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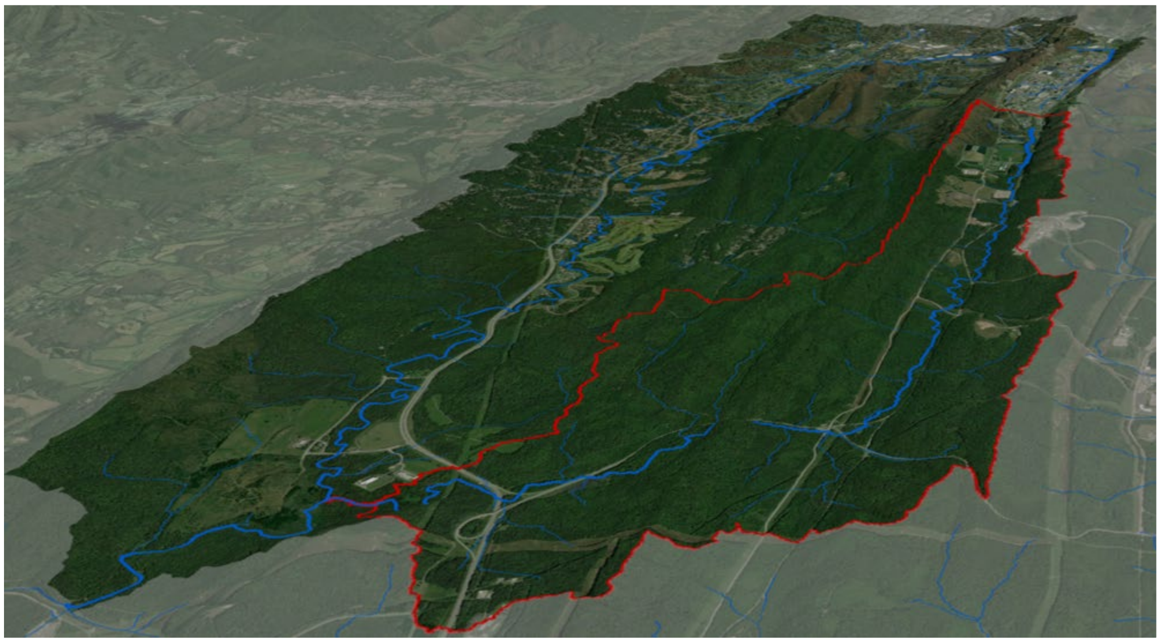
Towards Development of a Conceptual Model for Mercury in Bear Creek, Oak Ridge, Tennessee – FY23 Update



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September 2023

OAK RIDGE National Laboratory

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Environmental Sciences Division

TOWARDS DEVELOPMENT OF A CONCEPTUAL MODEL FOR MERCURY IN BEAR CREEK, OAK RIDGE, TENNESSEE – FY23 UPDATE

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ABBREVIATIONS

BCK	Bear Creek kilometer
BCV	Bear Creek Valley
BSWTS	Big Springs Water Treatment System
CMTS	Central Mercury Treatment System
DEM	digital elevation model
DOE	US Department of Energy
D&D	decontamination and decommissioning
EFPC	East Fork Poplar Creek
FY	fiscal year
Hg	mercury
MeHg	methylmercury
MTF	mercury treatment facility
NAP	nitric acid pipeline
OF	outfall
ORNL	Oak Ridge National Laboratory
ORR	Oak Ridge Reservation
RMPE	Reduction of Mercury in Plant Effluent
ROD	Record of Decision
SWAT	Soil Water Analysis Tool
UEFPC	Upper East Fork Poplar Creek
WEMA	West End Mercury Area
Y-12	Y-12 National Security Complex

EXECUTIVE SUMMARY

Mercury concentrations in fish in Bear Creek are elevated and comparable to concentrations seen in fish in East Fork Poplar Creek (EFPC) on the Oak Ridge Reservation, even though aqueous inorganic mercury concentrations are orders of magnitude lower in Bear Creek than in EFPC. Acknowledging that the relationship between aqueous and fish tissue mercury concentrations is not linear, and that methylmercury (MeHg) concentrations are likely more related to fish tissue concentrations than aqueous total mercury (Hg_T) concentrations, MeHg production is not easily predicted or controlled. In Bear Creek, where aqueous Hg_T concentrations are low, factors other than mercury loading drive the transformation of mercury to MeHg and subsequent trophic transfer to fish.

Initial development of a waste disposal facility (Environmental Management Disposal Facility; EMDF) in Bear Creek Valley has begun, and operation of the EMDF has the potential to increase mercury inputs to Bear Creek. Consequently, understanding the factors contributing to elevated MeHg concentrations in water and fish in Bear Creek has increased relevance and importance. This report summarizes data from recent and historical compliance and investigatory studies with an eye toward building a conceptual model to understand the processes affecting mercury transport and transformation in the Bear Creek watershed, as well as to highlight key knowledge gaps in our understanding of these processes that warrant further investigation. The conceptual model will provide a strong technical basis for prioritizing and optimizing potential mitigation actions or best management practices to minimize potential negative effects of the EMDF related to mercury in Bear Creek.

1. INTRODUCTION

The Bear Creek Valley (BCV) watershed (Figure 1) lies within the boundaries of the Oak Ridge Reservation (ORR) in Roane and Anderson counties. Bear Creek is a 12.9 km second/third-order stream whose valley is bounded to the north by Pine Ridge and to the south by Chestnut Ridge. The stream originates near the former S-3 Waste Management Area within the boundaries of the Oak Ridge Y-12 National Security Complex (Y-12) and flows into East Fork Poplar Creek at kilometer (EFK) 2.6. The watershed has a drainage area of 18.52 km^2, and although it contains no privately owned lands, it contains numerous disposal areas previously used for waste management by Y-12. Disposal of uranium, waste oils, solvents, and radionuclides other than uranium have historically contaminated groundwater as well as the surface water draining the waste areas.

In contaminated environments, conceptual models can aid in visualizing and understanding the dynamic nature of the hydrologic, geochemical, and physical environment. These conceptual models integrate data in an internally consistent manner to understand processes that control the fate and transport of contaminants. Over the past few decades of environmental investigation at Y-12, many conceptual models have been developed to identify and define various technical processes at various scales. In Bear Creek, the primary contaminants of concern have been identified to be uranium, ^{99}Tc, nitrate, thorium, and volatile organic compounds (e.g., acetone, methylene chloride, toluene, and tetrachloroethylene) originating from the former S-3 Waste Disposal Ponds. As a result, a considerable amount of effort over a number of years went into developing conceptual models and fundamental research to understand the transport and transformations of these contaminants in the BCV watershed (Moss et al. 1999). In contrast, because aqueous mercury concentrations in Bear Creek are relatively low, mercury has not been a focus of extensive monitoring.

However, mercury concentrations in fish in Bear Creek are elevated and comparable to concentrations seen in fish in East Fork Poplar Creek (EFPC) on the Oak Ridge Reservation, even though aqueous inorganic mercury concentrations are orders of magnitude lower in Bear Creek than in EFPC. Although it is recognized that the relationship between aqueous and fish tissue mercury concentrations is not linear, and that methylmercury (MeHg) concentrations are more likely to be related to fish tissue concentrations than aqueous total mercury (Hg_T) concentrations, MeHg production is not easily predicted or controlled. This suggests that in Bear Creek, factors other than mercury loading drive the transformation of mercury to MeHg and subsequent trophic transfer to fish.

Initial development of a waste disposal facility (Environmental Management Disposal Facility; EMDF) in Bear Creek Valley has begun, and EMDF operation has the potential to increase mercury inputs to Bear Creek. Consequently, understanding the factors contributing to elevated MeHg concentrations in water and fish in Bear Creek has increased relevance and importance. This report summarizes data from recent compliance and investigatory studies with an eye toward building a conceptual model to understand the processes affecting mercury transport and transformation in the Bear Creek watershed, as well as to highlight key knowledge gaps in our understanding of these processes that warrant further investigation. The Bear Creek mercury conceptual model will provide a strong technical basis for prioritizing and optimizing potential mitigation actions or best management practices to minimize potential negative effects of the EMDF related to mercury in Bear Creek.

Our focus for fiscal year (FY) 2022 was on compiling and summarizing available mercury data in Bear Creek as well as other water chemistry parameters that may explain MeHg concentrations in the watershed. During FY23, as part of the EMDF Record of Decision (DOE/OR/01-2794&D2/R2), it was agreed upon that a Remedial Site Evaluation (RSE) should be conducted. Thus, our focus this FY pivoted to prioritizing and planning additional mercury characterizations in the watershed to be conducted in early FY24 under the RSE. The overall goal of the planned activities is to "evaluate potential sources of mercury and methylmercury within the Bear Creek valley watershed" (Draft Bear Creek Valley Mercury Sources Remedial Site Evaluation Sampling and Analysis Plan Oak Ridge, Tennessee). The data quality objectives (DQO) planning for the RSE followed the seven-step iterative process outlined by the United States Environmental Protection agency in EPA/240/B-06/001 (US EPA 2006) to determine a sample collection design to meet stated goals and objectives of the activity. This status report summarizes our activities from this FY in support of developing a conceptual model for mercury in Bear Creek and also includes the historic data inventory and summary from the FY22 report.

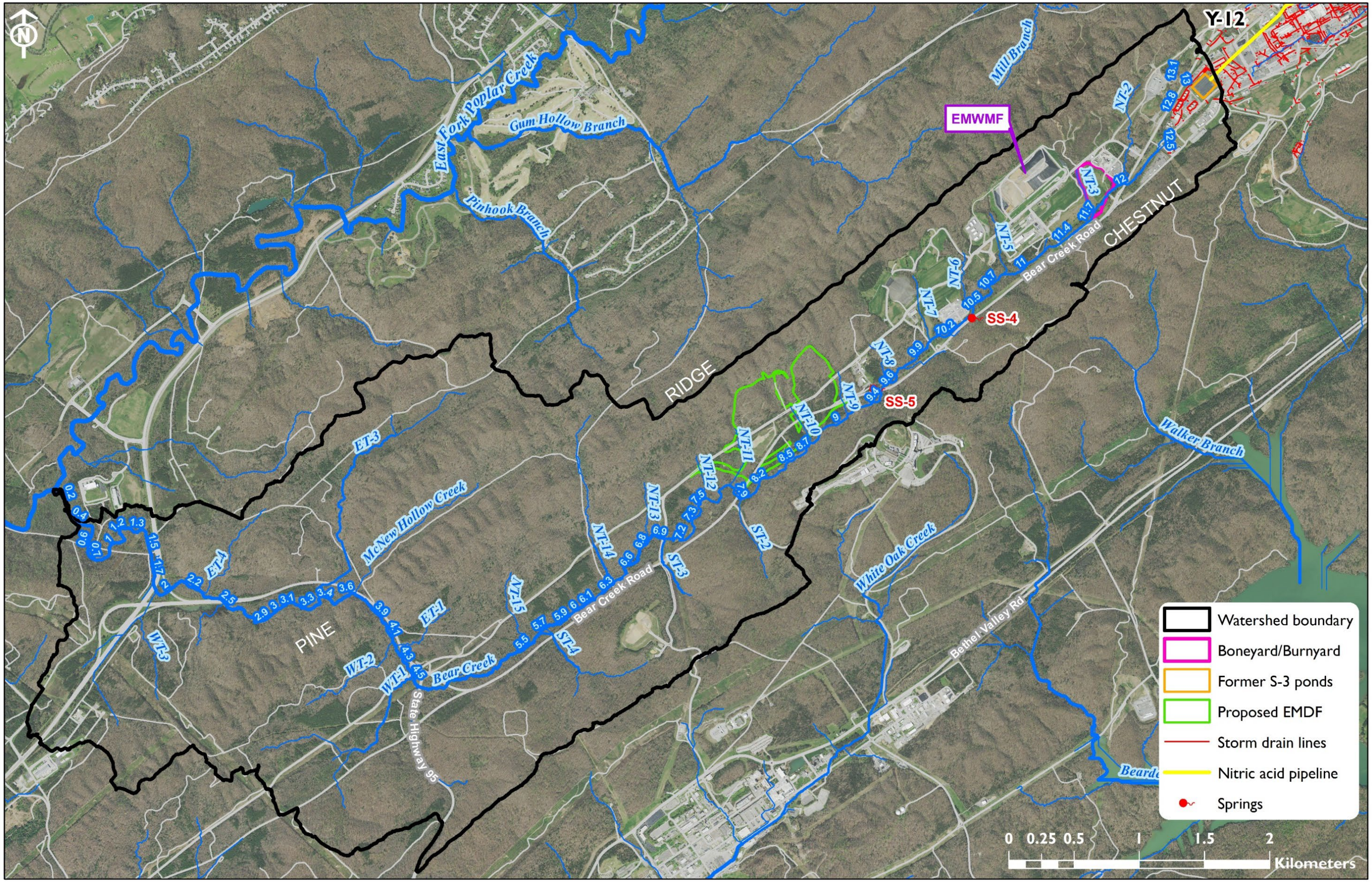

Figure 1. Bear Creek Watershed.
All locations approximate. Distance from creek mouth in kilometers shown in labels along Bear Creek mainstem.

2. HISTORIC DATA INVENTORY AND SUMMARY

In FY22, we compiled historic and recent data relevant to mercury biogeochemistry and transformations in Bear Creek from the Oak Ridge Environmental Information System (OREIS), including rainfall/streamflow, total and dissolved mercury concentrations in surface water and groundwater, MeHg in surface water, temperature, nitrate, and sulfate. Maps and infographics that summarize the OREIS data available for these parameters are presented in the following sections.

2.1 RAINFALL AND STREAMFLOW

We compiled rainfall data from multiple sources to include locations in the vicinity of Bear Creek Valley and integrated the data into a daily rainfall record for the watershed for 1999 through 2021 (Figure 2). These rainfall data were also used in the ongoing watershed model development for the Bear Creek watershed described in Section 3.3. Larger rainfall events in the watershed can approach or even exceed daily totals of 100 mm rainfall. Creek flow has been measured at numerous points in the watershed, with the most extensive flow records between Bear Creek kilometer (BCK) 9.2 and BCK 12.34, including several tributaries to Bear Creek within that reach (Figure 2). In general, flows in the upper part of the mainstem at BCK 12.34 average 0.01 cubic meters per second (cms), with flows reaching over 0.5 cms following larger rainfall events. In the lower parts of the mainstem, for which fewer flow records are available, average flows are 0.2 cms, with peak flows reaching 9 cms near BCK 3.3 and station 3538273, which is located upstream of BCK 3.3 between WT-2 and McNew Hollow Creek. Average flows in tributaries NT-1, NT-3, NT-4, NT-5, NT-7, and NT-8 range from 0.002 cms to 0.007 cms. Average flows in EFPC upstream of the Bear Creek confluence are approximately 1 cms.

2.2 MERCURY AND METHYLMERCURY IN SURFACE WATER AND GROUNDWATER

The source(s) of mercury to Bear Creek are not well understood, but mercury has historically been monitored in and around the S-3 ponds and the nitric acid pipeline. Located adjacent to the head of Bear Creek, the former S-3 disposal ponds comprised four unlined infiltration pits and covered an area of about 1.44 ha (Brooks 2001, Jeter 1978, Union Carbide Corporation 1983). These ponds had a storage capacity of about 9.5 million liters and received wastes for 32 years, from 1951 to 1983. The waste stream was primarily acidic uranium nitrate solution (pH ~1) generated within Y-12, although there were contributions from other processes and sites (e.g., K-25, X-10, Idaho National Laboratory) contributing other components to the mixture, such as organic solvents, polychlorinated biphenyls, and radionuclides other than uranium.

Detailed records of the waste composition were not kept during the operational lifetime of the S-3 ponds, but the liquids and sludges were characterized after waste disposal stopped and before the ponds were closed and capped with a multilayer RCRA cap and asphalt (the site now serves as a parking lot). Mercury concentrations in the liquid samples ranged between 3 $\mu g \cdot L^{-1}$ and 320 $\mu g \cdot L^{-1}$, and mercury concentrations in the sludges ranged between 0.21 $mg \cdot kg^{-1}$ and 12 $mg \cdot kg^{-1}$.

Most of the liquid waste disposed in the S-3 ponds was delivered via a buried pipeline—the nitric acid pipeline (NAP). During the NAP remedial investigation, the precaution was taken to monitor for mercury vapor concentrations for all excavations. Mercury vapor was detected above background levels at several locations. This precaution may have been put in place because the pipeline transited areas of known or suspected liquid mercury spills as opposed to mercury-contaminated liquid wastes in the pipeline leaking into surrounding soil. No results for mercury concentration in soils were reported.

Groundwater contamination and migration along Bear Creek Valley have been well documented through remedial investigations and intensive scientific research (e.g., NABIR Field Research Center and the

Integrated Field Research Challenge). Among the outcomes of those studies was the characterization of complex groundwater flow paths resulting in several distinct arms of the contaminant plume, some of which discharge to the headwaters and tributaries that feed into the mainstem of Bear Creek. Contaminants from the S-3 ponds appear in the SS-5 spring more than 3 km from the S-3 site and continue downstream in the surface water.

Exit pathways exist for mercury originating in the S-3 ponds to contribute to mercury loading in Bear Creek. Even though the sensitivity of instruments during the early characterization and remedial investigations was poor relative to current capabilities, mercury was identified as the "most widespread of all inorganic contamination" in soils at the S-3 site (Lockheed Martin Energy Systems Inc. 1997). Mercury from the S-3 site was identified as contributing to elevated concentrations in water at BCK 12.7. Mercury was also detected up to 20× above established background levels (0.34 $mg \cdot kg^{-1}$) in creek sediments and floodplain soils up to 40 cm deep and more than 5 km downstream from the S-3 site, although some of that mercury may have originated from waste burial grounds through Bear Creek Valley. Additionally, mercury concentrations in sediment and water samples collected from BCK 12.3 in 2008 were comparable to mercury-contaminated reaches of White Oak Creek at Oak Ridge National Laboratory (ORNL) and substantially higher than a local creek with no known source of mercury contamination (Mosher 2012). Reported mercury concentrations in groundwater in the vicinity of the S-3 ponds ranges between 0.9 $\mu g \cdot L^{-1}$ and 80 $\mu g \cdot L^{-1}$. Sampling conducted in recent years has largely been in the lower two-thirds of Bear Creek, which has left an incomplete understanding of mercury concentrations in the upper reaches.

As shown in Figure 3, many samples have been collected throughout the watershed from the early 1990s to the present; these samples have been analyzed for Hg_T for different projects and purposes and archived in OREIS or collected recently as part of special studies (see Section 3.1). There have been Hg_T detections throughout the watershed, with the highest detections seen at NT-1, NT-3, BCK 12.34/BCK 12.47, and at BYBYFD-EFF near the former boneyard/burnyard, all in the upper part of the watershed (Figure 4). BCK 12.31, NT-1, and NT-3 are the monitoring locations with the most substantial Hg_T sample record available in OREIS. The sample record for dissolved mercury (Hg_D) is much sparser than that for Hg_T; the most recent sampling and detections throughout the watershed from the limited sampling are associated with special studies (Figure 5; Section 3.1).

Total methylmercury ($MeHg_T$) samples have been collected at multiple points throughout the watershed (Figure 6) from 2009 to the present (Figure 7). $MeHg_T$ concentrations are influenced by the time of year the sample was collected, and concentrations can fluctuate at a sampling location within the same year and across years, as shown in Figure 7.

Groundwater throughout the watershed (Figure 8) has been analyzed for Hg_T (Figure 9) and Hg_D (Figure 10) for the past three decades, with many more samples analyzed for Hg_T. Most samples have been below laboratory detection limits, but elevated levels of Hg_T have consistently been detected at GW-246 over the past 15 years. GW-246 lies to the south of the former S-3 Waste Disposal Ponds, between the former pond site and the headwater channel of the Bear Creek mainstem.

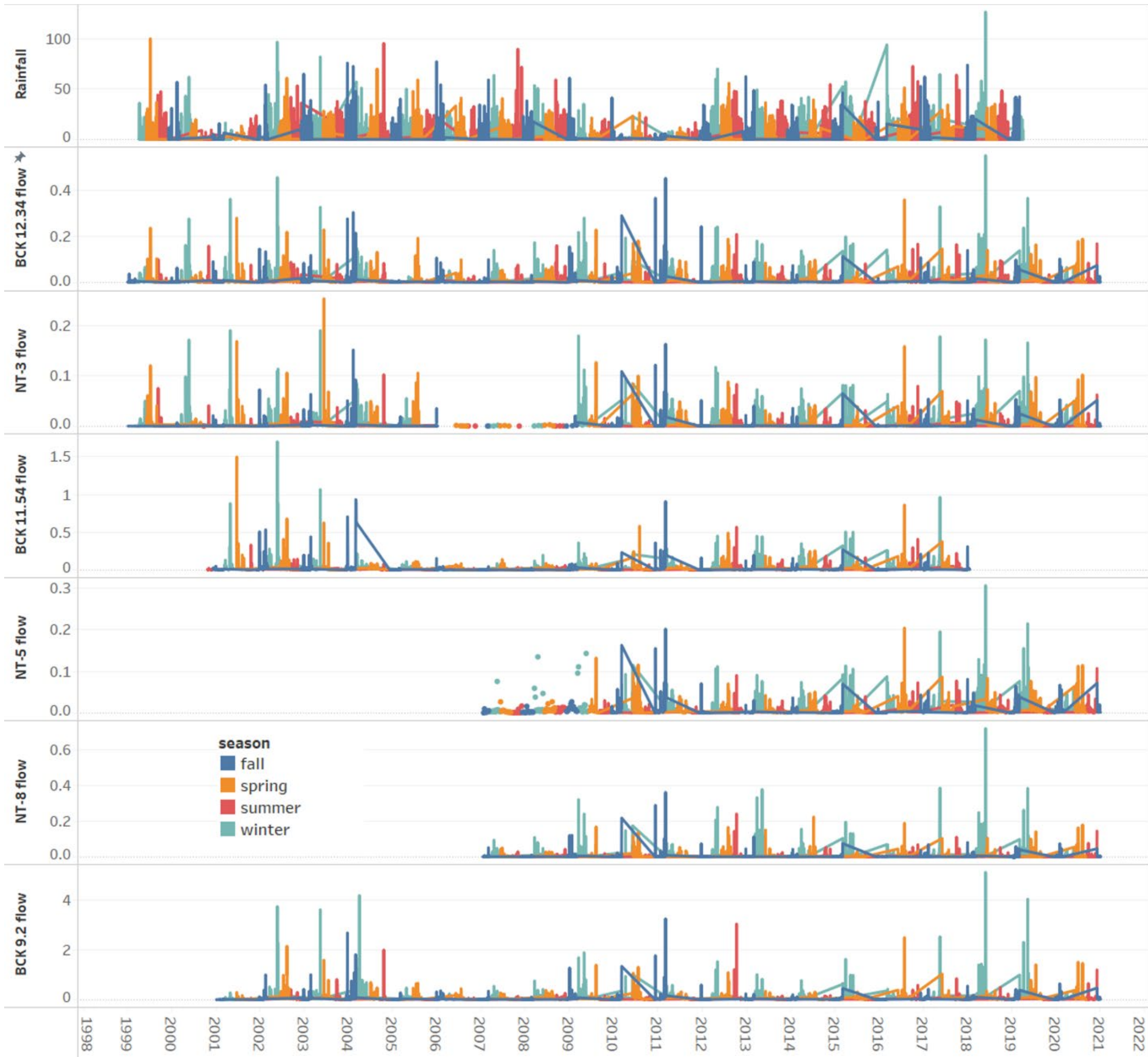

Figure 2. Watershed rainfall inputs and flows for mainstem and tributaries.
Daily rainfall (mm) and flows (cms) for Bear Creek mainstem and tributaries with significant flow records over time. Note the y-axis varies by location.

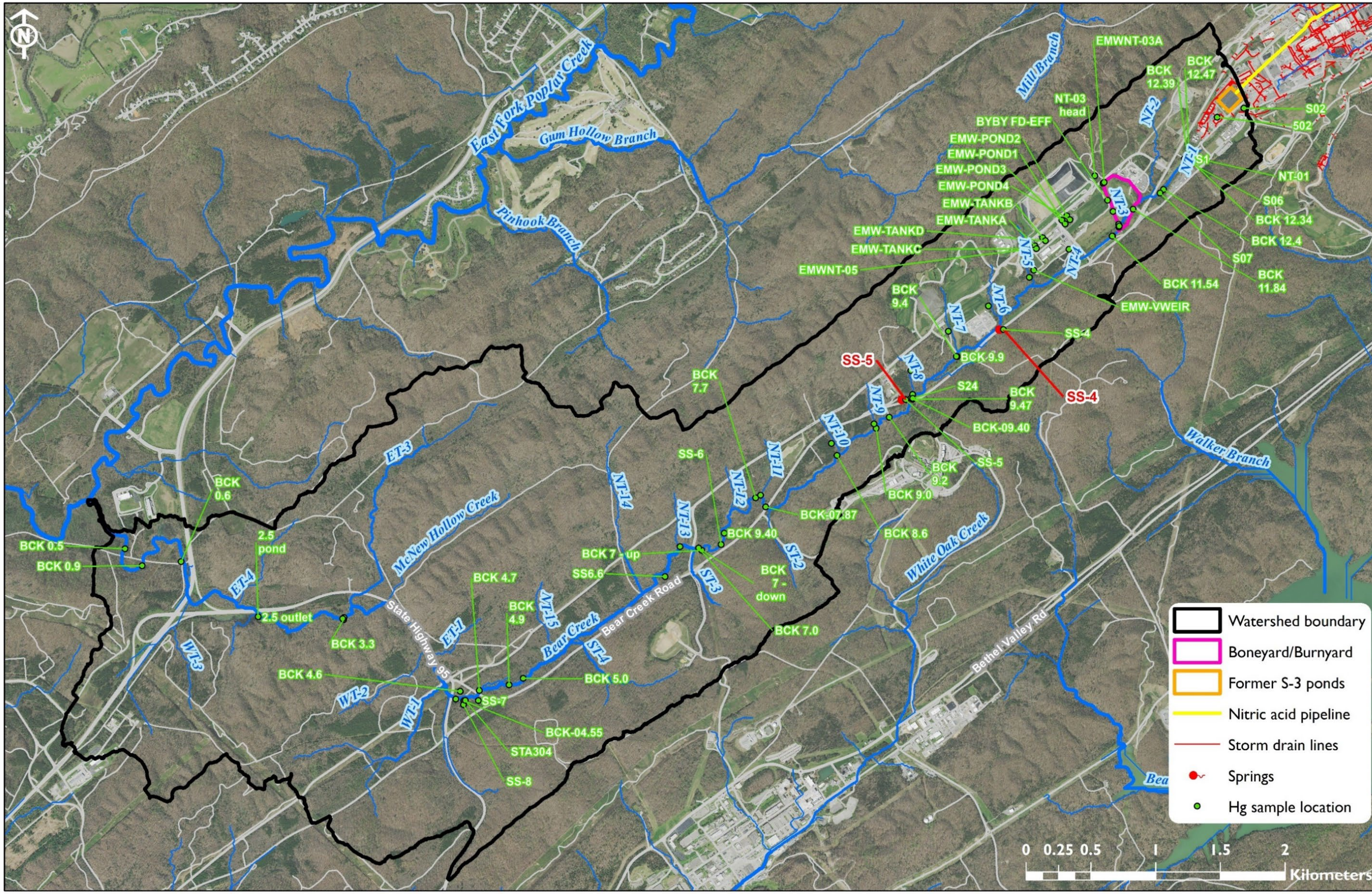

Figure 3. Surface water mercury sampling locations in Bear Creek watershed.
Only sites with at least ten sample results and with at least one sample within analytical method detection limits included on map.

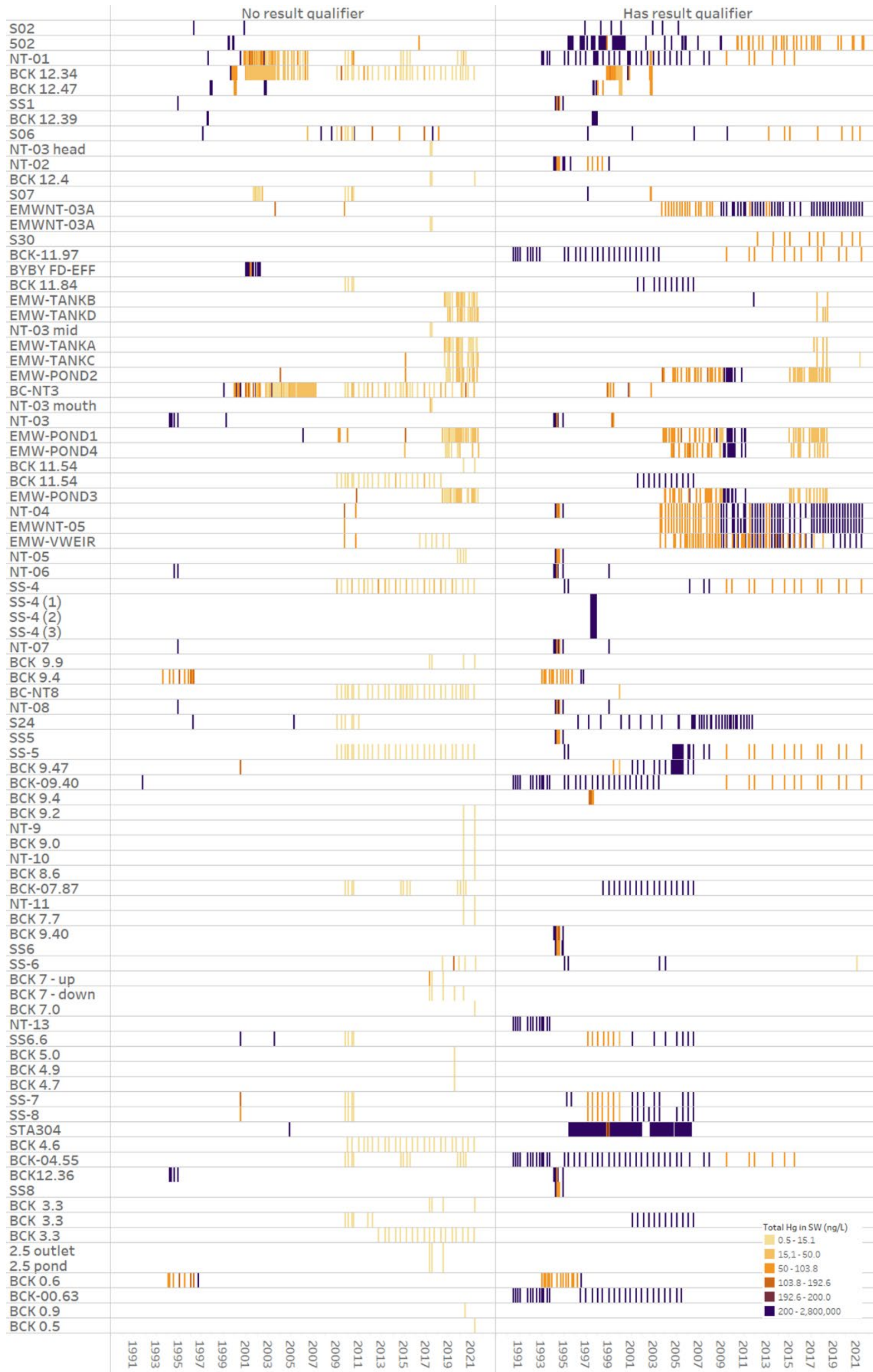

Figure 4. Total mercury concentrations in surface water.

Sample locations are sorted based on distance to the mouth of Bear Creek at its confluence with EFPC, with the headwaters at the top of the graphic and the mouth at the bottom. Results are shown only for locations with at least 10 sample results or locations included in special studies conducted from 2017 to 2021. Quantile classification applied for visualization so that each class contains an equal number of data points. Graphic is not intended to show specific concentrations but shows spatial and temporal sampling patterns and relative concentrations throughout the watershed. The 'No result qualifier' column shows results within analytical method detection limits, while the 'Has result qualifier' column shows results below method detection limits or those with some other flagged issue.

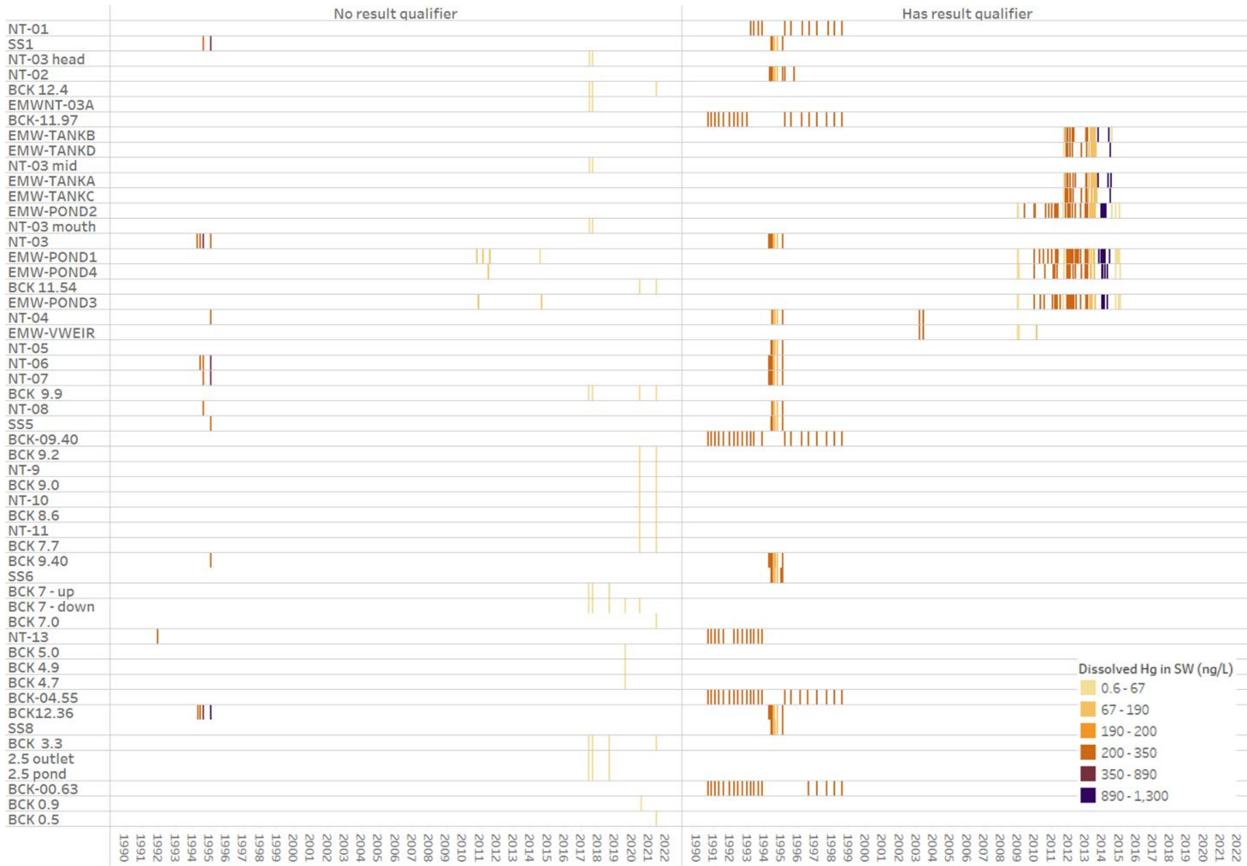

Figure 5. Dissolved mercury concentrations in surface water.

Sample locations are sorted based on distance to the mouth of Bear Creek at its confluence with EFPC, with the headwaters at the top of the graphic and the mouth at the bottom. Quantile classification applied for visualization so that each class contains an equal number of data points. Graphic is not intended to show specific concentrations but shows spatial and temporal sampling patterns and relative concentrations throughout the watershed. The 'No result qualifier' column shows results within analytical method detection limits, whereas the 'Has result qualifier' column shows results below method detection limits or those with some other flagged issue.

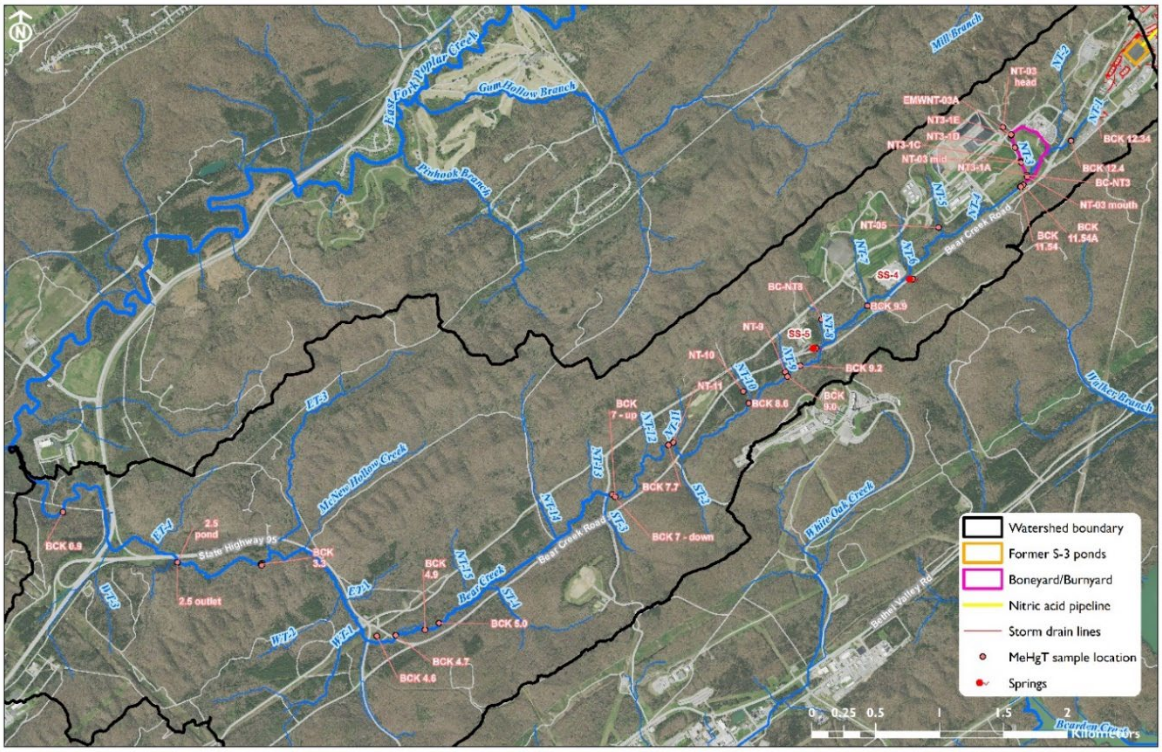

Figure 6. Methylmercury surface water sampling locations.

Figure 7. Total MeHg concentrations in surface water.

Sample locations are sorted based on distance to the mouth of Bear Creek at its confluence with EFPC, with the headwaters at the top of the graphic and the mouth at the bottom. Quantile classification applied for visualization so that each class contains an equal number of data points. Graphic is not intended to show specific concentrations but shows spatial and temporal sampling patterns and relative concentrations throughout the watershed. The 'No result qualifier' column shows results within analytical method detection limits, whereas the 'Has result qualifier' column shows results below method detection limits or those with some other flagged issue.

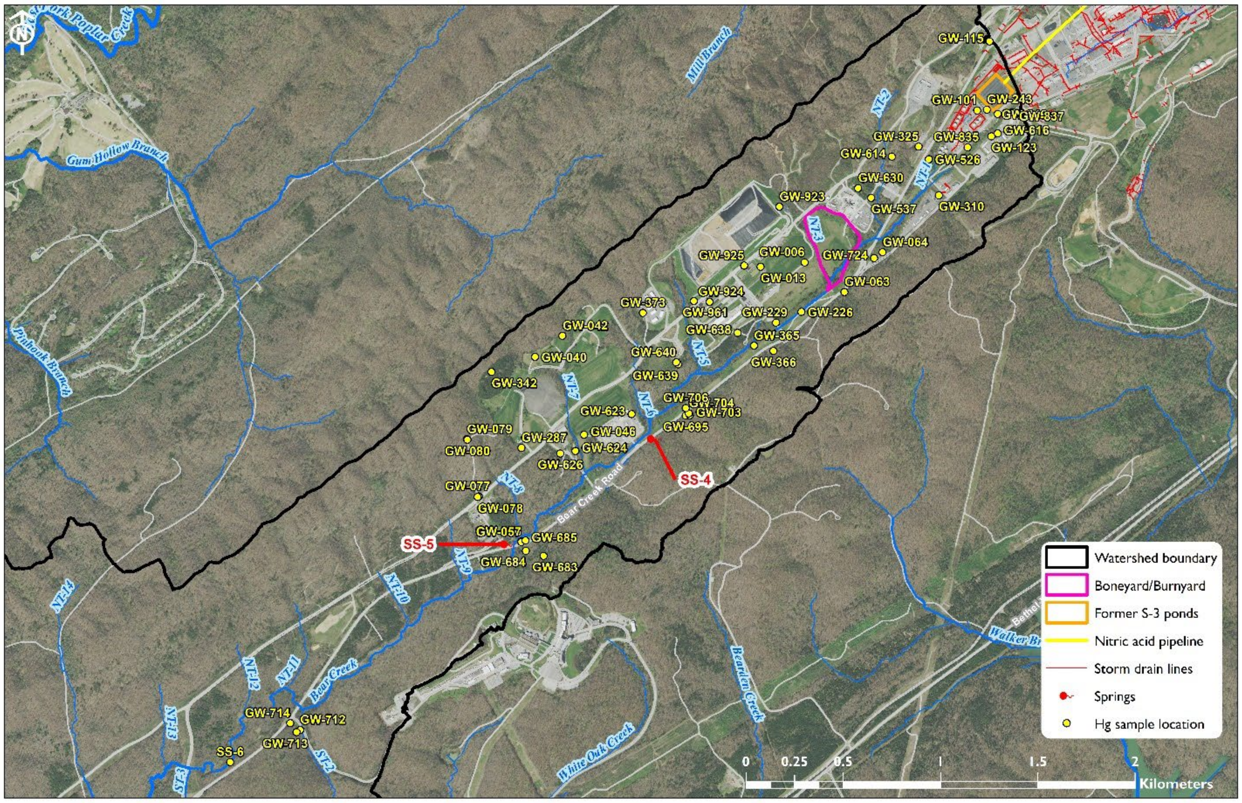

Figure 8. Groundwater mercury sampling locations.

Only sites with at least 10 sample results and with at least 1 sample within analytical method detection limits included on map.

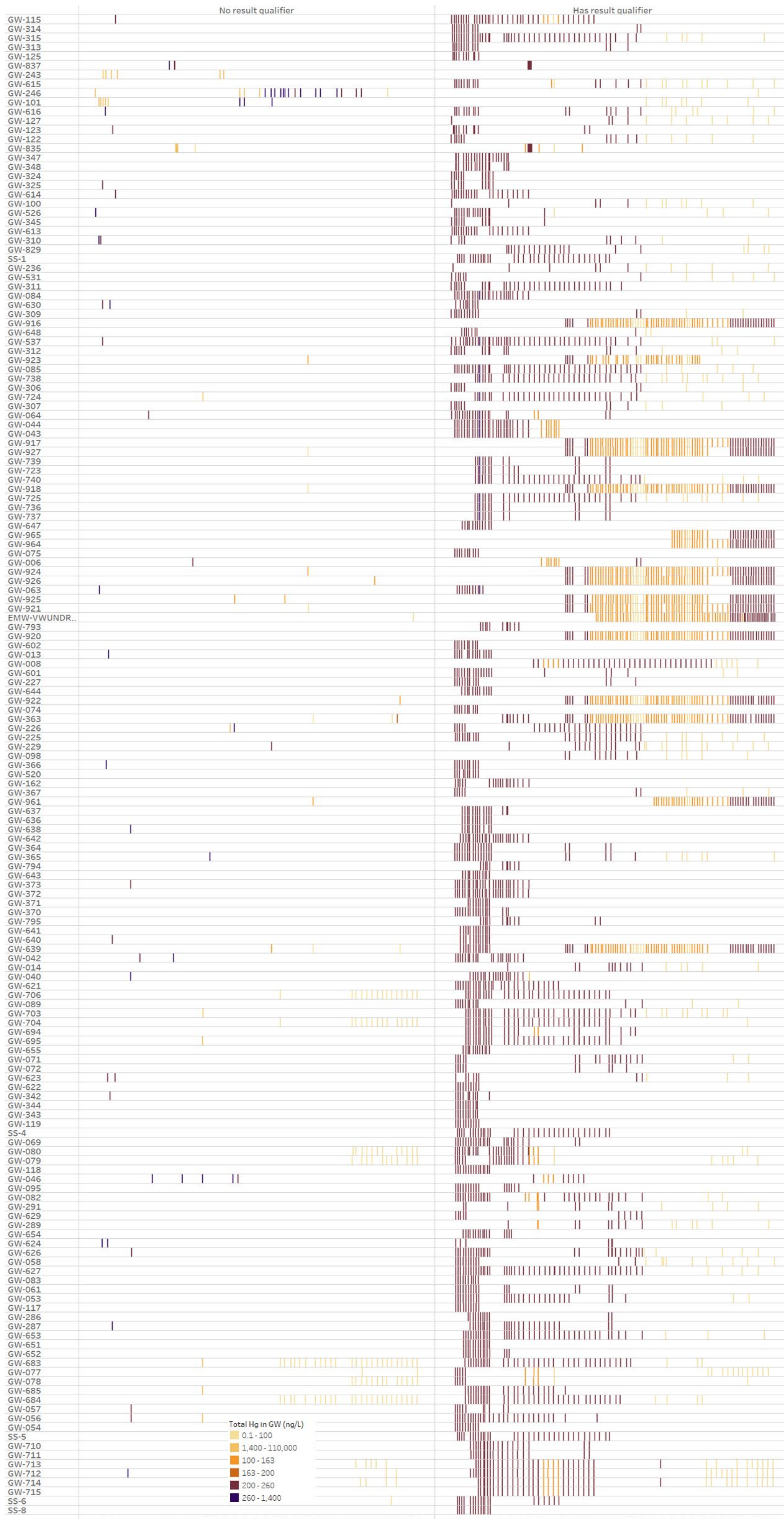

Figure 9. Total mercury concentrations in groundwater.

Sample locations are sorted based on distance to the mouth of Bear Creek at its confluence with EFPC, with the headwaters at the top of the graphic and the mouth at the bottom. Results are shown only for locations with at least 10 sample results. Quantile classification applied for visualization so that each class contains an equal number of data points. Graphic is not intended to show specific concentrations but shows spatial and temporal sampling patterns and relative concentrations throughout the watershed. The 'No result qualifier' column shows results within analytical method detection limits, whereas the 'Has result qualifier' column shows results below method detection limits or those with some other flagged issue.

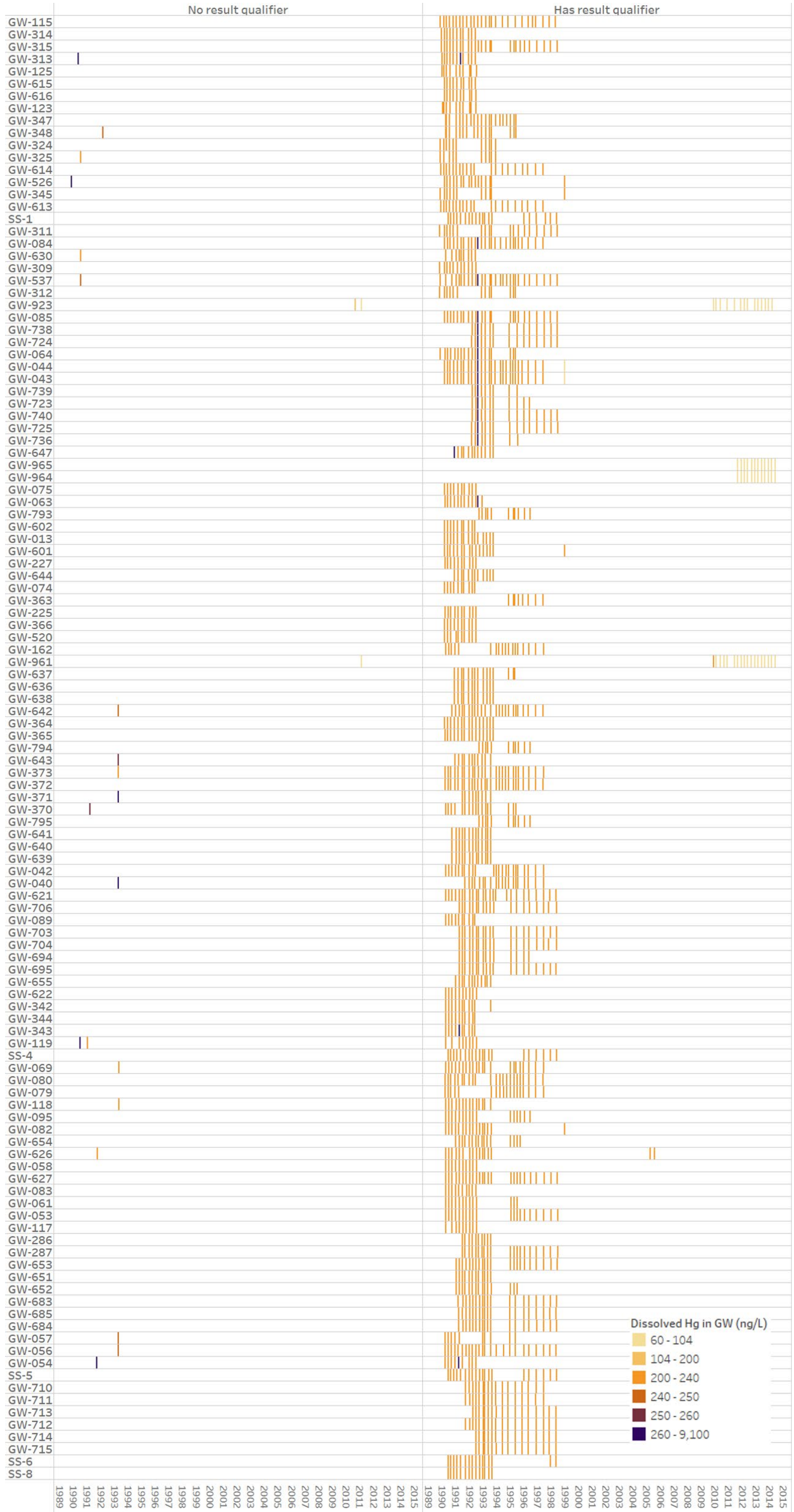

Figure 10. Dissolved mercury concentrations in groundwater.
Sample locations are sorted based on distance to the mouth of Bear Creek at its confluence with EFPC, with the headwaters at the top of the graphic and the mouth at the bottom. Results shown only for locations with at least 10 sample results. Quantile classification applied for visualization so that each class contains an equal number of data points. Graphic is not intended to show specific concentrations but shows spatial and temporal sampling patterns and relative concentrations throughout the watershed. The ‘No result qualifier’ column shows results within analytical method detection limits, whereas the ‘Has result qualifier’ column shows results below method detection limits or those with some other flagged issue.

2.3 TEMPERATURE, SULFATE, AND NITRATE IN SURFACE WATER

We also compiled and visualized data from OREIS on several other environmental and water chemistry parameters known to influence the mercury cycle and methylation in streams and rivers. Field measurements of water temperature have been collected at various points along Bear Creek and its tributaries (Figure 11) over the previous two decades. These data are plotted seasonally in Figure 12. In general, temperatures range from 3–4°C in some of the small tributaries in the upper part of the watershed in the winter to 26–28°C in both some tributaries and the mainstem in the summer.

Many samples collected throughout the watershed (Figure 13, Figure 14) over the past three decades (Figure 15, Figure 16) have been analyzed for nitrate and sulfate. Elevated nitrate and sulfate levels have been detected at numerous monitoring points over the past decade in both tributaries and the mainstem in upper Bear Creek watershed. Sulfate data for the previous decade is lacking for the lower parts of the watershed.

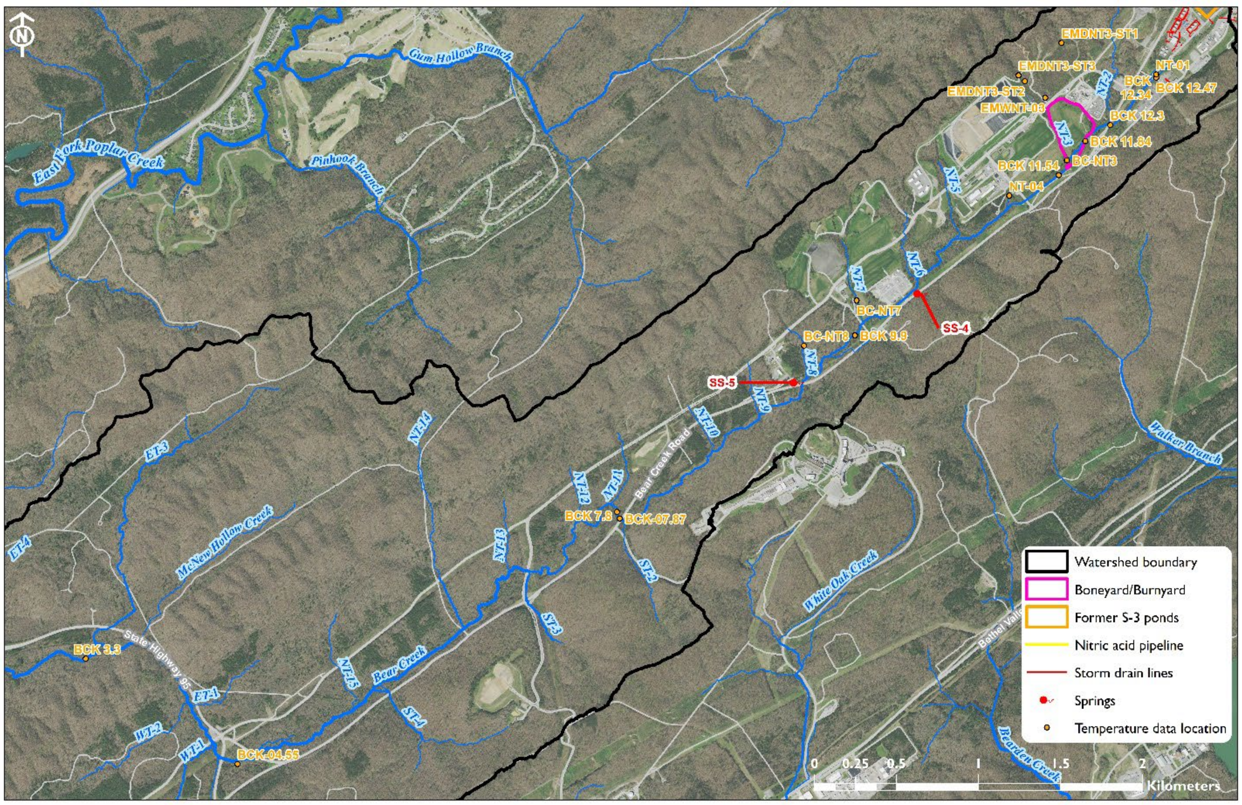

Figure 11. Surface water temperature measurement locations.

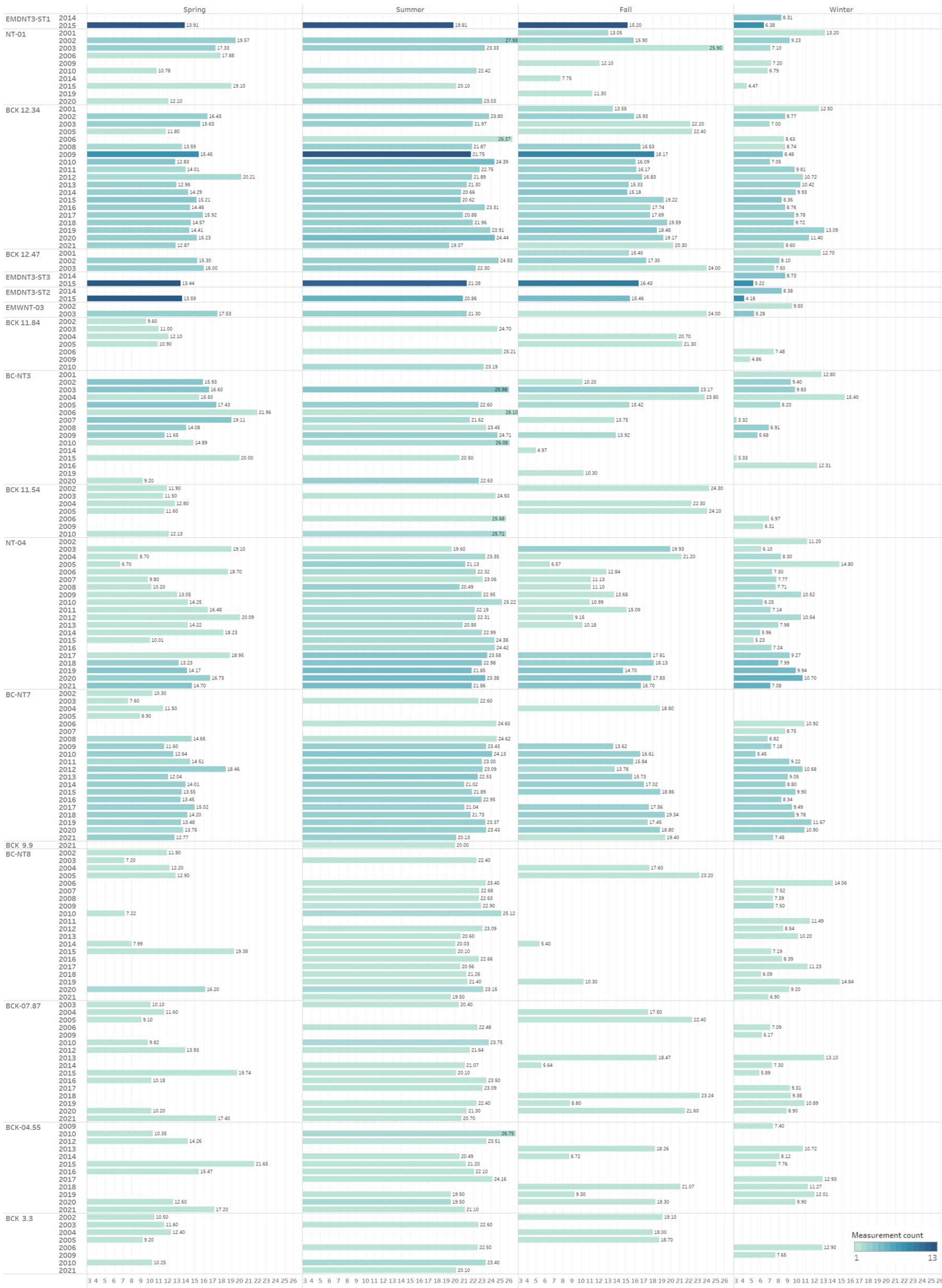

Figure 12. Surface water temperatures.

Measurement locations are sorted based on distance to the mouth of Bear Creek at its confluence with EFPC, with the headwaters at the top of the graphic and the mouth at the bottom. Results are shown only for locations with at least 10 temperature measurements. Bar labels show average temperature at location for each year and season where data are available. Measurement counts for each location, year, and season range from 1 to 13 and are indicated by the bar colors.

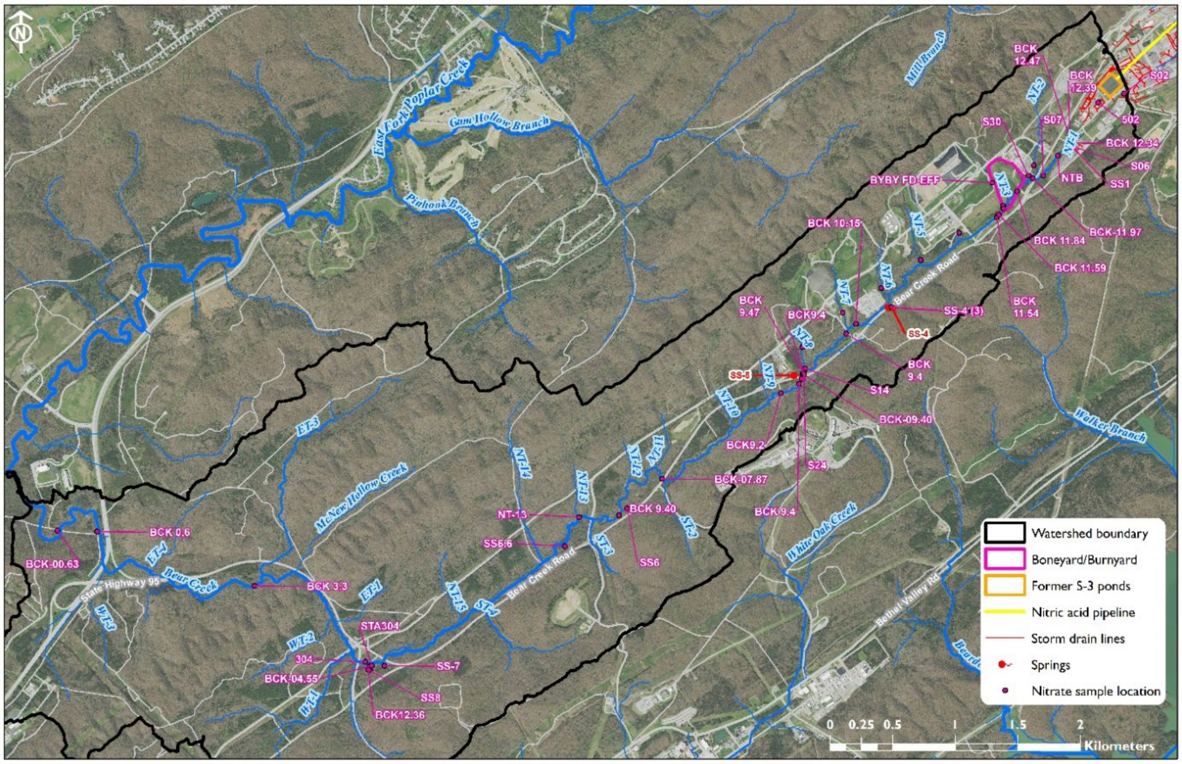

Figure 13. Nitrate surface water sampling locations.
Only sites with at least 10 sample results included on map.

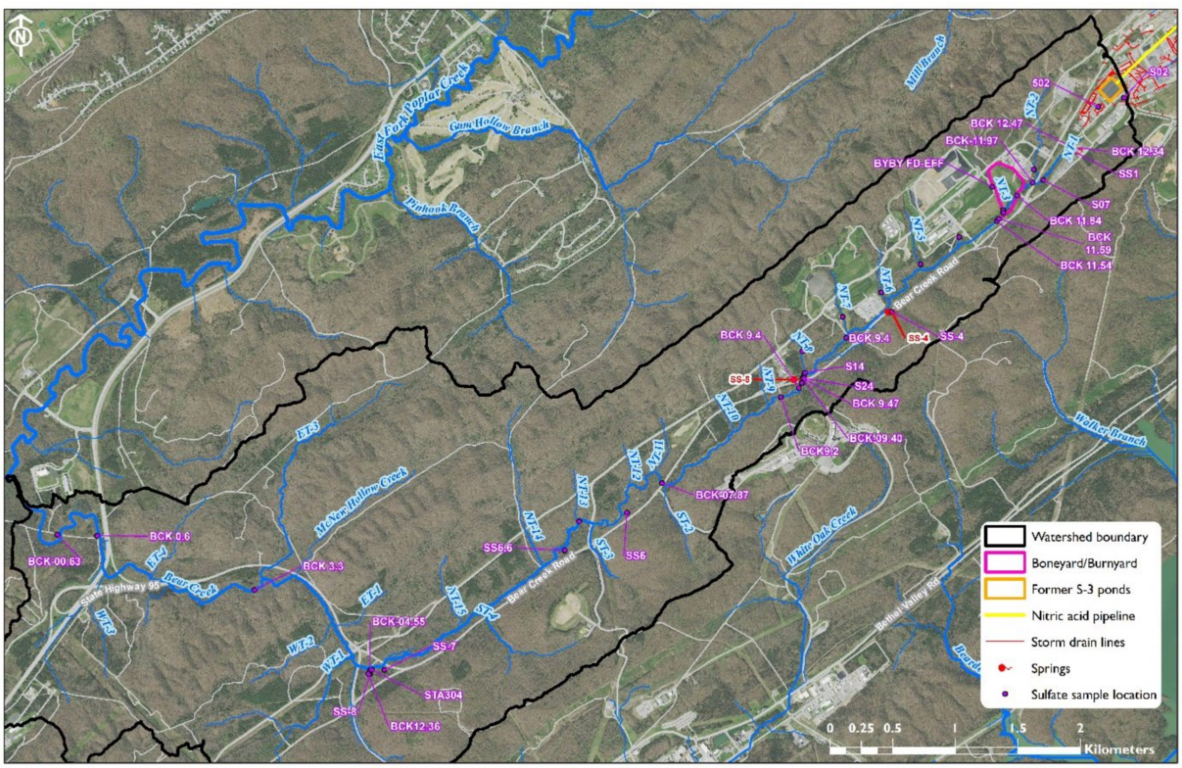

Figure 14. Sulfate surface water sampling locations.
Only sites with at least 10 sample results included on map.

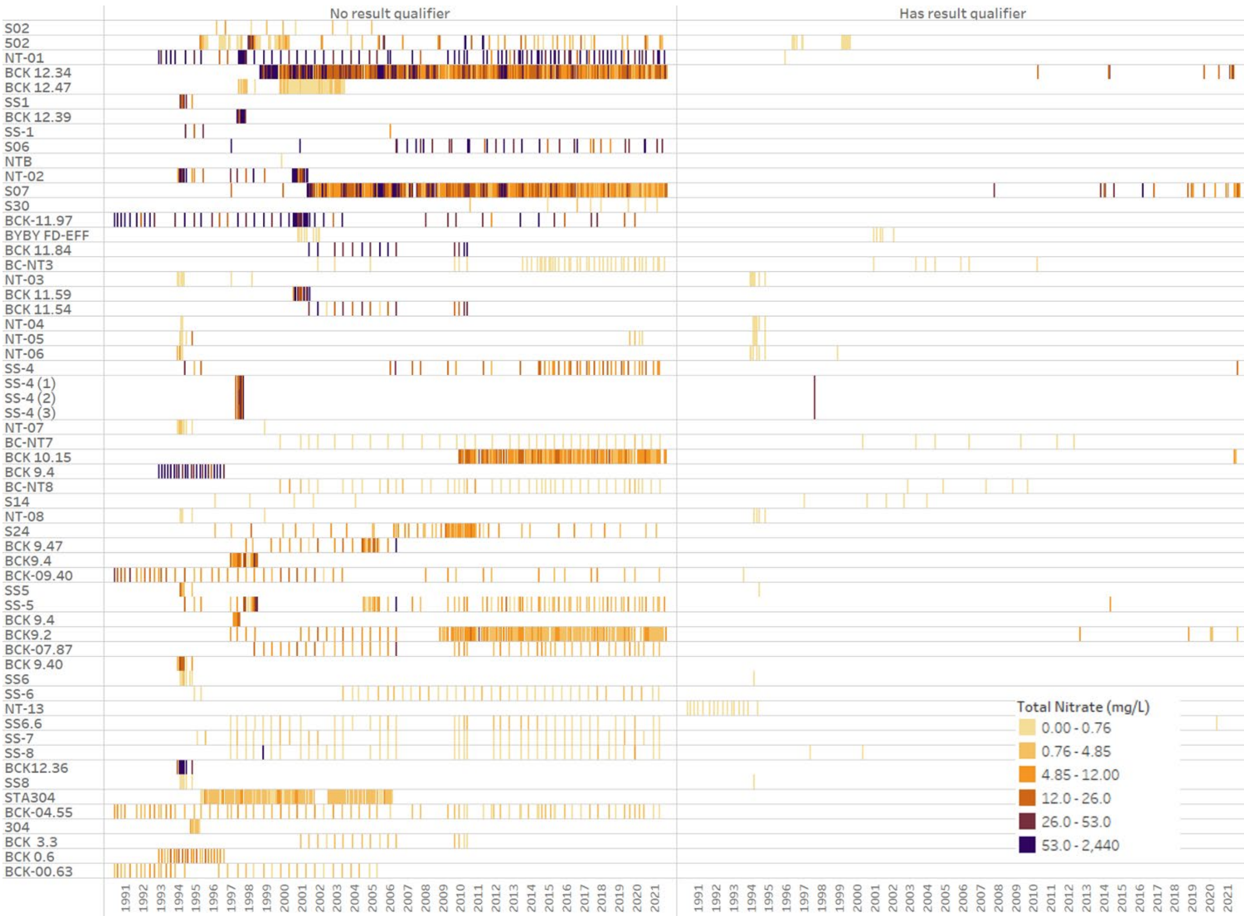

Figure 15. Total nitrate concentrations in surface water.

Sample locations are sorted based on distance to the mouth of Bear Creek at its confluence with EFPC, with the headwaters at the top of the graphic and the mouth at the bottom. Results are shown only for locations with at least 10 sample results. Quantile classification applied for visualization so that each class contains an equal number of data points. Graphic is not intended to show specific concentrations but shows spatial and temporal sampling patterns and relative concentrations throughout the watershed. The 'No result qualifier' column shows results within analytical method detection limits, whereas the 'Has result qualifier' column shows results below method detection limits or those with some other flagged issue.

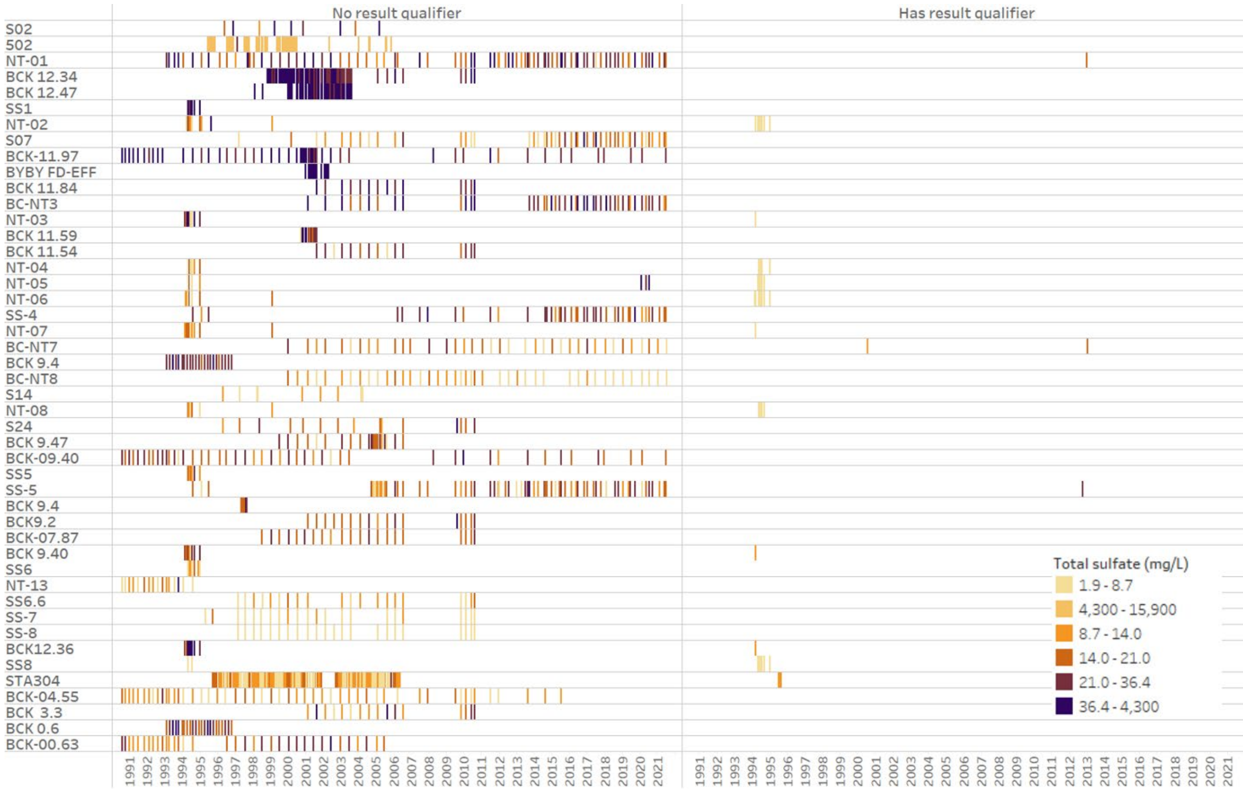

Figure 16. Total sulfate concentrations in surface water.

Sample locations are sorted based on distance to the mouth of Bear Creek at its confluence with EFPC, with the headwaters at the top of the graphic and the mouth at the bottom. Results are shown only for locations with at least 10 sample results. Quantile classification applied for visualization so that each class contains an equal number of data points. Graphic is not intended to show specific concentrations but shows spatial and temporal sampling patterns and relative concentrations throughout the watershed. The 'No result qualifier' column shows results within analytical method detection limits, whereas the 'Has result qualifier' column shows results below method detection limits or those with some other flagged issue.

3. RECENT AND ONGOING ASSESSMENTS

3.1 SPECIAL STUDIES

From 2017 to 2021, a series of limited, targeted field studies were conducted in Bear Creek to assess the biotic and abiotic factors contributing to increasing mercury concentrations in fish (Figure 17). The focus of these field studies varied from year to year, following the understanding of the important processes controlling mercury methylation and bioaccumulation, and beaver dams and periphyton were key areas of interest. Beaver dams create deep pools of slow-flowing water and organic matter accumulation, which has been correlated with increased MeHg concentrations. In addition, the deep pools created by beaver dams create habitat suitable for larger, longer-lived fish, which tend to accumulate mercury.

Periphyton biofilms are complex assemblages comprising algae, bacteria, fungi, detritus, extracellular polymers, invertebrates, and mineral particles (Mathews et al. 2021). Previous research in EFPC suggested that key controls on net MeHg concentration occurred in the stream or on the stream bed, and several lines of evidence pointed to benthic algal biofilms as central agents of MeHg production. Controlled laboratory experiments using field-derived periphyton biofilms and ambient creek water from EFPC have demonstrated that actively photosynthesizing biofilms may generate a significant fraction of the MeHg in EFPC (Olsen et al. 2016, Schwartz et al. 2019). In addition to being a source of MeHg production, these biofilms accumulate significant amounts of MeHg from their surroundings. Because periphyton biofilms form the base of the aquatic food web, the fact that they are a source of MeHg generation coupled with high biomagnification factors is of central importance to understanding and perhaps controlling MeHg concentrations in fish.

Studies in 2017–2018 focused on the role of beaver dams in contributing to mercury dynamics in Bear Creek, and limited sampling in 2019 addressed pools and fine-grained sediment deposits in previously unsampled locations. Sampling in 2020 and 2021 addressed the potential role that tributaries to Bear Creek may have on mercury and MeHg in the main channel. The 2020 sampling effort occurred over a 4.5 km reach from BCK 7 to BCK 11.5. This reach encompasses the EMDF location (Figure 17). The 2021 sampling effort extended from near the mouth of EFPC upstream to Bear Creek kilometer 12.4 and included all the sites sampled in 2020.

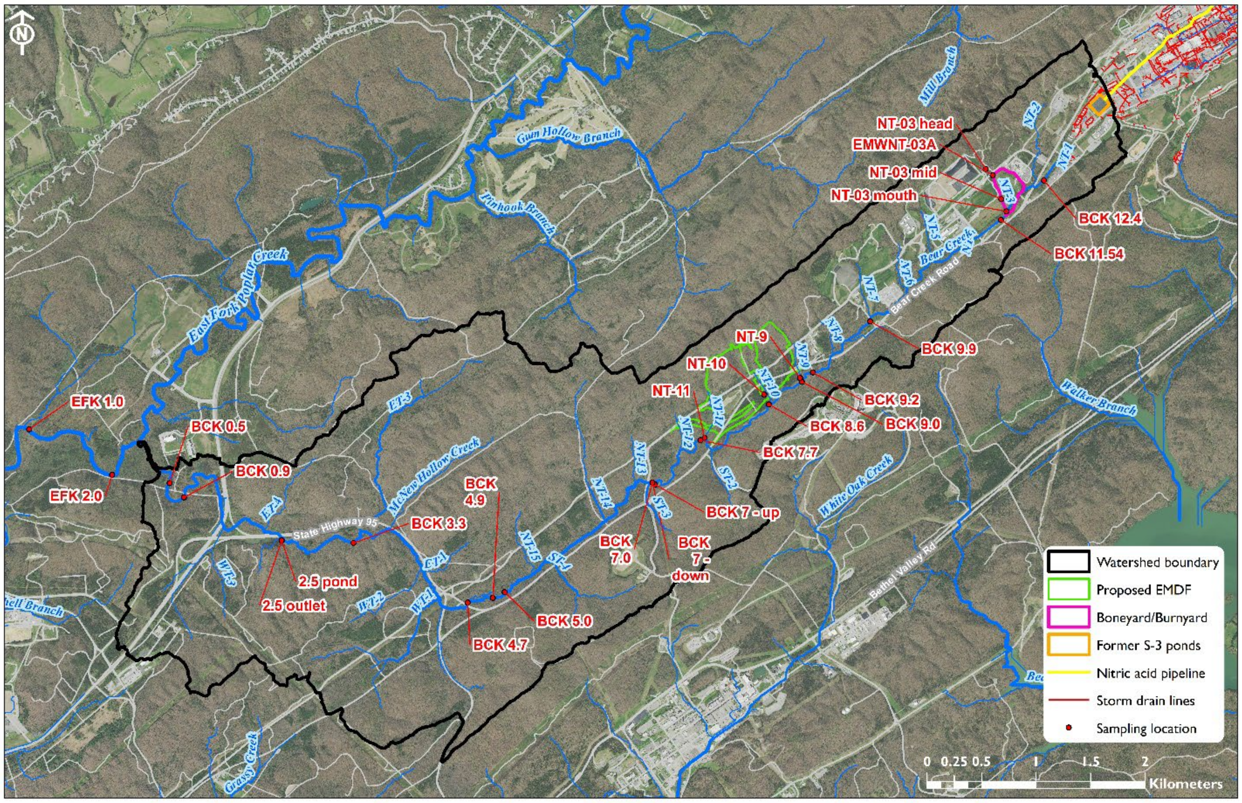

Figure 17. Special studies sampling locations.

3.1.1 Total and Dissolved Mercury

Total and dissolved mercury concentrations and particle-specific total mercury concentrations in Bear Creek are roughly 10× lower than those in EFPC (Figure 18). Tributary concentrations vary with location along Bear Creek. Concentrations in NT-03, near BCK 11.5, and in the beaver pond near the mouth (BCK 2.5) are higher than those in the main stem, whereas concentrations in NT-09, NT-10, and NT-11 are comparable to or lower than those in the main stem.

In general, concentrations of all three of these constituents slowly decline along Bear Creek from headwaters to the mouth of the creek (Figure 18). Nevertheless, there are notable downward departures from and subsequent recoveries to these trends between BCK 7 and BCK 10, the reach that encompasses the EMDF. The reason for the observed concentration decrease is unknown, although dilution caused by inputs of lower-concentration water sources may be a contributing factor (e.g., NT-08 confluence with Bear Creek just upstream of BCK 9.9). Diffuse groundwater inputs could contribute to the dilution. This reach was previously characterized as neutral to losing, meaning there was no net change in flow or a slight loss of water from the channel (Robinson and Johnson 1996), during a wetter-than-average year. More recent, detailed discharge measurements from which gaining, losing, or neutral conditions could be inferred are not available.

An open question is why concentrations increase at the end of this reach after the dip in concentration. Losing channel conditions would not affect the concentration, and there are no tributaries that could be adding mercury at that point. A potential explanation could be evaporative concentration. Concomitant increases in chloride, nitrate, and sulfate at the same location (Section 3.1.5) lend support to this mechanism. However, these observations do not eliminate the possibility of other, currently undefined additions of these solutes to the mainstem of Bear Creek. One approach to improve understanding of these dynamics would be to measure mass fluxes (concentration × discharge) at multiple locations along Bear Creek.

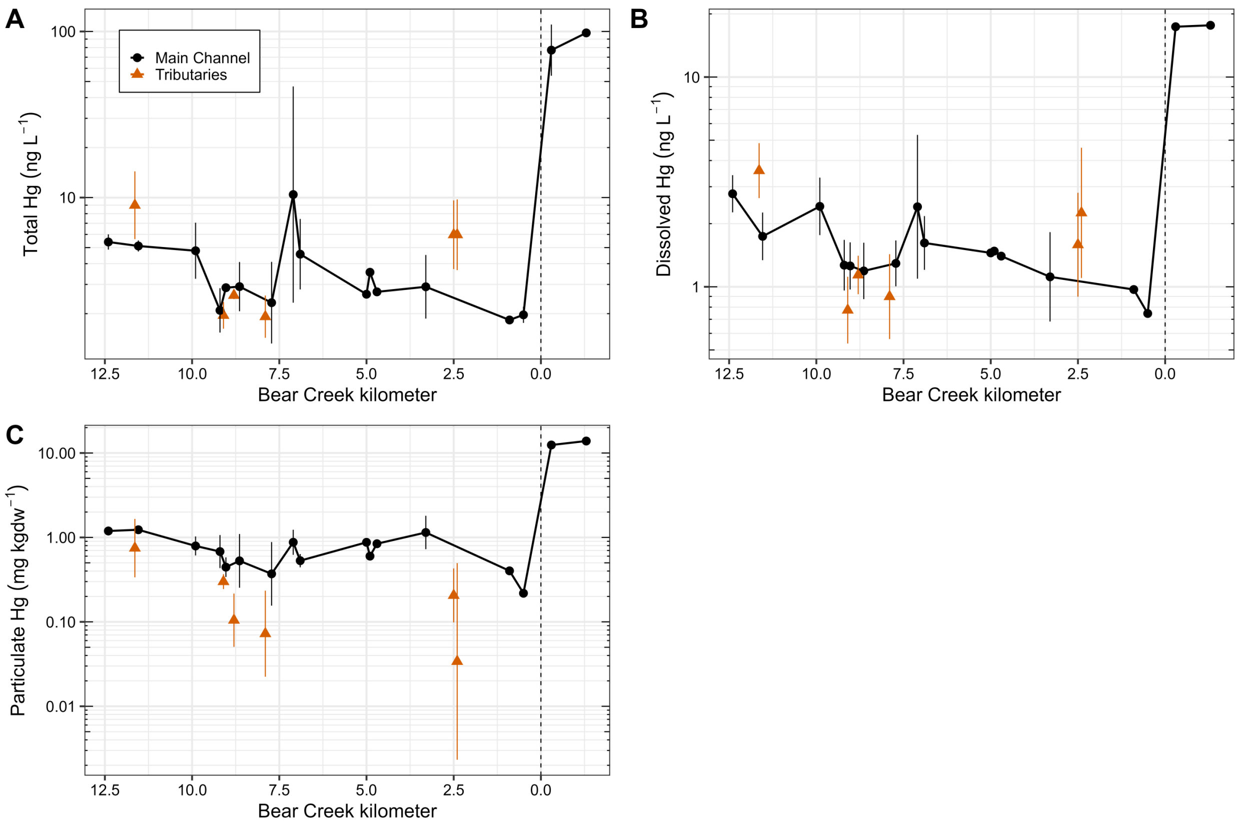

Figure 18. (A) Total mercury, (B) dissolved total mercury, and (C) particulate mercury concentrations along Bear Creek. Flow is from left to right on each panel. Filled circles represent samples from the main channel, and triangles represent samples from tributaries (geometric mean ± geometric SD of all results). Note the y-axis is log scale in each panel. The vertical dashed line marks the confluence of Bear Creek with EFPC.

3.1.2 Total and Dissolved Methylmercury

Total and dissolved MeHg concentrations measured in Bear Creek and its tributaries are seasonally comparable with those measured in the uppermost reaches of EFPC, which have the lowest concentrations in that creek (Figure 19). MeHg concentrations in tributaries to Bear Creek are comparable to or higher than those measured in the main stem.

In general, concentrations of total and dissolved MeHg and those of particle-specific total MeHg increase from upstream to downstream along Bear Creek. The influence of beaver dams on MeHg concentration is evident in the results for BCK 7 (former and current site of beaver dams) and at the long-established beaver pond along lower Bear Creek (BCK 2.5).

These results highlight an interesting parallel between Bear Creek and EFPC. In both streams, inorganic mercury concentrations decrease from upstream to downstream, whereas MeHg concentrations increase. Coupled measurements in Bear Creek of discharge and concentration to support mass flux calculations will improve understanding. For example, similar measurements and calculations in EFPC have demonstrated that MeHg flux increases from upstream to downstream, which indicates that the concentration increases cannot be attributed to accumulation or evaporative concentration effects.

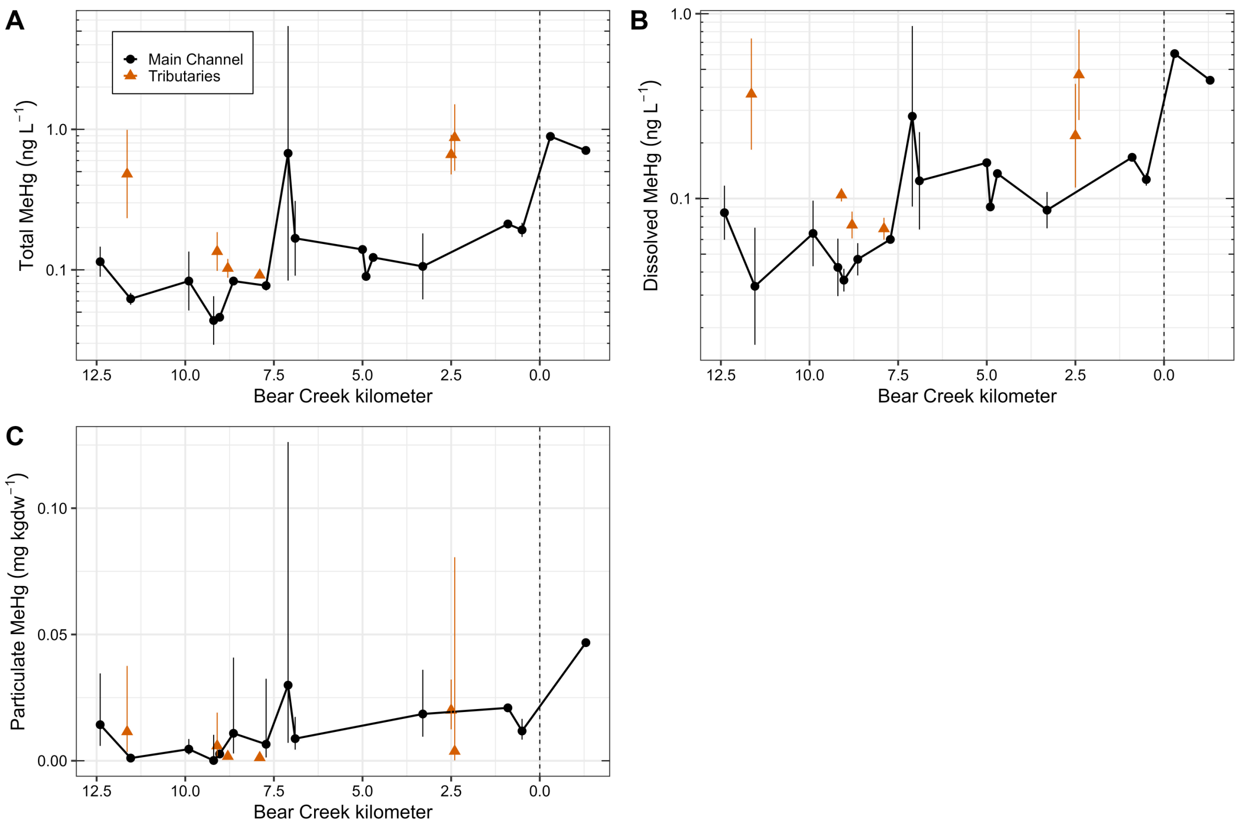

Figure 19. (A) Total MeHg, (B) dissolved MeHg, and (C) particulate MeHg concentrations along Bear Creek. Flow is from left to right on each panel. Filled circles represent samples from the main channel and triangles represent samples from tributaries (geometric mean ± geometric SD of all results). The vertical dashed line marks the confluence of Bear Creek with EFPC.

3.1.3 Sediments and Periphyton

Sediment mercury concentrations are highest at the upstream end of Bear Creek and decline almost exponentially with distance downstream to BCK 7.5 (Figure 20). Sediment concentrations are significantly higher at BCK 7, which was sampled at the time an active beaver pond was present. Sediment mercury concentrations then decrease steadily downstream. Total mercury concentrations in periphyton were highly variable in the upper half of Bear Creek and remained relatively steady from BCK 7.5 downstream. For both sediments and periphyton, mercury concentrations for samples collected from tributaries were comparable to those from the mainstem. At most sampling locations, Hg_T concentrations in periphyton were within 50% of that measured in sediments. However, at several sampling locations, periphyton mercury concentrations were more than 2× greater than in colocated sediment samples.

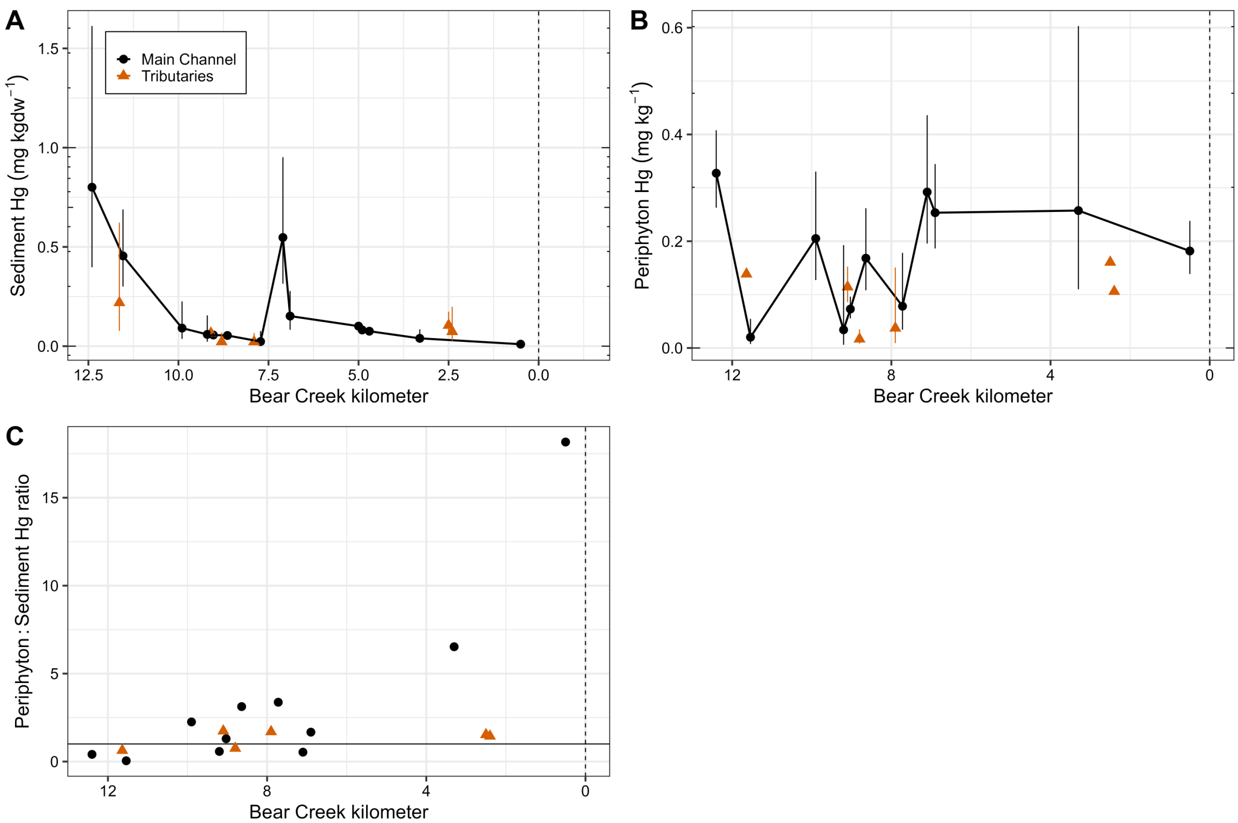

Figure 20. Total mercury in sediments (A) and periphyton (B) and periphyton to sediment concentration ratio (C) along Bear Creek. Flow is from left to right on each panel. Filled circles represent samples from the main channel, and triangles represent samples from tributaries (geometric mean ± geometric SD of all results, except C). The solid horizontal line indicates a 1:1 ratio. The vertical dashed line marks the confluence of Bear Creek with EFPC.

$MeHg_T$ in Bear Creek sediment decreases gradually downstream (Figure 21). Concentrations of MeHg in periphyton were higher in the lower half of Bear Creek compared to the upper half. For most sampling locations, MeHg concentrations in periphyton were more than 3× higher than those in collocated sediments, and in the lower portions of Bear Creek, MeHg periphyton:sediment concentration ratios exceeded 60×. Higher MeHg concentrations in periphyton relative to the sediments suggest that these biofilms are a source of MeHg production and/or selectively accumulate MeHg.

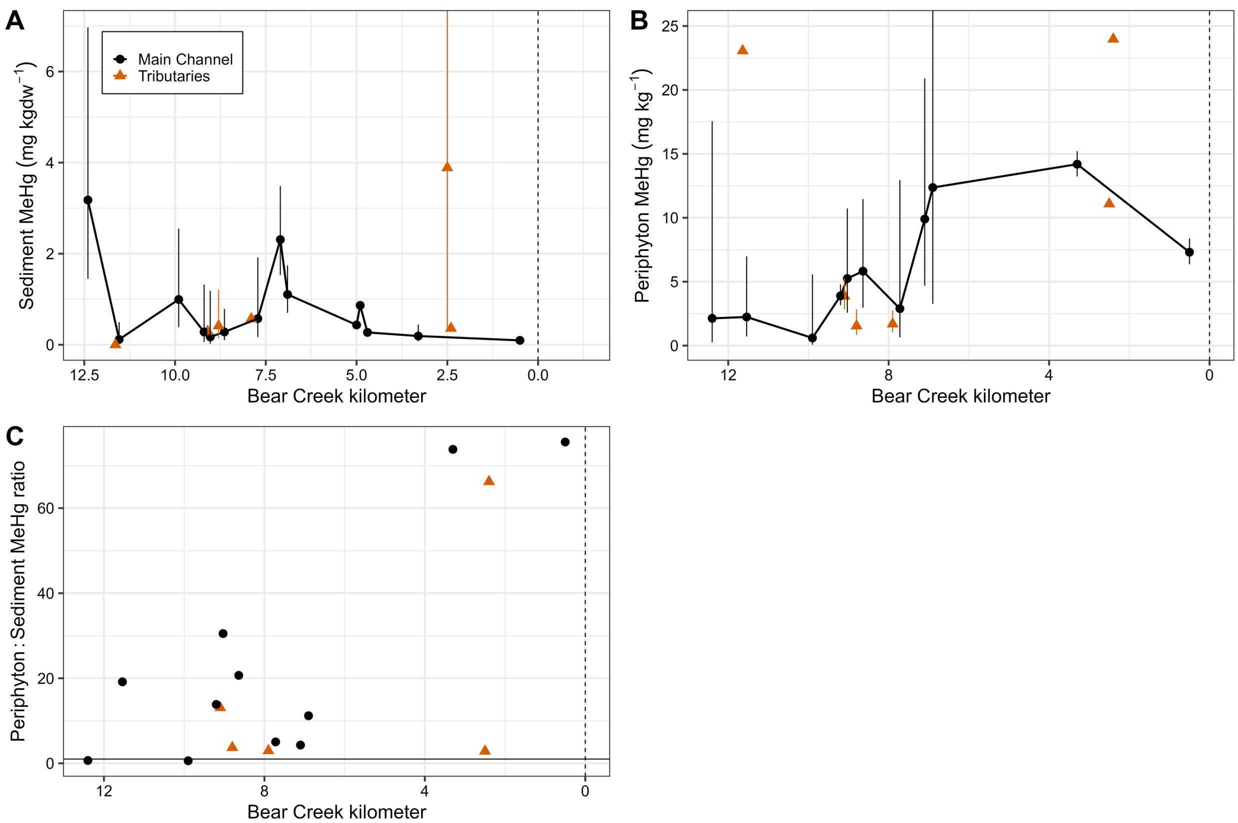

Figure 21. Total MeHg in sediments (A) and periphyton (B) and periphyton to sediment MeHg concentration ratio (C) along Bear Creek. Flow is from left to right on each panel. Filled circles represent samples from the main channel, and triangles represent samples from tributaries (geometric mean ± geometric SD of all results, except C). The solid horizontal line indicates a 1:1 ratio. The vertical dashed line marks the confluence of Bear Creek with EFPC.

3.1.4 Field Measured Parameters

Field measured parameters temperature, pH, and dissolved oxygen are typical for streams in this region. A well-documented contaminant plume discharges into the headwaters of Bear Creek, causing high specific conductance values that decrease rapidly downstream. Notably, temperature and specific conductance exhibit substantial drops between BCK 9.9 and BCK 9.2, supporting the suggestion that water sources between these sampling locations (e.g., groundwater sources) that discharge from NT-08 affect stream chemistry (see Section 3.1.1).

3.1.5 Anions and Trace Metals

As mentioned previously, a groundwater contaminant plume discharges into Bear Creek, elevating chloride and nitrate concentrations at the headwaters. The results of this study agree with previous observations of elevated nitrate and chloride levels near the headwaters that decrease with downstream distance (Figure 22). Chloride concentrations in NT-09 is substantially greater than those in Bear Creek and the other tributaries. Nitrate concentrations in NT-09, NT-10, and NT-11 were considerably lower than those in Bear Creek. The concentrations of chloride and nitrate decrease between BCK 9.9 and BCK 9.2. Sulfate concentrations were fairly consistent within the sampled reach and are elevated relative to NT-09, NT-10, and NT-11.

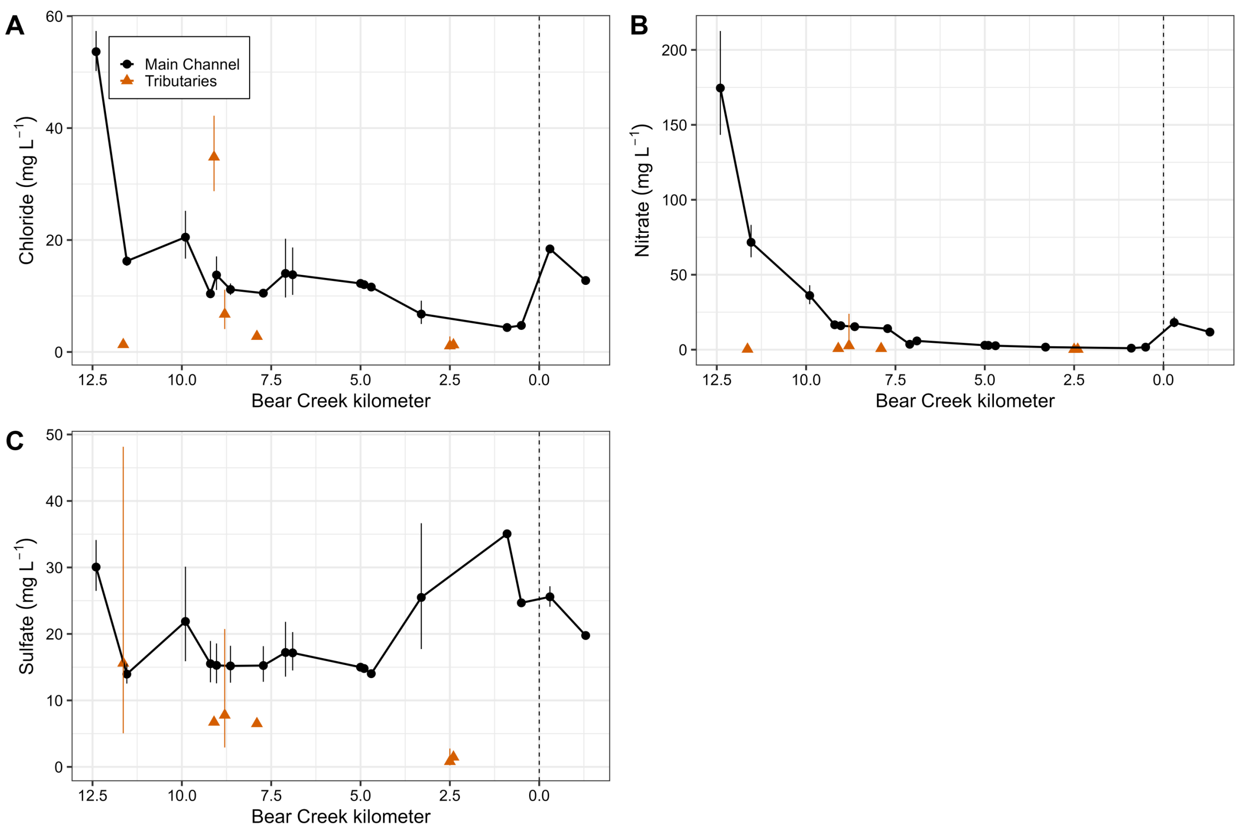

Figure 22. (A) Chloride, (B) nitrate, and (C) sulfate concentration along Bear Creek and in tributaries. Flow is from left to right on each panel. Filled circles represent samples from the main channel, and triangles represent samples from tributaries (geometric mean ± geometric SD of all results). The vertical dashed line marks the confluence of Bear Creek with EFPC.

3.1.6 Fish

Despite low aqueous mercury concentrations, mercury concentrations in sunfish collected in Bear Creek are high, though the past five years have seen precipitous declines in mean mercury concentrations in rock bass collected in Bear Creek such that mercury concentrations in fish in 2023 at BCK 3.3 and BCK 9.9 approached the EPA-recommended fish-based ambient water quality criteria of 0.3 µg/g. However, concentrations remain slightly elevated with respect to fish collected from the reference site (HCK 20.6; Figure 23). Decreases in fish tissue mercury concentrations have coincided with decreases in aqueous MeHg concentrations in Bear Creek. The decrease in aqueous MeHg concentrations and availability of larger fish could be due to the significant changes in habitat due to fluctuations in beaver activity over the past few years. The habitat through the middle-lower stretches of the stream had historically been poor for rock bass such that, in the early 2010s, this species could not be found, and redbreast sunfish were collected as a surrogate species. Starting in 2015, as beaver-impounded sections of this stream created deeper pools, rock bass were present in larger numbers and in larger sizes for bioaccumulation collection. The lack of large beaver dams in 2022 may have led to the smaller sizes of rock bass available for collection (Figure 23), but beaver activity was found to have increased in 2023 (Section 3.2). Future monitoring will demonstrate whether this decreasing trend in mercury concentrations in fish continues.

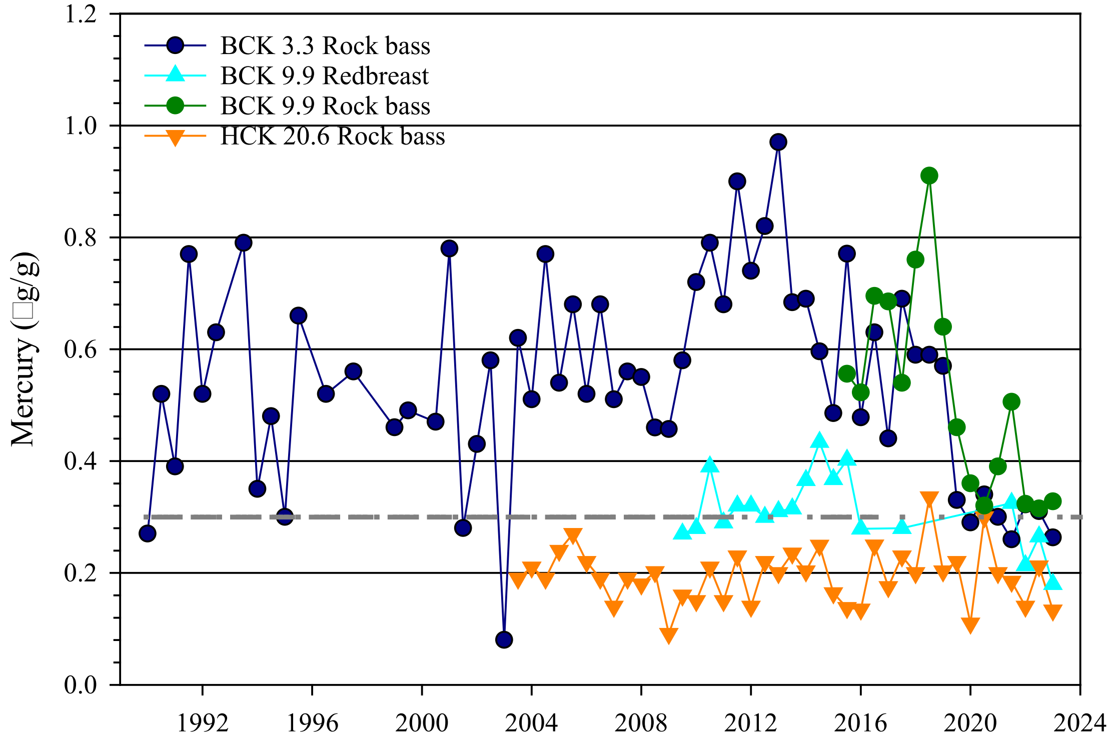

Figure 23. Mean concentrations of mercury in rock bass from BCK 3.3, redbreast sunfish and rock bass from BCK 9.9, and rock bass from the Hinds Creek reference site, 1990 to 2023. Dashed line indicates EPA-recommended ambient water-quality criterion for mercury (0.3 µg/g in fish).

3.2 BEAVER ACTIVITY

Beaver dams create deep pools of slow-flowing water and organic matter accumulation, which has been correlated with increased MeHg concentrations. Beaver dams and other interruptions to the natural flow of Bear Creek were surveyed between October and December 2021 and February 2022 (Figure 24). Surveys began approximately 150 m downstream from the State Route 95 bridge upstream to Burial Ground Access #4 Road near the Y-12 West Portal. Additional surveys were conducted in January and August 2023, approximately 250 m upstream from Reeves Rd. downstream to State Route 95 (Figure 25). For both sets of surveys, the location of beaver dams, lodges, and other beaver activity was recorded using a handheld Trimble Geo 7x. All beaver activity was photographed where possible. Additional sources of flow interruption such as log jams and blocked culverts were also recorded. Since February 2022, beaver activity has increased substantially in the vicinity of Reeves Rd. down to Gum Branch Rd. This includes at least 11 new dams since February 2022.

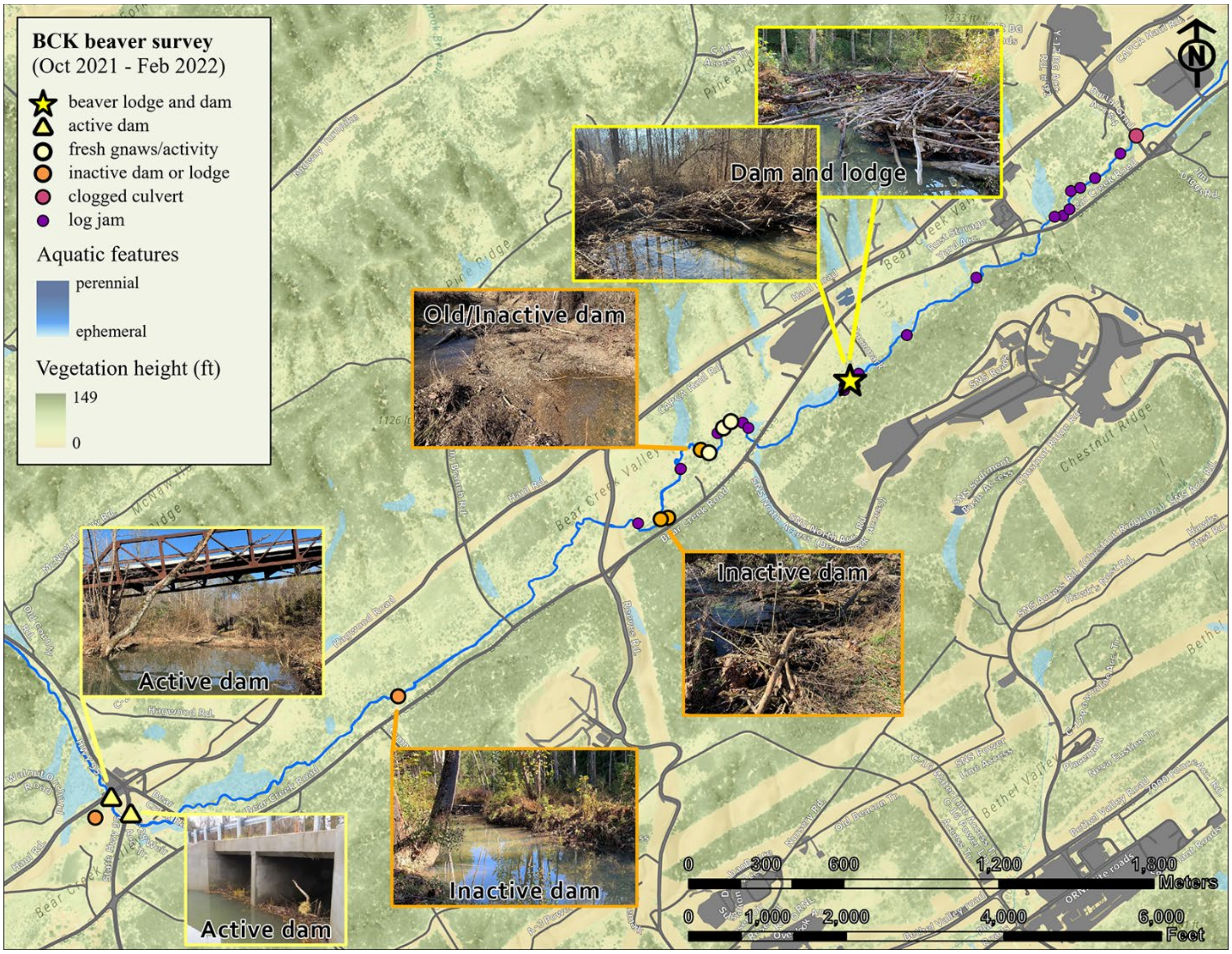

Figure 24. An assessment of beaver activity in Bear Creek October 2021 – February 2022.

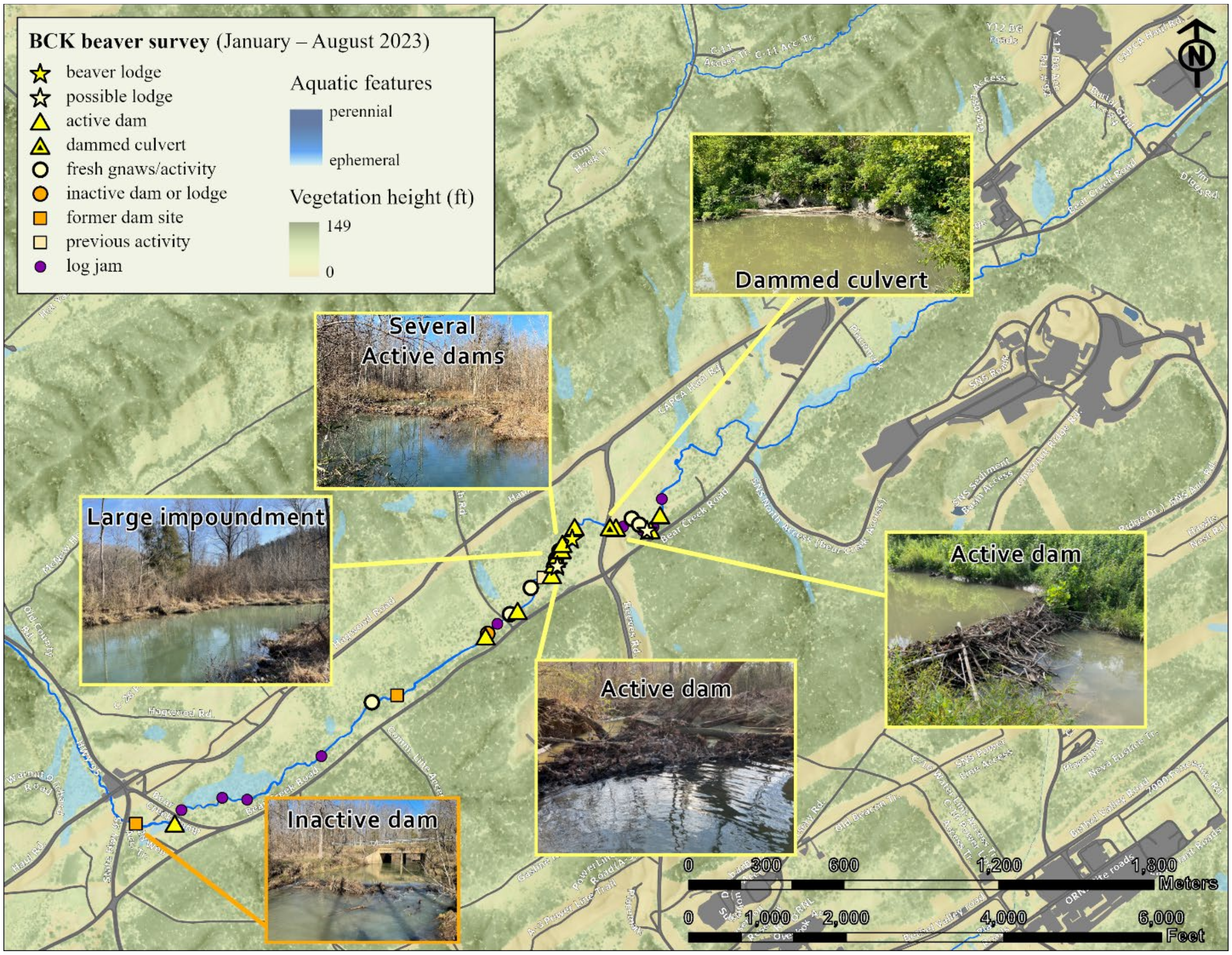

Figure 25. An assessment of beaver activity in Bear Creek in January and August of 2023.

In addition to the field surveys, we also compiled an archive of airborne and satellite imagery we thought could potentially be of use for mapping beaver impoundments in the watershed over time. Although we were able to compile 37 images from 1972 through 2022 from across various sources that have an average spatial resolution of approximately 1.1m (Table 1), Bear Creek is small enough that creek features, including small impoundments, are generally still difficult to distinguish in these relatively high-resolution airborne and satellite images. Further visual review of these images is warranted to fully determine their usefulness for our purposes.

Table 1. Bear Creek watershed imagery.

Date	Leaf	Number of Bands	Source	System	Spatial resolution (m)	Image quality
4/15/1972	off	3	USGS	Airborne	3	good
3/12/1981	off	3	USGS	Airborne	1.5	good
2/23/1997	off	1	USGS	Airborne	1	good
2000	off	1	State of TN	Airborne	0.6	good
9/29/2004	on	4	Quickbird	Satellite	2.5	low
9/29/2004	on	1	Quickbird	Satellite	0.65	good
2006	off	3	ORNL	Airborne	0.25	good
12/8/2006	off	3	NAIP	Airborne	1	good
2007	off	3	TDOT	Airborne	0.3048	good
9/18/2007	on	3	NAIP	Airborne	1	good
4/17/2008	off	4	NAIP	Airborne	1	good
1/1/2009	off	4	GeoEye-1	Satellite	2	low
1/1/2009	off	1	GeoEye-1	Satellite	2	good
1/1/2010	off	8	WorldView-2	Satellite	2	good
4/11/2010	on	4	NAIP	Airborne	1	good
5/24/2010	on	4	GeoEye-1	Satellite	1.7	low
10/28/2010	on	8	WorldView-2	Satellite	2	good
2011	off	3	TDOT	Airborne	0.3048	good
1/4/2012	off	1	WorldView-1	Satellite	0.5	good
3/21/2012	off	1	WorldView-1	Satellite	0.5	good
5/6/2012	on	4	Quickbird	Satellite	2.5	good
5/27/2012	on	4	NAIP	Airborne	1	good
2013	on	3	ORNL	Airborne	0.25	good
4/27/2014	on	4	NAIP	Airborne	1	good
9/20/2014	on	8	WorldView-2	Satellite	2	good
2015	off	3	TDOT	Airborne	0.3048	good
8/1/2015	on	3	NEON	Airborne	0.25	good
4/24/2016	on	4	NAIP	Airborne	1	good
6/7/2016	on	3	NEON	Airborne	0.1	good
10/24/2018	on	4	NAIP	Airborne	1	good
2019	off	3	TDOT	Airborne	0.3048	good
2/10/2021	off	3	WorldView-3	Satellite	0.3	good
4/3/2021	off	4	NAIP	Airborne	1	good
6/1/2021	on	3	NEON	Airborne	0.1	good
7/6/2021	on	8	WorldView-2	Satellite	2	good
7/22/2021	on	8	WorldView-2	Satellite	2	good
4/3/2022	off	8	WorldView-2	Satellite	2	Good

Leaf column indicates whether imagery was collected during leaf-on or leaf-off conditions. Source indicates agency or imagery program responsible for collecting imagery. System indicates whether imagery was collected via airplane or satellite. Spatial resolution indicates imagery pixel size.

3.3 SPATIAL MODELING OF PERIPHYTON

Traditional methods for the quantification of periphyton distribution and abundance in the field are time-consuming and labor-intensive. Even the most ambitious sampling campaigns will have large spatial gaps, requiring tenuous interpolation and extrapolation methods to fill those gaps but introducing significant uncertainty (Mathews et al. 2021). Recent advances in in situ fluorescence-based techniques and portable field instruments provide an opportunity to better estimate the distribution and abundance of periphyton-associated algal biomass in the field (Catherine et al. 2012).

One approach to address the scaling issue associated with the labor-intensive periphyton field data collection is to use remote sensing approaches in combination with field data collection to map periphyton distribution and abundance. Near-surface remote sensing via unmanned aircraft systems (UAS) is a cost-effective (Sankey et al. 2017) and flexible alternative for targeted data collections in smaller study areas such as the Bear Creek watershed. Periphyton distribution and abundance in streams are driven by light availability and shading, water depth, substrate type, water velocity, nutrient concentrations, and water temperature. These conditions, especially shading, water depth, substrate, and water velocity, can vary widely within small stretches of stream, so collecting remote sensing data at a very high resolution is necessary to understand, model, and map periphyton dynamics.

As part of the Mercury Remediation Technology Development (TD) program, we are actively researching spatial modeling approaches and their associated data needs to help us understand the most influential drivers of periphyton distribution and abundance in Bear Creek and EFPC (Mathews et al. 2021). To date, we have collected and analyzed lidar, hyperspectral, multispectral, and sub-canopy true color UAS data, and we have augmented this remote sensing data with lab and field studies to map/model periphyton distribution and abundance in sections of Bear Creek. Our focus this FY was on improving our UAS and field data stream for substrate and habitat mapping (Figure 26), light availability/shading mapping, water depth mapping, and instream sample collection to inform periphyton spatial modeling (Figure 27). Modeling that can inform management decisions on watershed, riparian corridor, and instream habitat management to reduce MeHg production is ongoing under the TD program and should improve our understanding of periphyton drivers and how they fit into the mercury conceptual model.

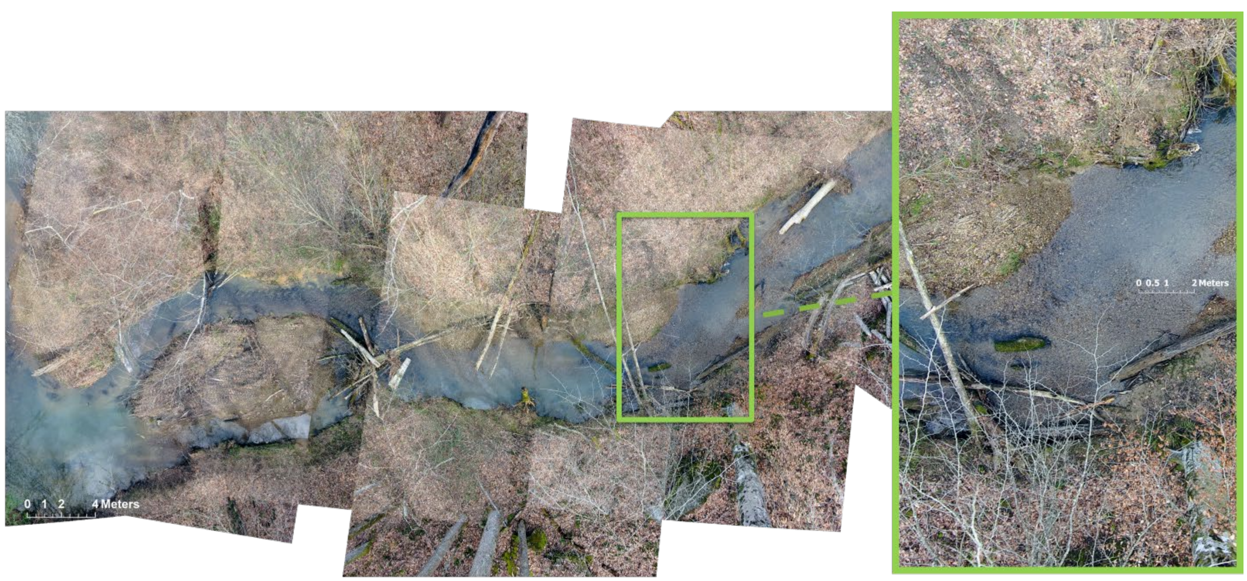

Figure 26. Closeup of ~75 m section of Bear Creek subcanopy imagery collected on March 1, 2023. The spatial resolution of 0.3 cm achieved by flying 10 ft above the stream channel allows for mapping instream features with exceptional detail.

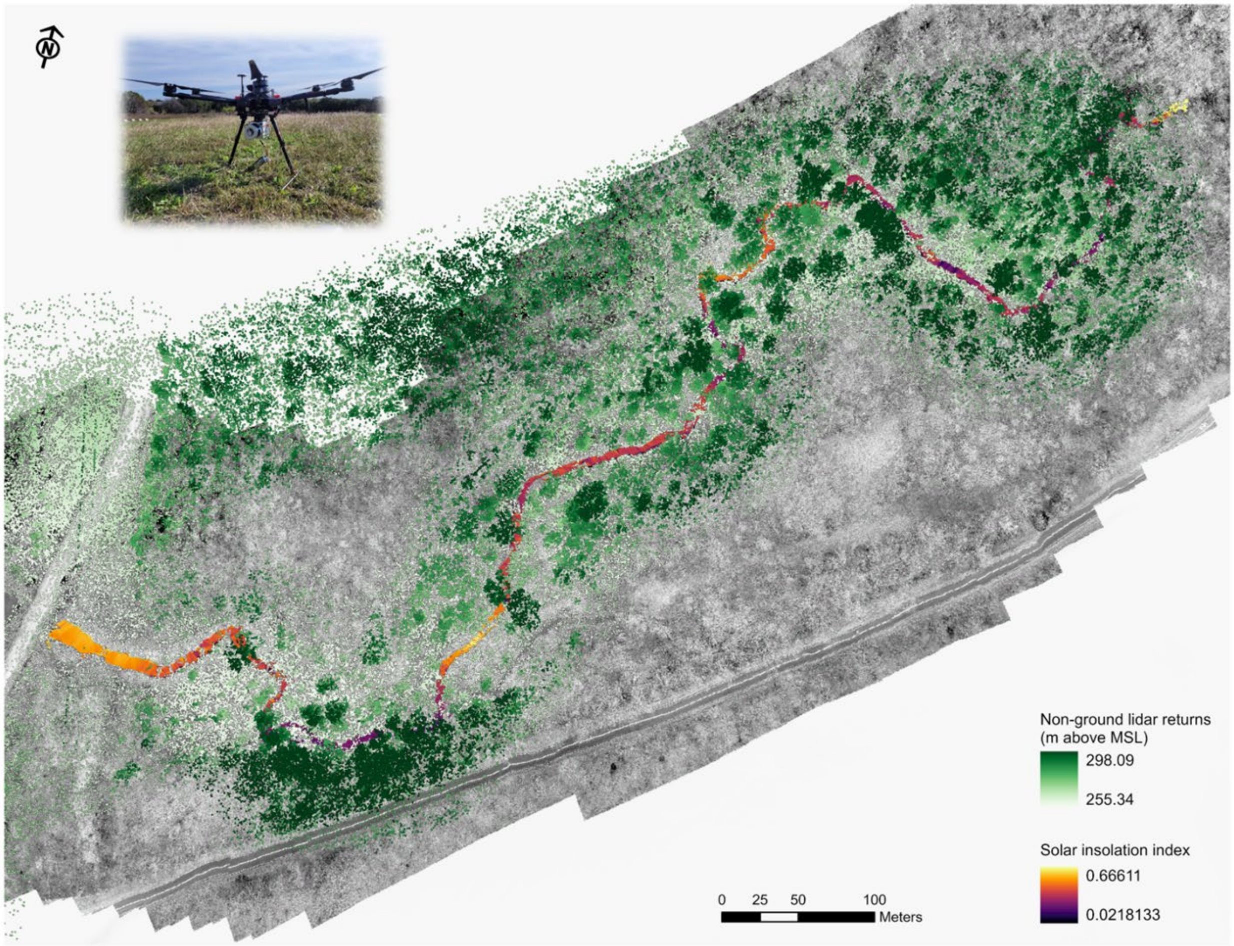

Figure 27. Non-ground lidar point cloud returns and solar insolation index.
Data collected on March 1, 2023, in Bear Creek. Non-ground returns shown in green, with darker green indicating taller trees. Solar insolation index shows estimate of solar exposure throughout stream channel, with higher values indicating higher exposure. Phoenix Lidar onboard the DJI M600 shown in inset.

3.4 WATERSHED MODELING

It is essential to accurately identify and measure the sources of mercury and understand how it moves through a contaminated watershed to downstream environments. Therefore, an analysis of mercury sources and transportation routes in the Bear Creek watershed has been initiated. The goal is to assess the factors that influence mercury movement from upstream to downstream using a conceptual approach. Creating a Bear Creek watershed model will aid in forming a conceptual model for mercury movement. This will be done by quantifying the factors that influence the transportation of mercury from upstream to downstream in a spatially explicit manner.

We are using the Soil Water Analysis Tool (SWAT), a widely recognized watershed model, to model conditions in the Bear Creek watershed. This model can be adapted to include natural and human factors that affect surface flow and quality (Gassman et al. 2007). Our goal is to create a SWAT model that can simulate streamflow and suspended solid transport in the watershed to identify the pathways of mercury pollution (Nair et al. 2020). The SWAT model is a continuous-time simulation model that is based on the physical characteristics of the watershed and integrates climate, landscape, and hydrology on a daily and sub-daily time scale. It also allows for analyzing flow and sediment management options (Arnold et al.

1998). Using a digital elevation model (DEM), the model divides watersheds into smaller sub-watersheds, which are then classified based on soil type, land use, and slope to create hydrologic response units (Figure 28). The model accurately simulates the water and nutrient budget for all response units and considers human activities like water extraction, reservoir storage, and diversion. This allows us to better understand flow and sediment management options on a watershed scale.

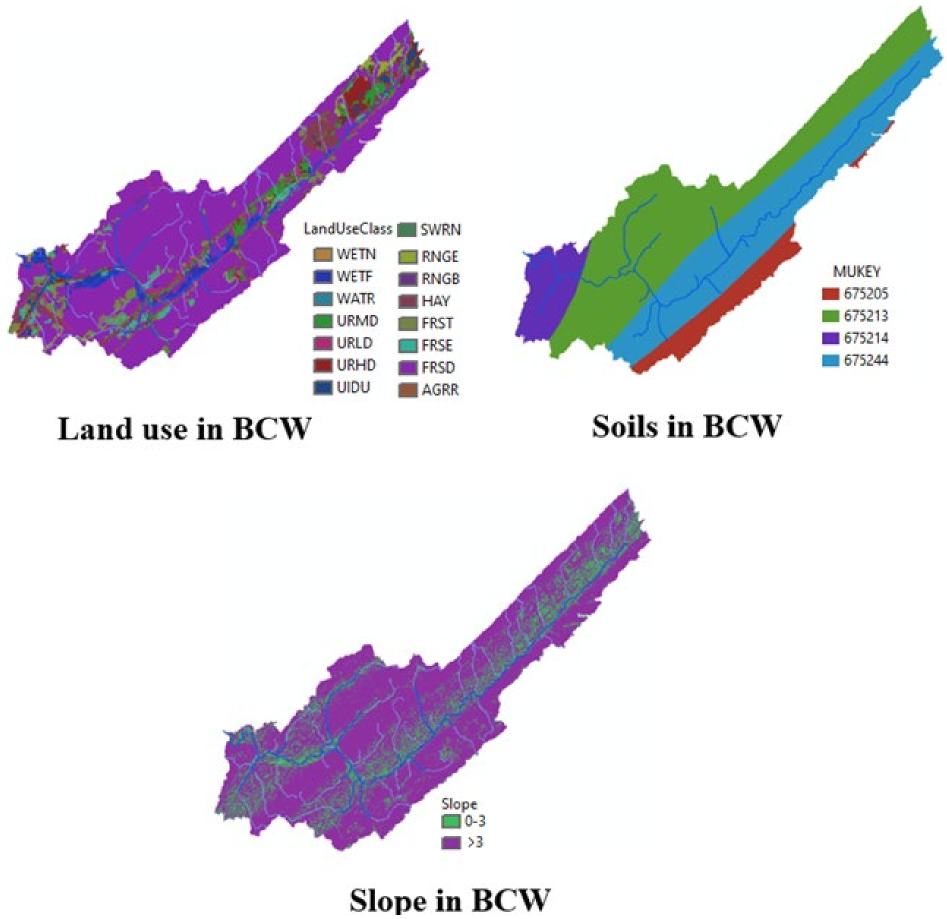

Figure 28. Land use, soils, and slope maps of the Bear Creek watershed.

Our hypothesis is that by using the SWAT model to simulate surface flow and sediment load in the Bear Creek watershed, we can derive valuable information that will help us connect different system components in a conceptual model for the transport of mercury. This modeling exercise can also uncover specific data gaps in the watershed that impede our understanding of the factors that drive mercury flux dynamics.

Data requirements for the initial setup of the SWAT model are shown in Table 2. A high-resolution (0.72 m) LiDAR-derived DEM and a high-resolution (1:24,000 scale) stream network from the National Hydrography Dataset were used to develop the SWAT model. Using the National Land Cover Dataset and medium-resolution (1:250,000 scale) STATSGO soil data, the SWAT model delineated 40 sub-watersheds and 91 hydrologic response units (HRUs) in the watershed (Figure 29).

Weather variables (e.g., temperature, precipitation, solar radiation, humidity, and wind velocity) drive the water budget, which drives suspended solid transport in the watershed. We used thirty years (1990–2019) of measured or modeled weather variables from within or nearby the Bear Creek watershed as inputs for the SWAT model. The daily measured precipitation from the ORR (Figure 29) was used along with temperature, solar radiation, and vapor pressure data from the DAYMET dataset as weather inputs to the model. Additionally, water flow from seeps, flumes, and springs must be accounted for in watershed modeling (Figure 30). The measured flow and sediment loading from different instream points were obtained from the OREIS database, which will be used to calibrate and validate the flow and sediment load of the SWAT model. There were 41 instream flow measuring locations in the watershed with a varied length of record and 13 instream flow measurement locations with enough data points for calibration of the model (Table 3).

Table 2. Data inputs and sources for initial SWAT setup.

Data Input	Spatial resolution	Source
Topography (DEM)	0.72 m	TNGIS[1]
Climate		Oak Ridge Reservation[2]
Soils	Medium	STATSGO USDA[3]
Land use	30 m	USGS[4]
Stream network	High	NHDPlus V2[5]

1. http://tngis.org/lidar.htm
2. https://metweb.ornl.gov/page4.htm
3. https://datagateway.nrcs.usda.gov/
4. https://www.mrlc.gov/data
5. http://www.horizon-systems.com/NHDPlus/NHDPlusV2_home.php

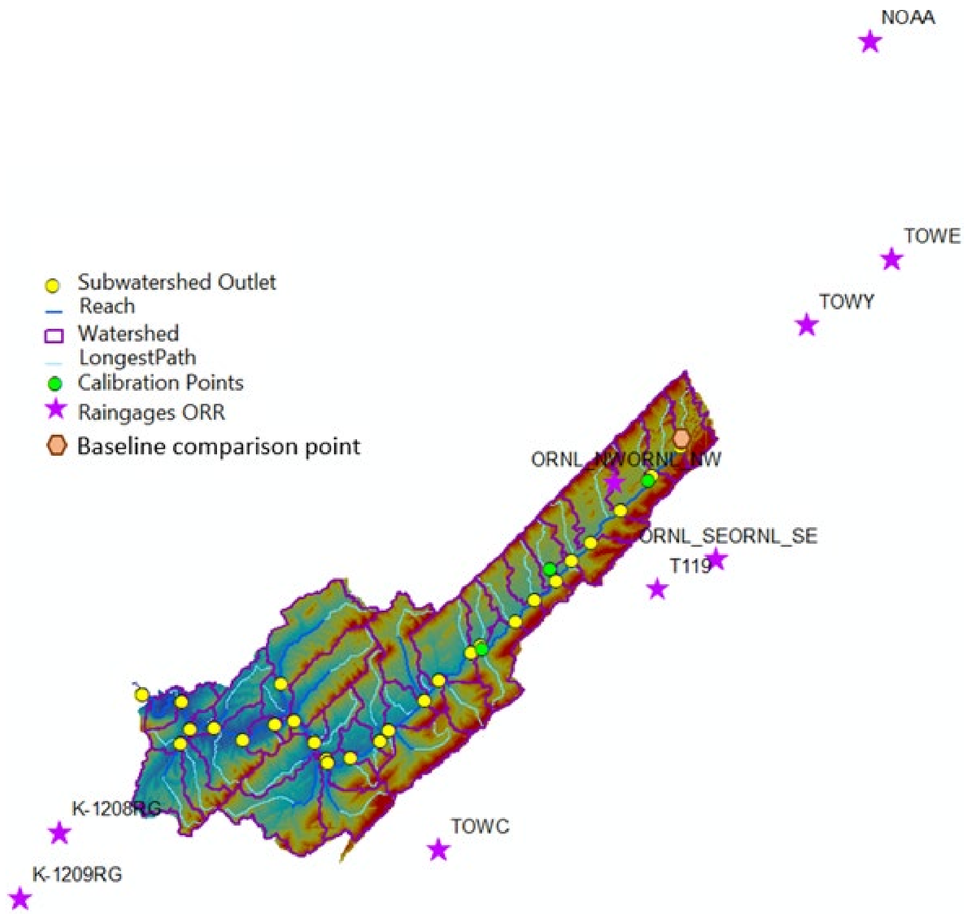

Figure 29. Delineated watershed and sub-watersheds along with locations of the measured precipitation inputs used in the SWAT model.

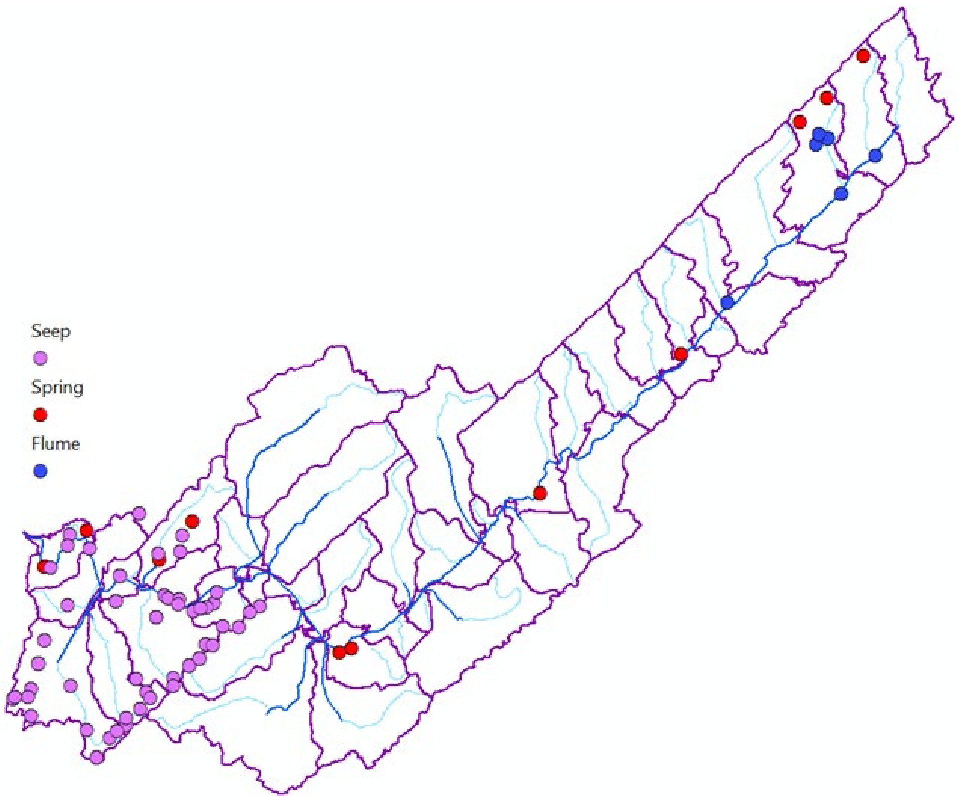

Figure 30. Spatial distribution of seeps, springs, and flumes in the Bear Creek watershed.

Table 3. Instream flow monitoring points in the Bear Creek watershed.

Monitoring point	Start date	End date
BCK 11.54	7/18/2001	10/15/2018
BCK 11.84	7/4/2001	8/30/2010
BCK 12.34	**10/1/1999**	**9/30/2021**
BCK 12.47	10/1/2000	9/30/2003
BC-NT3	10/1/1999	9/30/2021
BC-NT8	10/10/2007	9/30/2021
BCK9.2	**10/1/2001**	**9/30/2021**
NT-05	10/17/2007	9/30/2021
S02	8/10/1995	1/24/2006
S06	10/11/1995	9/15/2021
S08	10/5/1995	5/31/2006
S24	7/1/1995	6/29/2006
STA304	7/1/1995	12/14/2005

Shaded rows show monitoring points with sufficiently long data record for calibration.
BCK 12.34 and 9.2 used for calibrating the SWAT-BCK modeled flow.

3.4.1 Calibration Run Results

To accurately calibrate the SWAT model for precipitation flow simulation, we needed to determine the amount of water in the stream that did not come directly from rainfall runoff. This water could have flowed into the stream through natural means such as seepage, flumes, springs, or human intervention. To estimate the excess water, we relied on data from two monitoring points, namely BCK 9.2 and BCK 12.34. Using the flow data from these points, we could compare the flow rate at BCK 12.34 to the flow rate at BCK 9.2 through regression analysis. Despite these sites being about 3 km apart and receiving similar rainfall, the analysis revealed that the flow rate of BCK 9.2 could be up to six times greater than the flow rate of BCK 12.34 (Table 4). Therefore, water other than runoff enters the stream between 12.34 and 9.2.

We followed the method given by Nair et al. (2020) to quantify the excess water, which was added as the outflow from corresponding sub-watersheds in SWAT. The curve numbers (CN2) were decreased by 10% for all HRUs in the basin to adjust the runoff, which essentially increases water movement into the soil profile. The soil available water capacity (SOL_AWC) was increased by 10% to augment the water storage in the soil profile, permitting the water entering the soil to be available to compensate for the evapo-transpiration demand for the vegetation in the watershed. The parameter that controls daily runoff as a fraction of total available water, the surface runoff lag coefficient, was changed to 1 to account for lagged flows from a rainfall event.

Table 4. Results of regressing flow at BCK 12.34 against flow at BCK 9.2.

	Beta value	t-value
Intercept	0.0118	7.2834
Flow at 12.34	5.9614	112.502
Adj-R^2	0.71	

The results of the flow calibration are displayed in Figure 31–Figure 34. The calibration accurately captured the flow changes and produced high values for key performance indicators, namely the Nash–Sutcliffe coefficient of efficiency (E) and R^2 value of monitored flow compared to the simulated flow. These indicators were consistently high for both daily and monthly flow evaluations. The two monitoring points used for calibration are in the upper portion of the watershed. It is best practice for calibration and validation of a SWAT model to have flow values from both upstream and downstream in the watershed. Unfortunately, we lack flow data in the downstream portion of the watershed, so the current calibrated SWAT model can account only for natural flow generation processes and may not capture the heterogeneity in upstream-to-downstream flow generation and progression downstream of BCK 9.2.

SWAT model development is ongoing and will be used as an integrator for water inputs and outputs to the watershed, landscape influences, and instream dynamics to inform conceptual model development.

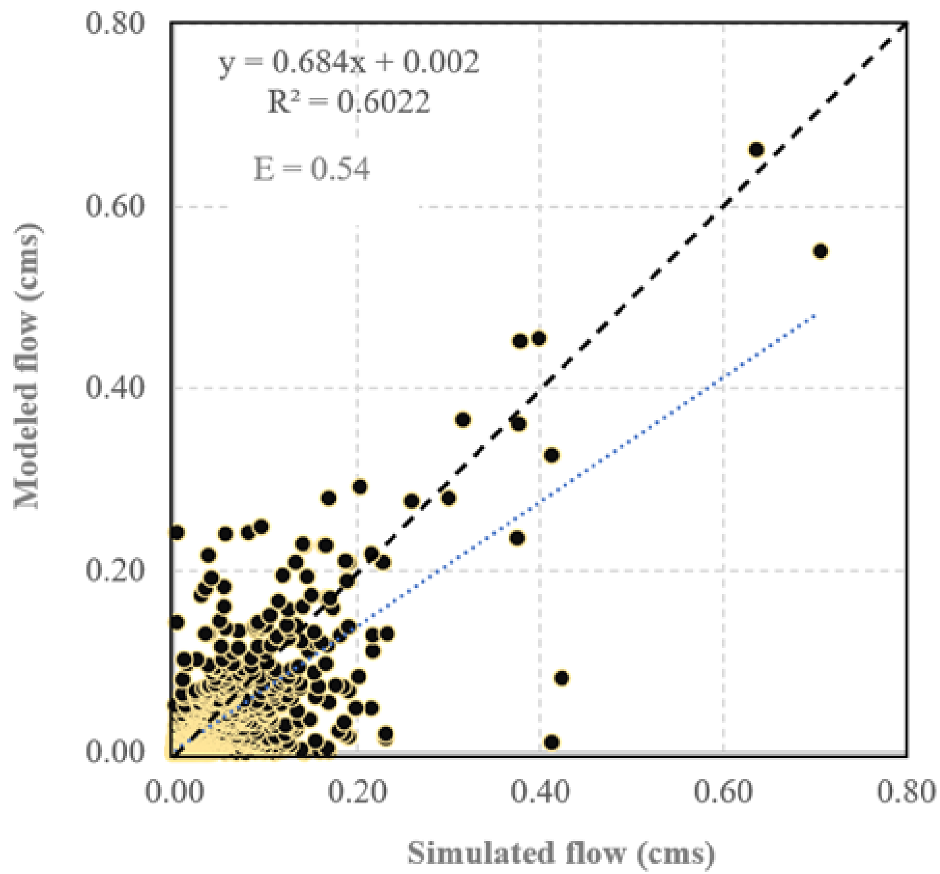

Figure 31. Comparison of daily flow of the calibrated model against measured flow for BCK 12.34.

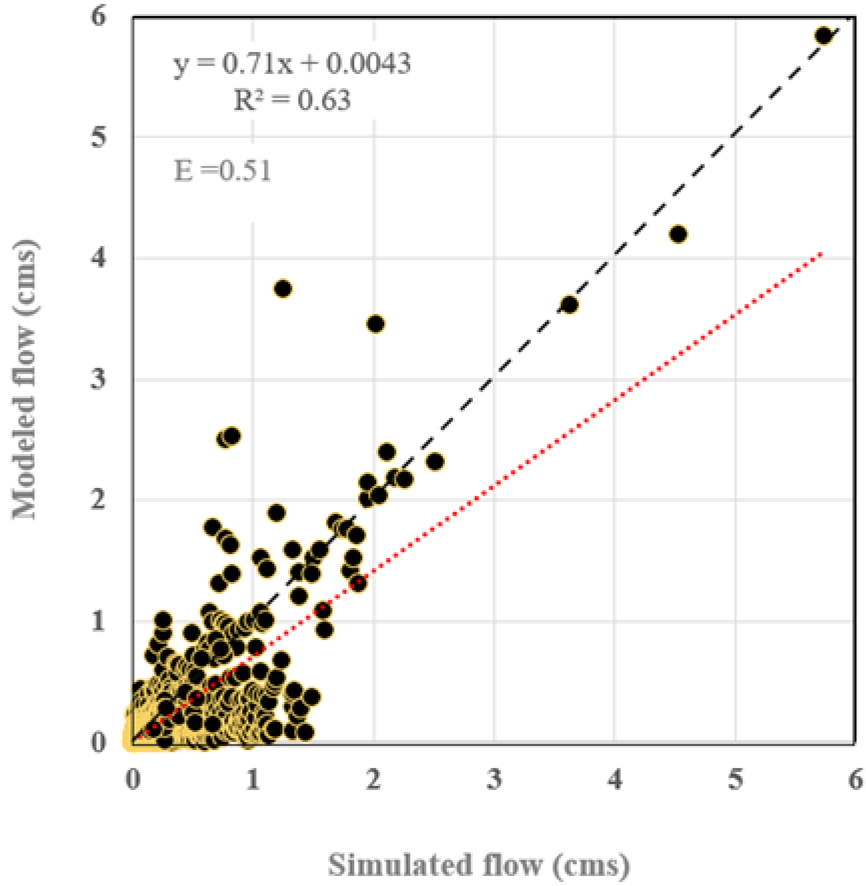

Figure 32. Comparison of daily flow of the calibrated model against measured flow for BCK 9.2.

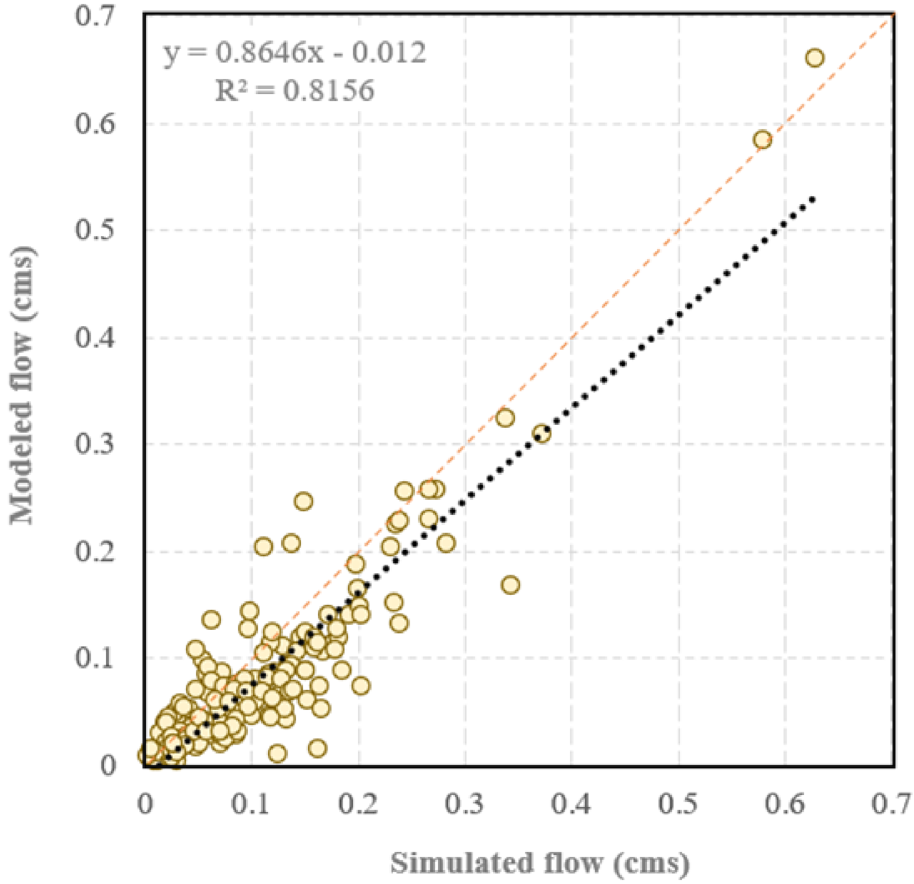

Figure 33. Comparison of monthly flow of the calibrated model against measured flow for BCK 12.34.

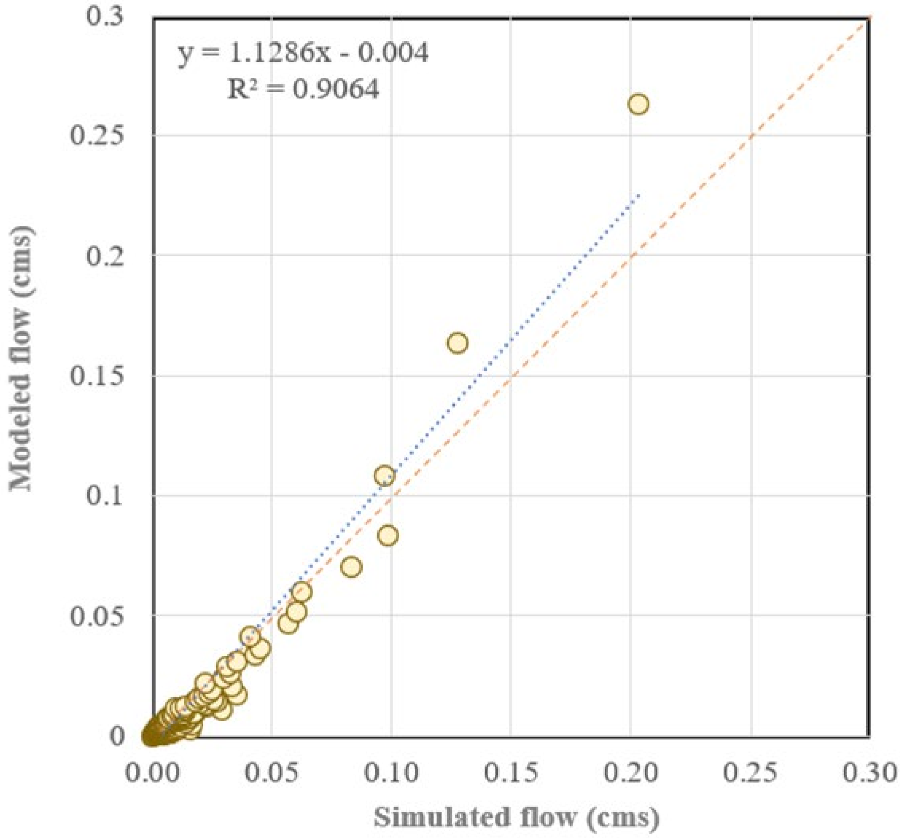

Figure 34. Comparison of monthly flow of the calibrated model against measured flow for BCK 9.2.

4. FUTURE WORK

Additional activities related to construction and operation of the EMDF include an RSE to be conducted in early FY24. The broad goal of that activity is to “evaluate potential source of mercury and methylmercury within the Bear Creek valley watershed” (draft Bear Creek Valley Mercury Sources Remedial Site Evaluation Sampling and Analysis Plan Oak Ridge, Tennessee). The DQO planning for the RSE followed the seven-step iterative process outlined by the EPA in EPA/240/B-06/001 (2006) to determine a sample collection design to meet stated goals and objectives of the activity. Briefly, the RSE includes sampling floodplain and bank soils, creek sediments, and water at fifteen cross-channel transects along Bear Creek and one transect at a reference site. Quantifying total mercury and MeHg concentrations is the priority, and many ancillary parameters and properties will also be measured to help interpret and give context to the results. Sampling locations are being selected specifically to assess the effects of beaver dams. Results of the RSE will be incorporated into development of the conceptual model.

As summarized throughout this report, there are an impressive number of both water quality and discharge measurements along Bear Creek. Nevertheless, there are far fewer *coordinated* water quality plus discharge measurement campaigns with a specific emphasis on mercury and MeHg. Additionally, we are not aware of any event-based (i.e., precipitation-driven floods), coordinated discharge and water quality sampling campaigns. Solute concentration data are valuable for describing water quality but are limited in the sense that concentration changes, in the absence of accompanying concurrent discharge data, cannot be used to distinguish simple accumulation from production, evaporative concentration from new source additions, and so on. We have successfully coupled concentration and discharge data under baseflow and flood events in EFPC to identify key source areas contributing mercury and MeHg to the creek and to demonstrate mass loading even as concentrations decrease (Riscassi et al. 2016, Mathews et al. 2021). This lack of concentration–discharge data will be partially addressed with the installation of a new gauging station in lower Bear Creek (~BCK 1). The BCK 1 station is under construction at this time and will be equipped with a pressure transducer and multiparameter sonde (temperature, pH, specific conductance, dissolved oxygen, nitrate). These continuous monitoring instruments will need to be supplemented with manual sampling for mercury and MeHg or other constituents. Additionally, these measurements provide flux data at one location—to assess loading will require one or more additional locations along Bear Creek. We advocate for dedicated coupled concentration–discharge measurements under both baseflow conditions and over several flood hydrographs.

Future work will build on the summary presented in this report to develop a conceptual model that outlines the key environmental parameters that correlate with MeHg concentrations and bioaccumulation in Bear Creek. A conceptual model will help to provide an understanding of the processes affecting mercury transport and transformation in the Bear Creek watershed and will continue to highlight key knowledge gaps in our understanding of these processes. This conceptual model will provide a strong technical basis for prioritizing and optimizing potential mitigation actions or best management practices to minimize mercury methylation in a cost-effective and efficient manner.

5. REFERENCES

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