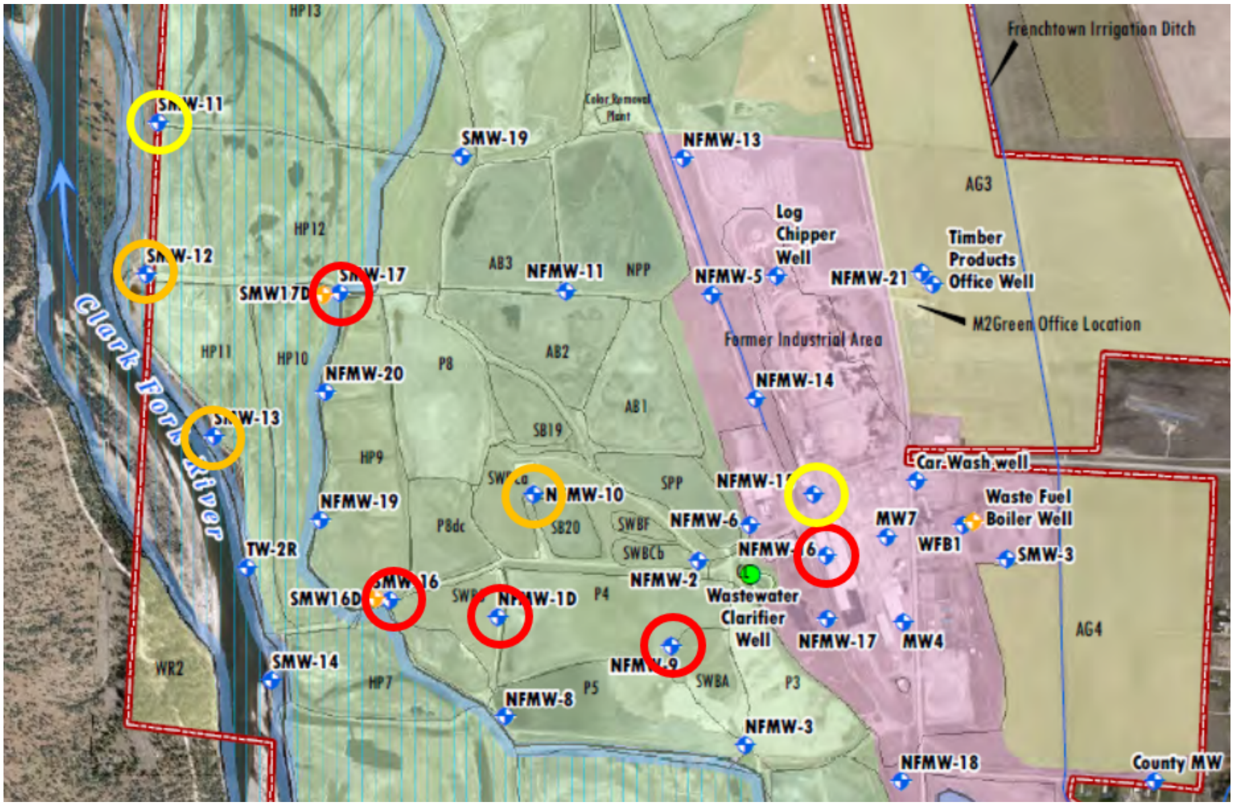
2.4 Transport of Dioxins/Furans in Surface Water and Groundwater

BERA Section 2.4.3, Fate and Transport, p. 10: "*Soil erosion and surface runoff can transport dioxins into surface water. Dioxins do not easily dissolve in water and therefore, most of the dioxins that enter surface water become strongly attached to particles that eventually settle in the sediment.*"

Despite the hydrophobic properties of dioxins and furans, several studies have found that these chemicals can be mobilized, most likely via colloidal transport (e.g., Hofmann and Wendelborn, 2007). The BERA does not fully evaluate a potential groundwater pathway to the Clark Fork River, despite detectable dioxins in groundwater samples. Figure 1 shows shallow groundwater wells with elevated TCDD TEQ, calculated with non-detectable (ND) dioxin and furan congeners set to a value of 0 pg/L. The wells with red circles have a maximum TCDD TEQ > 2 pg/L (which exceeds the Montana Circular DEQ-7 groundwater standard). Wells with a maximum TEQ > 1 pg/L have yellow circles, and wells with a maximum TEQ > 0.5 pg/L have orange circles. The spatial pattern suggests a groundwater pathway that extends from waste disposal sites in OU2 to the Clark Fork River.

The TEQ calculations in Figure 1 include only detectable dioxin and furan congeners. To date, there has been no assessment of coplanar PCBs in groundwater. As noted elsewhere, the site may be a source of coplanar PCBs in addition to dioxins and furans.

Figure 1. Shallow groundwater wells with elevated TCDD TEQ. Red circles have exceeded 2 pg/L, yellow circles have exceeded 1 pg/L, and orange circles have exceeded 0.5 pg/L.



Source: Modified from NewFields, 2017, Figure 3.

Dioxins and furans can be toxic at extremely minute concentrations and difficult to assess in water using standard methods because of their hydrophobicity. Groundwater standards are generally based on practical quantitation limits (PQLs) rather than low or no adverse effects levels. There is no TCDD TEQ surface water standard for the protection of aquatic life, but it is likely that it would be lower than the detection limits in the water samples collected at the site. For comparison, the surface water aquatic life criterion for TCDD TEQ based on risk to human consumption of fish and water is 0.005 pg/L, which is two orders of magnitude lower than typically achieved using standard methods.

As noted in Figure 1, even with standard sampling methods, the data suggest possible groundwater transport of dioxins/furans to the Clark Fork River, although the data are variable. At one site, a shallow groundwater sample contained 2.4 pg/L TCDD and a duplicate sample had no detectable TCDD at a detection level of 0.18 pg/L. This highlights the difficulty of capturing highly hydrophobic POPs using these standard sampling methods.

To reduce this variability and greatly reduce the detection limit, specialized sampling methods have been developed to assess highly toxic hydrophobic chemicals such as dioxins. The alternate methods generally require a large volume of potentially contaminated water to flow-through materials that attract lipophilic compounds, allowing those compounds to become concentrated to levels that are easier to detect. This can be accomplished by actively pumping large volumes of water through a device such as a resin column that will attract the lipophilic compounds, or passively placing a device in flowing water and leaving it for a prolonged period of time.

These methods have been used for many years at other sites with known dioxin issues. For example, Dinkins and Heath (1998) used high-volume water sampling to achieve detection limits below 0.001 pg/L in a dioxin-contaminated river in Ohio. For each water sample, 1,000 liters of water were pumped through a resin column. Charlestra et al. (2008) deployed semipermeable membrane devices into the Androscoggin River in Maine for 36 days to evaluate dioxin releases from a paper mill. Although there is more uncertainty in the estimate of the volume of water sampled using this method, Charlestra et al. (2008) also achieved detection limits close to 0.001 pg/L.

Both the Department of Defense (Lohmann, 2016) and U.S. EPA (2012) have developed guidance for assessing organic contaminants using passive sampling. The techniques have been used at numerous Superfund sites. Burgess et al. (2015) deployed three different passive sampling devices at three different Superfund sites, concluding that the devices generated scientifically accurate data that were critical for making informed management decisions at contaminated sites.

Passive sampling can also be used to assess the transport of hydrophobic compounds in groundwater. The Interstate Technology & Regulatory Council (ITRC, 2007) published protocols for using five different passive samplers in groundwater wells. The U.S. Geological Survey recently published additional guidance for assessing groundwater chemistry using passive sampling methods (Imbrigiotta and Harte, 2020).

Despite the hydrophobic nature of dioxin-like compounds, the existing data suggest a potential pathway to biota that has not been entirely evaluated. Prior to concluding that this site poses no risk from releases of POPs, EPA should consider using these existing technologies to assess potential offsite transport of highly toxic hydrophobic chemicals.

2.5 Assessment of Dioxins/Furans and Coplanar PCBs in Fish Tissues

EPA evaluated dioxins, furans, and coplanar PCBs in northern pike and rainbow trout tissues, and dioxins and furans in longnose dace. We believe the data are inconclusive. The following comments are not addressing specific sections in the BERA, but rather addressing issues with the assessment approach and conclusions.

2.5.1 Insufficient Data

The 2018 northern pike and rainbow trout data collection had data quality issues that relegated the data to the discussion of uncertainty in the BERA. The 2019 rainbow trout data included three sample sites upstream of the mill, one site in Frenchtown, and only one site downstream of the site at St. Regis. Northern pike were collected at two sites upstream of the site and in Frenchtown; no pike were collected downstream in St. Regis. This is a very limited dataset.

Dioxins may have been transported upstream from the site via stack emissions or biota moving upstream, but the vast majority of dioxins released to the Clark Fork River would have been transported downstream. This BERA does not include data to assess potential exposure and risk to POPs that have come to be located in downstream depositional areas.

Longnose dace were collected at several locations adjacent to and immediately downstream of the site. This spatial coverage likely does not cover the entirety of areas where dioxins have come to be located. In addition, coplanar PCBs were not analyzed in longnose dace. This is a data gap and likely underestimates TCDD TEQ in longnose dace.

2.5.2 TEQ Calculations

In the BERA, TCDD TEQ is calculated using one-half of the detection limit of undetected congeners. This is a standard practice and would typically be considered “conservative” (i.e., potentially overestimating risk) because it assumes that all congeners are actually present at concentrations below the detection limit, even if they are not. However, when many of the congeners are undetected and data quality issues result in inconsistent and at times elevated detection limits, this method can cause samples with almost no dioxins to appear to have TEQs that are as high as or higher than samples with lower detection limits and many detected congeners. The substitution of a single value (such as half the detection limit) for censored data can introduce false or misleading trends (Helsel, 2005). At this site, calculating TEQs using half the detection limit for undetected congeners can lead to artificially high TEQ calculations in samples from upstream of the site, which could mask a trend of increasing TEQ downstream of the site. We have evaluated TEQs with undetected congeners set to a value of 0 rather than half the detection limit in our analyses presented in these comments.

2.5.3 Sample Aggregation

The method used to assess risk to higher trophic-level biota from exposure to TCDD TEQ required many calculations and assumptions, with little underlying data to support any conclusions. Many of the samples collected at this site were composited, where multiple samples were mixed together, which implicitly assumes that concentrations are uniform in the individual samples, or that organisms will be exposed to different concentrations across a home range and the composite sample provides a better point estimate of exposure. Large areas of the site are represented by a single data point, which may not be representative of the potential exposure. All of the data from 11 ponds in OU3 covering hundreds of acres were digested down to a single exposure area using one assumed value for each contaminant. The existing data may not capture the areas of contamination and potential risk when such broad areas are represented by composited samples and aggregated data.

To assess risk to osprey from dioxins, the BERA relies on longnose dace data from adjacent to and immediately downstream of the site, and rainbow trout data from Frenchtown and St. Regis. All of the fish data were aggregated to create a single estimate of ingested dose. The data are sparse, and each step of aggregation increases the uncertainty. Rather than relying on these calculations and literature values, EPA could assess risk to osprey by collecting data from Clark Fork River ospreys.

2.5.4 TCDD TEQ in Fish Tissues

To assess risk to fish, the BERA relies on TRBs from Steevens et al. (2005). Much of the underlying data in Steevens et al. (2005) are 30-year-old studies where adverse effects were measured at different concentrations of TCDD in fish eggs, including multiple studies where TCDD was injected into the eggs, and studies where reported exposure concentrations were nominal (i.e., estimated based on dilutions of a stock solution) rather than measured. In the

BERA, TCDD TEQ concentrations in rainbow trout and northern pike were calculated from composite fillet and carcass samples of adult fish (one sample site for northern pike, and two for rainbow trout), and compared to benchmarks developed to show harm in eggs. This requires assuming that benchmarks in eggs and adult fish tissues are the same when normalized to lipid weight.

Lipid concentrations for rainbow trout (fillet with skin on) collected for the BERA range from 5.7% to 43.5%; 5 of the 20 reported values exceed 35% lipid. This is considerably higher than literature ranges of lipid values reported for multiple salmonids. We examined data from several studies (Niimi, 1983; Miller, 1993; Russell et al., 1999; Kandemir and Polat, 2007) and found lipids ranged from 0.7% to 26%, with an average of 8.1% lipid (in whole fish, skin-on fillet, and fillet). Eighteen of the 20 Clark Fork River rainbow trout fillet lipid values exceed 8.1%, which could confound the assumption that the TCDD TEQ concentrations in these fish can be compared to literature TRBs.

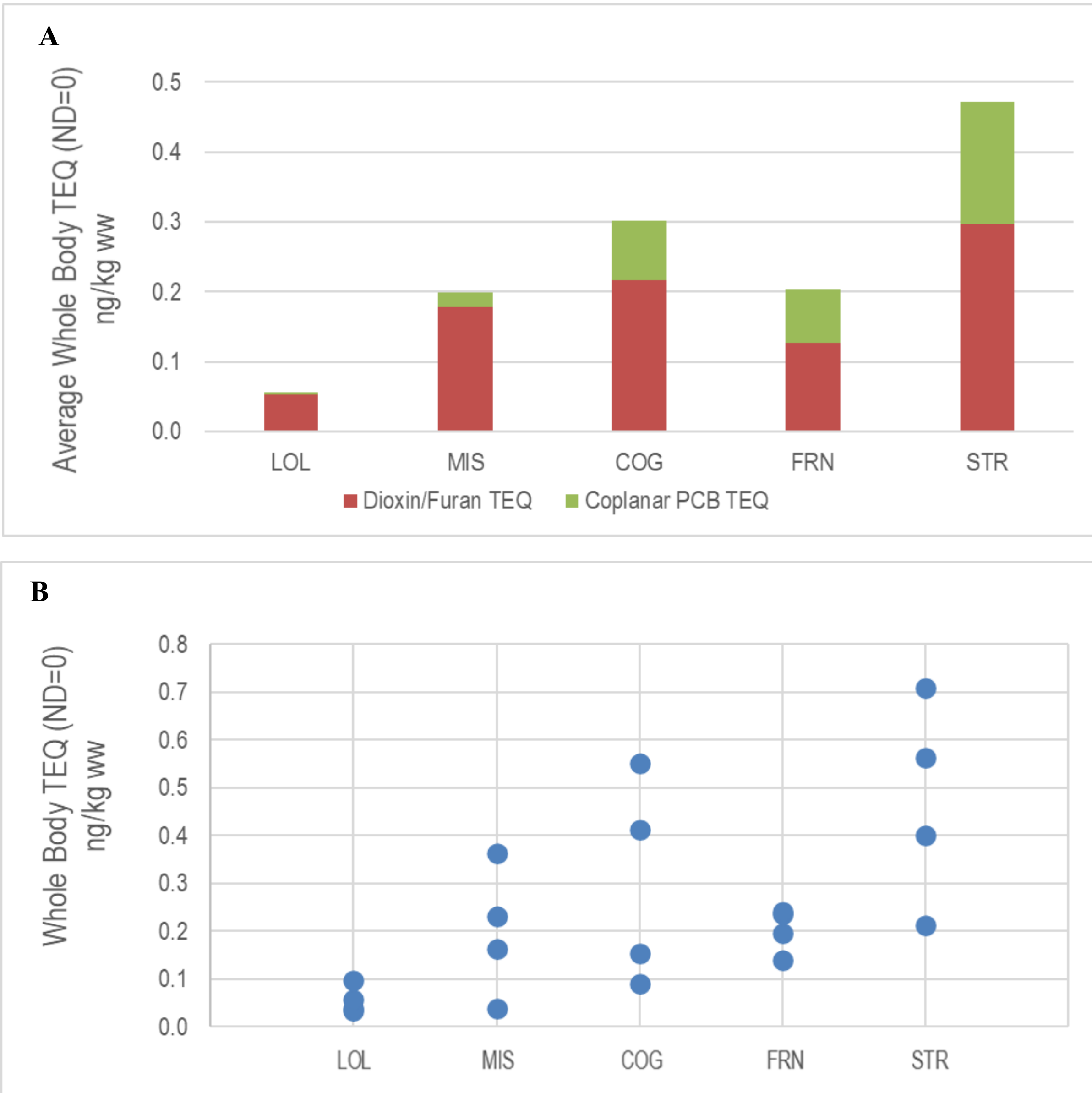
Furthermore, the Steevens et al. (2005) benchmarks are based on tissue residue effects levels in fish eggs, which are converted to "whole-fish" by Steevens et al. (2005) based on the 1:1 lipid-normalized chlorinated organic compound relationship reported by Russell et al. (1999). Though Steevens et al. (2005) describes these as adult "whole fish" concentrations, Russell et al. (1999) used adult dorsal muscle in their analyses, and they cited studies where "whole fish" included fish with eggs and gut contents removed (Niimi, 1983) or just skin-on fillets (Miller, 1993). Therefore, comparisons of lipid-normalized TEQ values derived from fish carcass samples may not represent a direct comparison to the Russell et al. (1999) analysis and subsequent benchmarks derived by Steevens et al. (2005). Additional risk thresholds should be evaluated and additional data collected to achieve a lower level of uncertainty.

There are other issues with the calculation of risks to fish in the BERA. Without belaboring the point, the assumptions and calculations required to compare the trout and pike data to fish egg residue benchmarks creates a level of uncertainty too high to allow for any conclusions about potential risk.

2.5.5 Spatial Trend of TCDD TEQ in Fish

We have not attempted to calculate risk to fish, but we have examined the spatial trend in fish tissues, using simpler analysis methods. For example, we examined average TCDD TEQ in rainbow trout ("whole body," using the method of adding fillet and carcass as described in the BERA) at each 2019 sampling location (Figure 2). While the data show potential sources of dioxins/furans and coplanar PCBs upstream of the site, the highest concentrations of both dioxins/furans and coplanar PCBs were in fish collected at St. Regis, downstream of the site. As noted in a previous comment, coplanar PCBs can be a byproduct of combustion, and they are present in OU2 soils in the only samples collected at the site that included PCB congeners. Given that the site is a known source of these POPs, and the concentrations are increasing downstream, additional data should be collected to properly assess potential risk from both past and ongoing releases into the Clark Fork River.

Figure 2. TCDD TEQ in composite rainbow trout samples ("whole body," i.e., fillet + carcass) collected in 2019. We set non-detectable congeners to zero (ND = 0) and examined the data on a wet weight (ww) basis. The sample locations are shown upstream to downstream (left to right on the X axis), including Lolo (LOL), Missoula (MIS), Council Grove (COG), Frenchtown (FRN), and St. Regis (STR). (A) Average TCDD TEQ in each reach. (B) TCDD TEQ (dioxin/furan/PCBs) in individual composite samples.



2.5.6 Summary

The assessment of risks to fish is based on a very limited dataset, with many assumptions and calculations required to compare the existing data to TRBs derived from fish eggs in Steevens et al. (2005). The existing data for rainbow trout suggest that TCDD TEQ increases downstream of the site. Additional research is required to properly assess exposure to fish and piscivores downstream.

References

Burgess, R.M., R. Lohmann, J.P. Schubauer-Berigan, P. Reitsma, M.M. Perron, L. Lefkovitz, and M.G. Cantwell. 2015. Application of passive sampling for measuring dissolved concentrations of organic contaminants in the water column at three marine superfund sites. *Environmental Toxicology and Chemistry* 34(8):1720–1733.

Charlestra, L., D.L. Courtemanch, A. Amirbahman, and H. Patterson. 2008. Semipermeable membrane device (SPMD) for monitoring PCDD and PCDF levels from a paper mill effluent in the Androscoggin River, Maine, USA. *Chemosphere* 72:1171–1180.

Dinkins, S.A. and J.P. Heath. 1998. Quantification of Dioxin Concentrations in the Ohio River Using High Volume Water Sampling. Presented at the Proceedings of the National Water Quality Monitoring Council National Monitoring Conference, Reno, NV. July 7–9.

Helsel, D.R. 2005. More than obvious: Better methods for interpreting nondetect data. *Environmental Science and Technology* 39(20):419A–423A.

Hofmann, T. and A. Wendelborn. 2007. Colloid facilitated transport of polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) to the groundwater at Ma Da Area, Vietnam. *Environmental Science and Pollution Research* 14:223–224.

Imbrigiotta, T.E. and P.T. Harte. 2020. Passive Sampling of Groundwater Wells for Determination of Water Chemistry. Techniques and Methods. Chapter 8, Section D, Book 1. U.S. Geological Survey. Available: https://doi.org/10.3133/tm1d8. Accessed February 10, 2021.

ITRC. 2007. Protocol for Use of Five Passive Samplers to Sample for a Variety of Contaminants in Groundwater. DSP-5. Interstate Technology & Regulatory Council, Diffusion/Passive Sampler Team, Washington, DC.

Kandemir, Ş. and N. Polat. 2007. Seasonal variation of total lipid and total fatty acid in muscle and liver of rainbow trout (*Oncorhynchus mykiss W.*, 1792) reared in Derbent Dam Lake. *Turkish Journal of Fisheries and Aquatic Sciences* 7:27–31.

Lemieux, P.M., C.W. Lee, J.D. Kilgroe, and J.V. Ryan. 1999. *Emissions of Polychlorinated Biphenyls as Products of Incomplete Combustion from Incinerators*. EPA 600/A-99/051. Presented at the International Conference on Incineration and Thermal Treatment Technologies.

Lohmann, R. 2016. Development of a Passive Multisampling Method to Measure Dioxins/Furans and Other Contaminant Bioavailability in Aquatic Sediments. SERDP Project ER-2538. November.

Miller, M.A. 1993. Maternal transfer of organochlorine compounds in salmonines to their eggs. *Canadian Journal of Fisheries and Aquatic Sciences* 50:1405–1413.

NewFields. 2017. Addendum 4 to the Remedial Investigation Work Plan: 2017 Groundwater Monitoring. Prepared for M2Green Redevelopment LLC, International Paper Company, and WestRock CP, LLC. Version 2, July.

Niimi, A. 1983. Biological and toxicological effects of environmental contaminants in fish and their eggs. *Canadian Journal of Fisheries and Aquatic Sciences* 40:306–312.

Russell, R.W., F.A. Gobas, and G.D. Haffner. 1999. Maternal transfer and in ovo exposure of organochlorines in oviparous organisms: a model and field verification. *Environmental Science and Technology* 33:416–420.

Steevens, J.A., M.R. Reiss, and A.V. Pawlisz. 2005. A methodology for deriving tissue residue benchmarks for aquatic biota: A case study for fish exposed to 2, 3, 7, 8-tetrachlorodibenzo-p-dioxin and equivalents. *Integrated Environmental Assessment and Management: An International Journal* 1:142–151.

U.S. EPA. 1990. U.S. EPA/Paper Industry Cooperative Dioxin Study: "The 104 Mill Study" Statistical Findings and Analyses. U.S. Environmental Protection Agency, Washington, DC. July 13.

U.S. EPA. 1997. *Ecological Risk Assessment Guidance for Superfund: Process for Designing and Conducting Ecological Risk Assessments*. Interim Final. EPA 540-R-97-006, OSWER 9285.7-25, PB97-963211. U.S. Environmental Protection Agency. June.

U.S. EPA. 1998. *Guidelines for Ecological Risk Assessment*. EPA/630/R-95/002F. U.S. Environmental Protection Agency.

U.S. EPA. 2012. Guidelines for Using Passive Samplers to Monitor Organic Contaminants at Superfund Sediment Sites. OSWER Directive 9200.1-110 FS. U.S. Environmental Protection Agency. December.

U.S. EPA and SRC. 2020. Draft Baseline Ecological Risk Assessment for Operable Units 2 & 3 of the Smurfit-Stone/Frenchtown Mill Site Located in Missoula County, Montana. Prepared by the U.S. Environmental Protection Agency, Region 8, with technical assistance from SRC, Inc, Denver, CO. November.