

# Reducing Acidification in Endangered Atlantic Salmon Habitat

## Third Year of Clam Shells

*April 2022*

Contact: Emily Zimmermann, Biologist  
Phone: (207) 446-1003



Photo credit: Tanya Rucosky, DSF

MAINE DEPARTMENT OF ENVIRONMENTAL PROTECTION  
17 State House Station | Augusta, Maine 04333-0017  
[www.maine.gov/dep](http://www.maine.gov/dep)



## Introduction

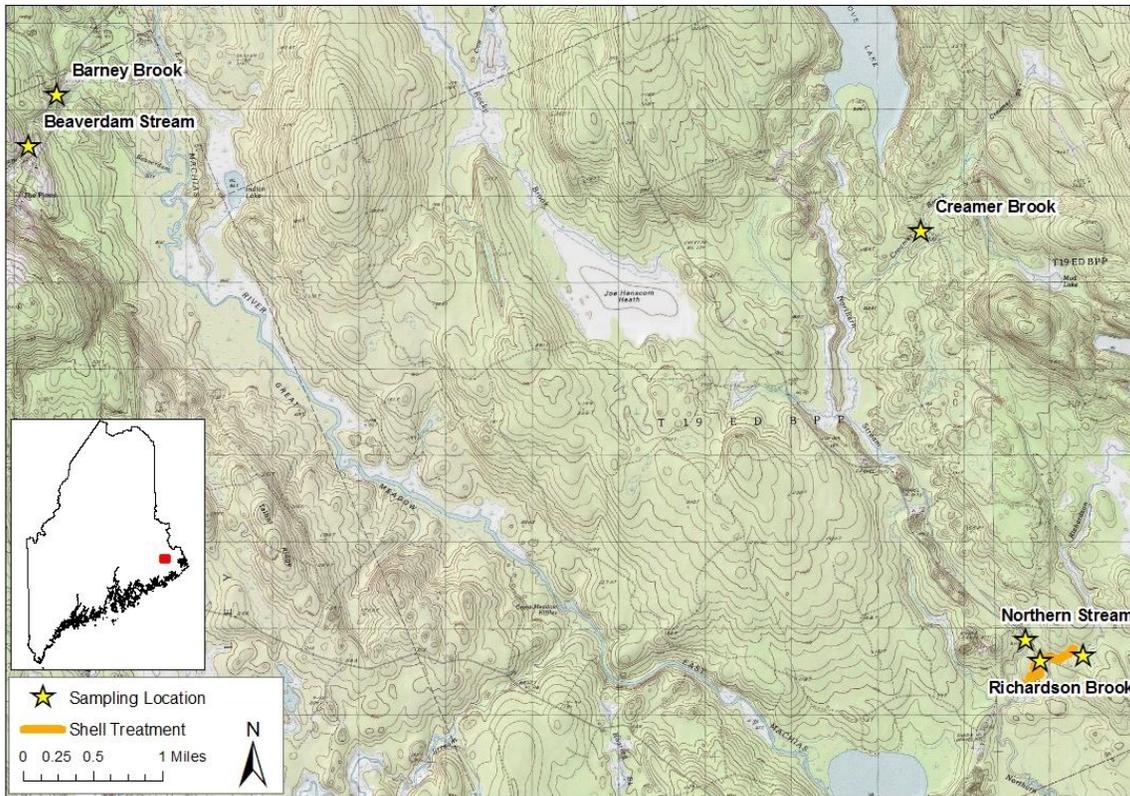
Despite restored access to historic Atlantic salmon (*Salmo salar*) habitat in eastern Maine, population sizes have remained low (USASAC 2020). Most Downeast rivers and streams have been identified as acidic (pH <6.5), with headwaters chronically acidic and main stems episodically acidic (Haines et al. 1990; Whiting and Otto 2008). Loss of fish populations due to acidification of surface waters has been well documented in the North Atlantic region (as reviewed by Clair and Hindar 2005; Dennis and Clair 2012). In addition, numerous studies have demonstrated that episodic exposure to low pH can have detrimental, sub-lethal impacts especially when coinciding with key salmon life stages during snow melt and spring runoff (e.g., Kroglund et al. 2008; Lacroix and Knox 2005; as reviewed by McCormick et al. 1998). Adding lime to acidic waters, through application of agricultural lime or lime slurry, has increased salmon populations in Scandinavia and Nova Scotia (as reviewed by Clair and Hindar 2005; Halfyard 2007; Hesthagen et al. 2011), and has been a recommended restoration action for Maine's acidic rivers and streams (NRC 2004). A 2009 Project SHARE pilot study investigating the efficacy of using clam shells to lime small streams suggested a trend towards improved habitat quality (Whiting 2014). For a more detailed project background, see Zimmermann (2018).

To further investigate this mitigation method, the Downeast Salmon Federation (DSF) started a multi-year liming project in the East Machias River watershed in 2019. Clam shells are being spread along the stream bottom, as well as along the banks to capture high flow events (i.e., rainfall and snowmelt, when episodic acidity is expected). The project goal is to increase juvenile salmon abundance by application of clam shells to achieve a target pH, and to evaluate changes in the macroinvertebrate community, which provides a food source for salmon. From 2017 through summer 2019, baseline data were collected (see Zimmermann 2019). Each summer starting in 2019, shells were spread along a treatment reach in Richardson Brook over multiple days, with shell additions occurring Sept. 7-14, 2021. Half of the shells spread in 2021 were quahog (*Mercenaria mercenaria*), in contrast with prior years when all were soft shell clams (*Mya arenaria*). This report investigates any impacts to water quality from the addition of shells.

## Methods

### Study Location

Four tributary streams to the East Machias River were monitored (Fig. 1; for physical characteristics see Appendix I Table 1 in Zimmermann 2020). These are within the homeland of the Passamaquoddy Tribe of Abenakis. The East Machias River watershed is typical of coastal eastern Maine, with extensive wetlands resulting in colored waters high in organic materials and low in pH, and with high summer temperatures (Project SHARE-USFWS 2009; Zimmermann 2020). The existing salmon population in the East Machias River system is low (median large parr density 13.1 per habitat unit, 100m<sup>2</sup> in 2019), with an estimated 1289 ± 233 smolts exiting the system in 2019 (Maine Department of Marine Resources, MDMR; DSF; USASAC 2020). In 2021, 12 redds were observed in the watershed (MDMR). Richardson Brook and Creamer Brook are stocked by DSF and MDMR, and the average large parr density observed during fall electrofishing is 11 parr/100m<sup>2</sup> and 16 parr/100m<sup>2</sup> respectively (Fig. 2, MDMR data). Five species of fish were present in Richardson Brook in 2021, matching the average of prior years (MDMR data; Appendix I Table 1 in Zimmermann 2020). The bedrock geology in the study area is predominantly marine sandstone and slate with some volcanic rocks, especially around

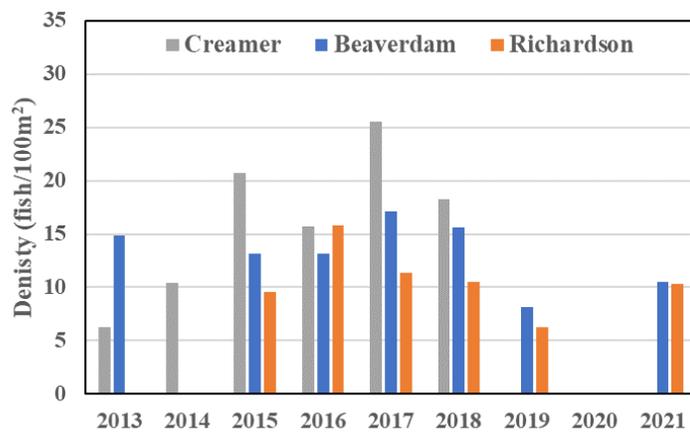


**Figure 1.** Map of the study sites on tributaries to the East Machias River. On Richardson Brook, samples were collected below (Rich-B) and above the shell treatment reach (Rich09). Northern Stream was only sampled for macroinvertebrates.

Creamer Brook (see Appendix I Table 2 in Zimmermann 2020; Maine Geological Survey – MGS 1985). Beaverdam Stream is stocked by DSF, has an average of 13 parr/100m<sup>2</sup>, and had 3 species present in 2021, lower than the average of the prior years (Fig. 2; MDMR data; Appendix I Table 1 in Zimmermann 2020).

Water Quality

All water quality monitoring activities followed the EPA-approved Salmon Habitat Monitoring Program Quality Assurance Project Plan (MDEP 2021). Continuous monitoring devices provided water quality data every half hour from April – November 2020 (see Zimmermann 2018 for detailed methods). Grab samples for acid neutralization capacity (ANC), calcium, aluminum species, dissolved organic carbon (DOC), closed-cell pH, and base cations were collected in April, August, September, and November (Appendix I Table 1; see Zimmermann 2018 for detailed methods). DSF staff collected macroinvertebrate data in October



**Figure 2.** Salmon density in three of the study streams from 2013-2021. High flows prevented data collection in Creamer Brook in 2019 and 2021. Extremely low flows prevented data collection in 2020. Data from MDMR electrofishing surveys.

at three locations using rock bags, following the Izaak Walton League of America's stream-side identification methods (IWLA 2021).

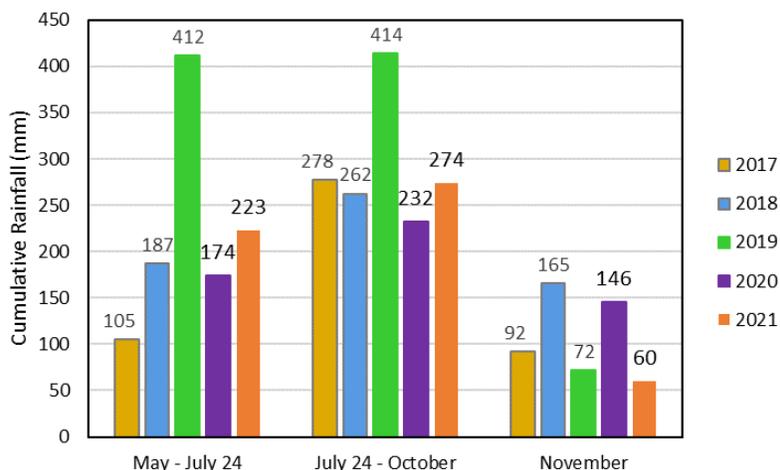
### Statistical Analysis

Water quality data were analyzed using the Water Resources Database 6.1.0.101 (Wilson Engineering 2021) and R 4.1.2 (R Core Team 2021). Plots were created using *ggplot2* (Wickham 2009). All data are presented as mean  $\pm$  standard deviation (SD), unless otherwise stated. Due to the small sample sizes, non-parametric Kruskal-Wallis tests were used to compare water grab sample results between sites, seasons, and years, with pairwise Wilcoxon rank sum post-hoc tests with Holm adjustment. Continuous data across all sites had 4.8% of pH data and 1.9% of specific conductance data rejected due to quality control issues. Equipment malfunction, primarily battery failure, resulted in loss of 4.9% of continuous data for all parameters except DO, which lost 11.9% of the record. Flagged data (based on data corrections as per MDEP 2021 or best professional judgement) represented on average 11% of specific conductance, pH, and DO data.

## Results and Discussion

### Weather

In 2021, Maine experienced an unusually warm winter with low snowpack followed by a warm, dry spring (NOAA 2021; U.S. Drought Monitor 2021). The unusually hot and dry summer was interrupted by a cool, wet July (NOAA 2021). Rainfall totals were similar to 2018 and 2020 (Fig. 3; Weather Underground 2021; Zimmermann 2021), however mean stream depths were most similar to baseline years (Zimmermann 2018 and 2019). Most rain events were of small volume (<20 mm) and occurred May to October, with the largest event occurring in early July (Weather Underground 2021).



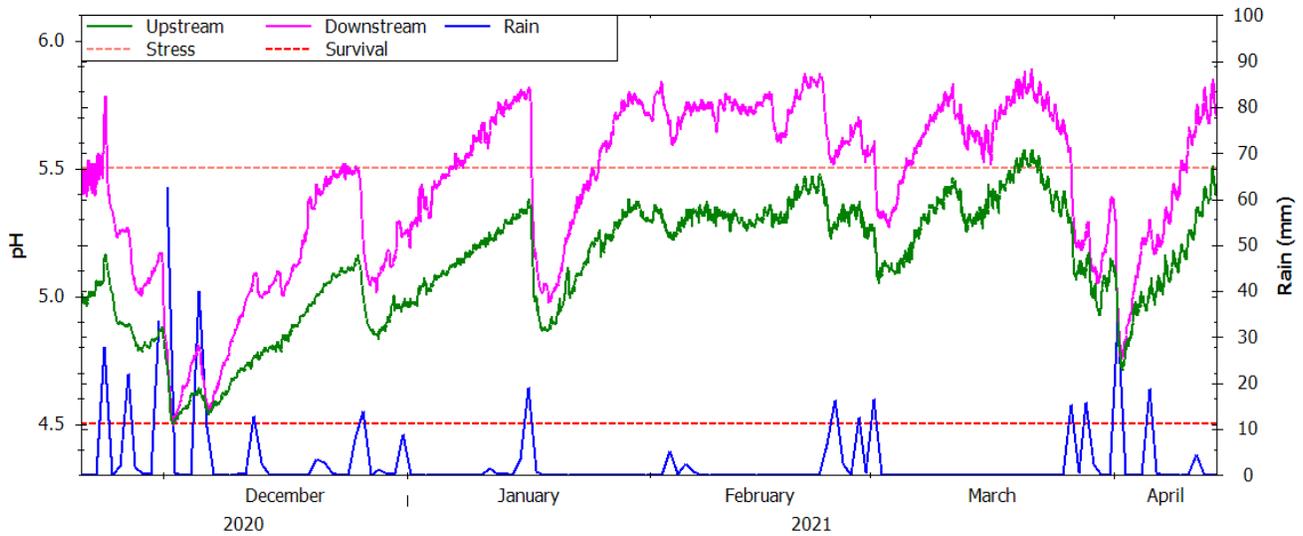
**Figure 3.** Cumulative annual rainfall. The three time periods represent spring pre-treatment, summer-fall shell treatment, and November post-treatment, based on shell applications in 2019-2021. Data from Weather Underground stations KMEALEXA2, KMEBAILE9 and KMEBARIN2.

### pH

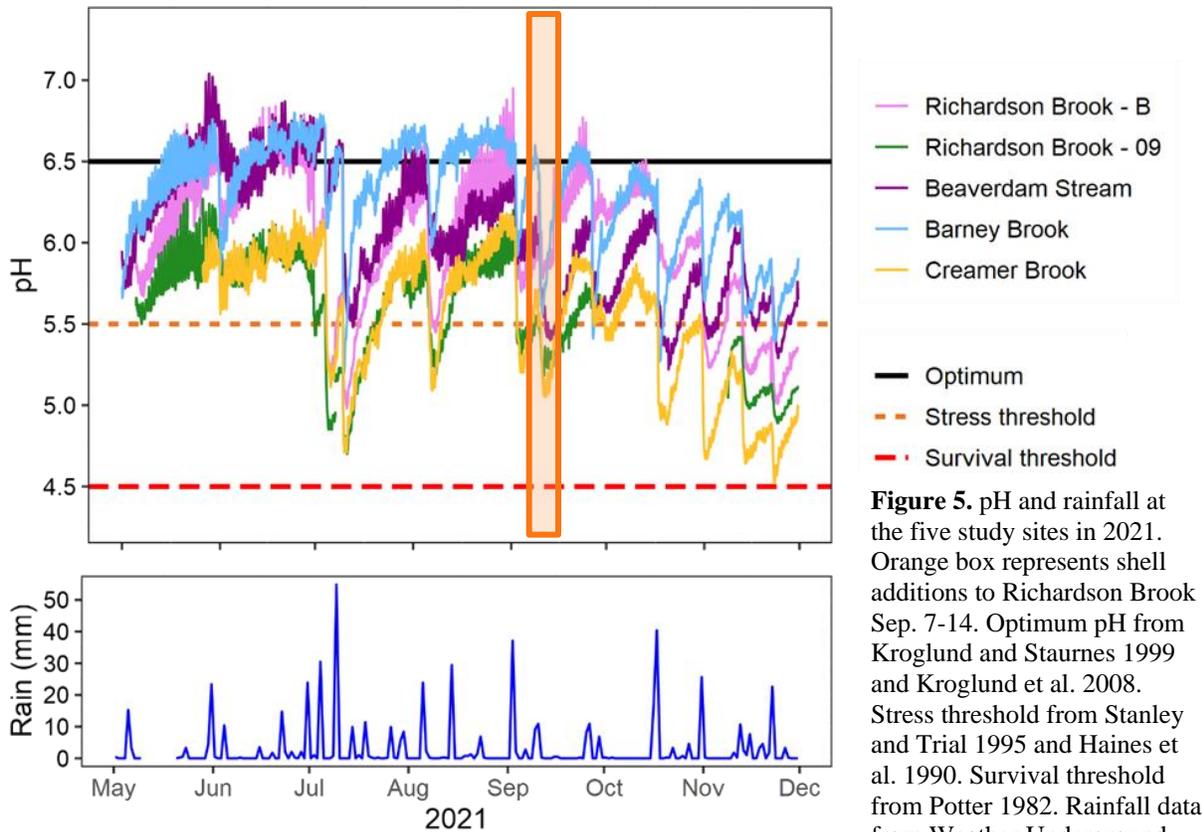
Salmon prefer pH values that are circumneutral (6.5-7.5), rather than acidic (<6.5). The impacts of acidity depend on 1.) duration, magnitude, and frequency of the episode, 2.) the ability of the fish to avoid adverse water quality conditions, 3.) the concentration of exchangeable aluminum (Al<sub>x</sub>), and 4.) the buffering capacity of the water (i.e., ANC and calcium; see Zimmermann 2018 for overview). pH thresholds used in this analysis are estimates of anticipated impacts to salmon populations and do not include a detailed analysis of the impact of other factors.

Winter pH was higher at the treated downstream Richardson Brook site in comparison with the upstream control from December 2020 to April 2021, however rainfall-driven episodic

acidity events continued to occur (Fig. 4). pH remained above the critical stress threshold of 5.5, below which adverse impacts to salmon populations are expected, for 74% of the time at the treated downstream site ( $5.4 \pm 0.3$ ) compared with 63% of the time in winter 2020, and only 3% of the time in 2021 at the upstream control ( $5.1 \pm 0.2$ ; Haines et al. 1990; Stanley and Trial 1995;



**Figure 4.** Winter 2020-2021 pH at the two Richardson Brook sites, the upstream control (Rich09) and the downstream treatment site (Rich-B). Stress threshold from Stanley and Trial 1995 and Haines et al. 1990. Survival threshold from Potter 1982. Rainfall data from Weather Underground.



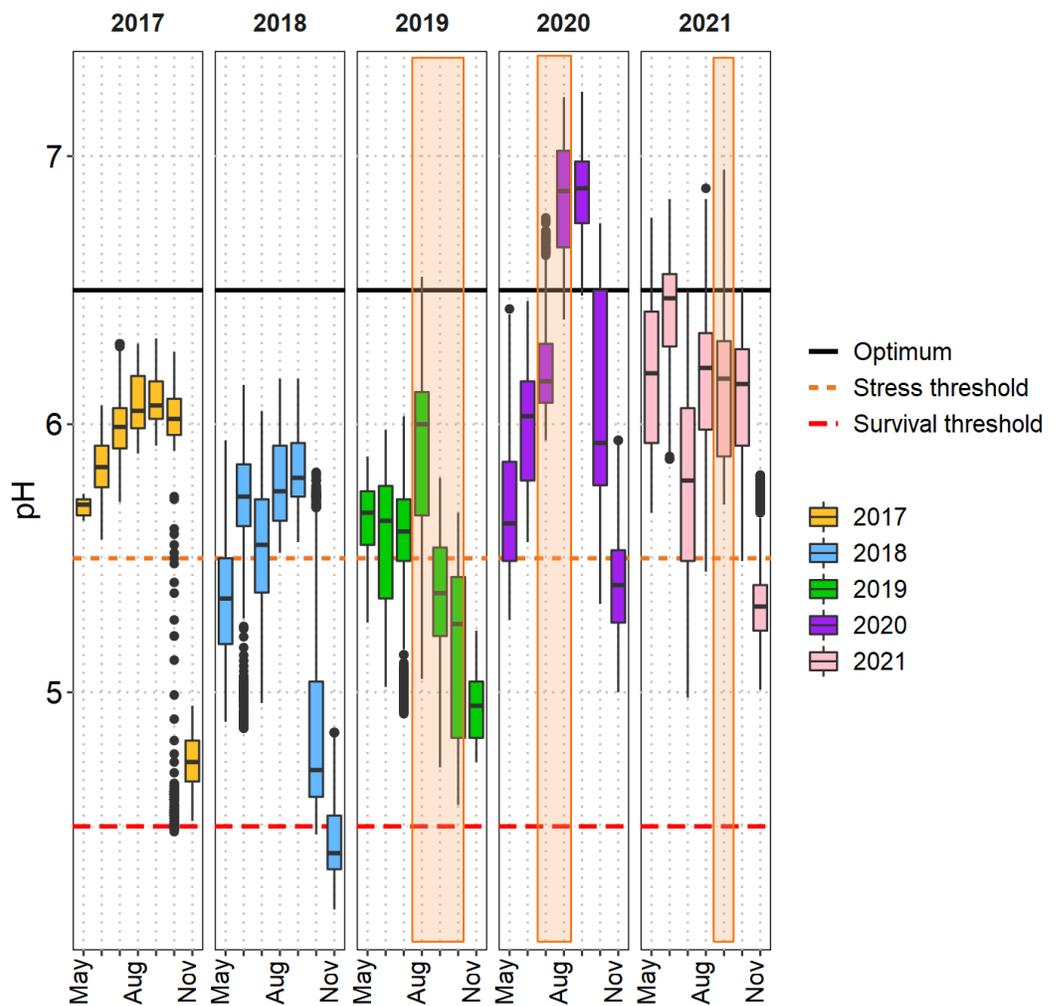
**Figure 5.** pH and rainfall at the five study sites in 2021. Orange box represents shell additions to Richardson Brook Sep. 7-14. Optimum pH from Kroglund and Staurnes 1999 and Kroglund et al. 2008. Stress threshold from Stanley and Trial 1995 and Haines et al. 1990. Survival threshold from Potter 1982. Rainfall data from Weather Underground.

Zimmermann 2021). The survival threshold of pH 4.5, lethal to all salmon life stages, was only reached once, lasting 3.5 hours at the treated site and 6.5 hours at the control site (Potter 1982). The data thus indicate that in Richardson Brook shell additions increased winter pH at the treatment site.

From spring through fall, at all sites combined, pH values remained mostly above the stress threshold of 5.5, as in prior years (Fig. 5; Appendix I Tables 2 and 6; Haines et al. 1990; Stanley and Trial 1995; Zimmermann 2021). Like prior years, the only streams with pH above the threshold of 6.5, an optimal minimum pH for the protection of the most sensitive salmon life stages (alevins and smolts), were Barney Brook (31%), Beaverdam Stream (14%), and the treated Richardson Brook site (Rich-B, 7%; Fig. 5; Appendix I Tables 2 and 5; Kroglund and Staurnes 1999; Kroglund et al. 2008). pH was slightly lower than compared to 2020 despite similar total

rainfall amounts, likely because more rain events occurred May to October 2021, with less rain in November (Fig. 3; Weather Underground 2021). As in 2020, most rain events were <20 mm, however the largest event (55 mm in July) resulted in significant pH depression at all sites (Fig. 5). Rain events were not large enough to lower pH below the survival threshold of 4.5 at any of the sites (Potter 1982).

The pH at the treated Richardson Brook site (Rich-B) was 0.4 units higher in 2021 than in baseline



**Figure 6.** Monthly pH at the downstream Richardson Brook site (Rich-B). Each box represents the interquartile range, with the horizontal line representing the median, and whiskers extending to the minimum and maximum values observed, except where values are considered statistical outliers (dots). Optimum pH from Kroglund and Staurnes 1999 and Kroglund et al. 2008. Stress threshold from Stanley and Trial 1995 and Haines et al. 1990. Survival threshold from Potter 1982. Orange boxes represent shell additions 2019-2021.

years (2017-2019; Fig. 6; Zimmermann 2020). Beaverdam Stream was the only other study site with higher minimum pH (by 0.54 units) and maximum pH (by 0.26 units) than baseline years. In 2021, there was no significant change in pH at the treatment site following the addition of shells (Fig. 6). Half the shells spread in 2021 were quahogs, which dissolve more slowly than soft shell clams, possibly reducing any immediate impacts on pH. In addition, more rain fell September to October 2021 as compared to the prior year when shells were spread during dry weather (Fig. 3). The treated site had higher autumn pH than the upstream control, as seen in 2020, and unlike baseline years when pH was similar at the two sites (Fig. 5; Appendix I Table 4; Zimmermann 2020). The data thus indicate that in Richardson Brook shell additions increased pH at the treated site to levels similar to streams with high buffering capacity (Beaverdam Stream and Barney Brook).

Although sub-lethal stress still occurred at the treated Richardson Brook site, particularly in the fall (November) when 80% of the data were below pH 5.5 in 2021 (Fig. 5; Baker et al. 1996; Henriksen et al. 1984; Lacroix and Knox 2005; Magee et al. 2003), this is an improvement over the baseline years of the study when values were <5.5 for all of November (Fig. 6; Zimmermann 2020). No significant change was observed in the duration of stressful acidic events (<5.5) in comparison with the upstream control (average duration 2 days at both sites). Both sites experienced a decrease in maximum duration compared with baseline years (Rich-B from an average maximum of 38 days during baseline to 17 days in 2021; Rich09 from an average maximum of 57 days to 22 days in 2021), however this cannot be attributed to shell additions as it occurred at both sites. Although pH at the treated site has increased following shell additions, acidic episodes are still occurring and recovery from these events has not improved.

### Stream Temperature

Salmon prefer cold waters. Stream temperatures in 2021 were similar to the prior years of the study in all study streams, remaining below the threshold for optimal growth of 20°C for most of the sampling period (89% at all streams combined; Appendix I Tables 2 and 6; Jonsson et al. 2001; USEPA 1986; Zimmermann 2021). At all streams combined, the stress threshold of 22°C was exceeded 4.1% of the time (Cunjak et al. 2005; Elliott and Elliott 2010; Lund et al. 2002), USEPA's short-term maximum for survival of 23°C was exceeded 2.3% of the time (USEPA 1986), the lethal temperature for adult salmon survival (26-27°C) was exceeded only 0.06% of the time (Shepard 1995 as cited in Frechette et al. 2018), and the lethal temperature for parr (28-29°C) was never exceeded (Elliott 1991 as cited in Stanley and Trial 1995; Garside 1973 as cited in Lund et al. 2002; Grande and Andersen 1991 as cited in Elliott and Elliott 2010). During the hot, dry summer, Barney and Creamer Brooks remained the coldest, possibly due to the relative influence of groundwater during low flows (Appendix I Table 2). Barney Brook never exceeded the stress threshold of 22°C, but other sites experienced stressful temperatures lasting on average 8 hours at a time. The longest period above 22°C occurred at the upstream Richardson Brook site at the end of June, lasting 3.5 days. As in prior years, sub-lethal stress may be occurring during the hottest parts of the summer.

### Specific Conductance

Specific conductance is a measure of the concentration of ions in the water, or the ability of water to conduct electricity. The streams in the study area have very low specific conductance ( $26 \pm 8 \mu\text{S}/\text{cm}$  at all sites combined), which can increase the difficulty of accurate pH

measurements and electrofishing (Hovind 2010 as cited in Garmo et al. 2014; Zimmermann 2018). Lab-analyzed closed-cell pH grab samples closely matched sonde pH ( $0.10 \pm 0.22$ ), with the closest match at the treatment site at Richardson Brook ( $-0.04 \pm 0.11$ ), providing confidence in the pH data discussed above. In 2021, specific conductance was similar to baseline years, with rain events causing periodic declines (Appendix I Table 2; Zimmermann 2020). The summer spike in specific conductance observed at the treated Richardson Brook site in 2020 was not observed in 2021 (Zimmermann 2021).

### Dissolved Oxygen (DO)

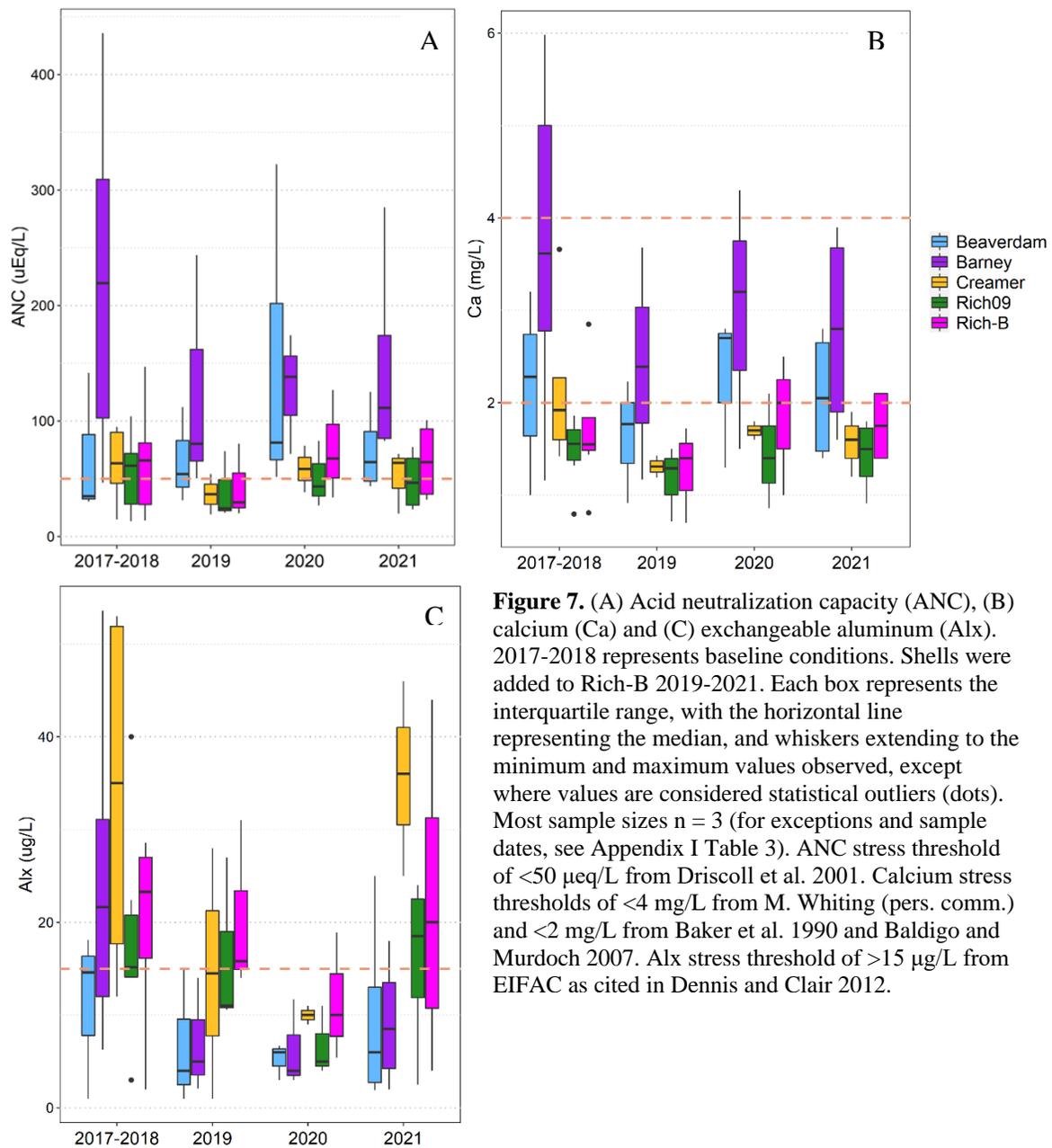
Salmon prefer well oxygenated waters. As in prior years, DO levels were within a healthy range for fish and aquatic life in addition to the preferred range for salmon of  $>6-7$  mg/L for most of the study period (98%; Appendix I Tables 2 and 6; Stanley and Trial 1995; USEPA 1986). DO concentrations fell below the Maine Water Quality Standard of 7 mg/L at all sites except Creamer Brook during the hot dry summer of 2021, lasting on average 4 hours, with a maximum duration of 18.5 hours at the upstream Richardson Brook site (38 MRS Section 465.2.B). DO never dropped below USEPA's threshold for acute impairment of 5 mg/L (USEPA 1986). The hot dry summer resulted in DO minima that coincided with the warmest temperatures, increasing stress, as well as coinciding with the lowest flows, possibly preventing movement of salmon to oxygen and temperature refugia, if any existed nearby.

### Acid Neutralization Capacity (ANC)

Streams with higher ANC have a higher capacity to buffer against changes in acidity. As in prior years, summer baseflow stayed consistently above the threshold of acid sensitivity for the protection of the most sensitive aquatic species and life stages of  $50 \mu\text{eq/L}$  (Driscoll et al. 2001; Zimmermann 2021). ANC minima were below the Norwegian  $20-30 \mu\text{eq/L}$  critical limit for salmon at two sites, the upstream control at Richardson Brook in spring and fall, and Creamer Brook in fall (Fig. 7 A; Appendix I Table 3; Baker et al. 1990; Lien et al. 1996; Kroglund et al. 2002; Zimmermann 2020). ANC is likely only high enough ( $>100 \mu\text{eq/L}$ ) for maintenance of the necessary calcium concentration (2 mg/L) during summer baseflows, and primarily only at Barney Brook and Beaverdam Stream (Fig. 7 A; Brocksen et al. 1992). Minimum ANC was higher in 2020 and 2021 compared with baseline years, however this trend was observed at all sites and cannot be attributed to shell additions (Zimmermann 2018). There were no statistically significant differences at the treated Richardson Brook site between years, compared to the other sites, or pre- and post- shell additions (Appendix I Table 4). In low DOC waters, ANC is an approximate surrogate for alkalinity (Garmo et al. 2014). As in prior years, no samples were above USEPA's recommended AWQC of 20 mg/L alkalinity (calculated from ANC), however this threshold does not apply where values are naturally lower (USEPA 1986). Relatively low ANC values indicate a deficit of buffering materials in the watershed due to thin soils (Potter 1982), allowing volatile swings in pH after rain inputs and increasing the potential for salmon mortality (Fig. 5; MacAvoy and Bulger 1995).

### Calcium

Higher calcium values enable faster growth and higher survival in fish. As in the prior years of the study, calcium was below the survival threshold of 2 mg/L at all sites for most (63%) of the sampling events (Fig. 7 B; Appendix I Tables 3 and 6; Baker et al. 1990; Baldigo and Murdoch 2007; Zimmermann 2021). No samples were above the suggested threshold of 4 mg/L to prevent



deformities and other stress (Marcus et al. 1986, as cited in Brocksen et al. 1992). The addition of clam shells to Richardson Brook has had no statistically significant impact on calcium (Appendix I Table 4). As in prior years, calcium minima coincided with low pH, high aluminum, and low ANC, however some buffering of Alx is expected to occur during summer baseflow.

### Aluminum

No significant changes in aluminum were observed between 2021 and prior years (Zimmermann 2021). Average total aluminum per stream ranged from 100 to 378  $\mu\text{g/L}$ , well below the Maine AWQC maximum of 750  $\mu\text{g/L}$  (based on a pH of 6.5-9 and dissolved organic carbon (DOC)  $<5 \text{ mg/L}$ , which are significantly different from values observed in the study streams; Appendix I Tables 2, 3 and 5; MDEP CMR Chapter 584). USEPA's site-specific maximum criterion (CMC; ranging from 5-1,400  $\mu\text{g/L}$  depending on DOC, total hardness, and

pH at each sample site; USEPA 2018) was exceeded in 58% of samples, similar to baseline years (Zimmermann 2020). As in prior years, organic aluminum was the dominant species.

Exchangeable aluminum (Alx) can cause respiratory distress when it binds to the gills of fish. Alx values were similar to baseline years, representing  $9.0 \pm 6.0\%$  of aluminum species (Zimmermann 2020). Approximately half of the 2021 samples exceeded the threshold for the protection of aquatic life of  $15 \mu\text{g/L}$  (Fig. 7 C; Appendix I Tables 5 and 6; Howells et al. 1990 as cited in Dennis and Clair 2012; Kroglund and Staurnes 1999; McCormick et al. 2009). There were no statistically significant differences at the treated Richardson Brook site between years, compared to the other sites, or pre- and post- shell additions (Appendix I Table 4). As in prior years, sub-lethal stress due to toxic Alx may decrease smolt tolerance to saltwater when they migrate out of their freshwater habitat (Kroglund and Staurnes 1999; McCormick et al. 2009; Monette et al. 2008; Staurnes et al. 1995).

### Dissolved Organic Carbon (DOC)

DOC can help buffer against the toxic impacts of Alx by binding aluminum into inert organic complexes (Baldigo and Murdoch 2007; Kroglund et al. 2008; Tipping et al. 1991). Downeast streams, including those studied here, are naturally highly colored, with relatively high organic content (and therefore high DOC) due to wetlands and coniferous forests (Haines et al. 1990). In 2021, DOC at all sites combined ranged from 7.2 to 27 mg/L, with an average of  $15.7 \pm 6.0 \text{ mg/L}$ , significantly higher than in 2019 or 2020 ( $\chi^2$  (4,  $n = 73$ ) = 17.24,  $p < .01$ ; Appendix I Table 3; Zimmermann 2021). DOC was higher during the ‘post’ shell addition time period (>July 26, 2020) at all sites compared to ‘pre’ shell additions, with a statistically significant increase at the two Richardson Brook sites (Rich-B  $\chi^2$  (1,  $n = 17$ ) = 3.95,  $p < .05$ ; Rich09  $\chi^2$  (1,  $n = 16$ ) = 4.50,  $p < .05$ ) and Beaverdam Stream ( $\chi^2$  (1,  $n = 13$ ) = 6.22,  $p < .05$ ). Based on the high DOC values, it is expected that some buffering of Alx is occurring in the study streams despite low pH values, however the increase cannot be attributed to the addition of shells.

### Base Cation Surplus

Base cation surplus (BCS) reduces the influence of natural acidity from DOC, to help distinguish the impacts of natural acidity versus anthropogenic acidification (Lawrence et al. 2007; Baldigo et al. 2009). BCS is the difference between the sum of cations (calcium, potassium, magnesium, and sodium) and anions (chloride, nitrate, sulfate, and strong organic anions as defined as  $0.071 \cdot \text{DOC} - 2.1$ ; Lawrence et al. 2007). The threshold for aluminum mobilization occurs at a BCS around 0, regardless of DOC values. In 2021, BCS ranged from a minimum of -7 at Creamer Brook to 221 at Barney Brook (Appendix I Table 7). Lowest values are observed in the spring and fall, corresponding with the lowest pH values. As expected, based on calcium, ANC, and pH (Figs. 5 and 7), Beaverdam Stream and Barney Brook had the highest average BCS, and thus the highest capacity to buffer against the mobilization of toxic aluminum.

### Macroinvertebrates

Samples showed high variability between replicates at each site. A water quality index can be calculated based on occurrence of different taxa groups, with the presence of sensitive taxa increasing the index score (IWLA 2021). Ephemeroptera (mayflies), Plecoptera (stoneflies) and Trichoptera (caddisflies), collectively referred to as EPT, have low tolerance to water pollution, and are therefore indicators of good water quality. In 2021, the control site on Northern Stream (only sampled for macroinvertebrates; Fig. 1) had the highest abundance, EPT

family richness, and water quality index score. No significant differences have been observed following shell additions at Richardson Brook, however in 2020 macroinvertebrate assemblages may have been negatively impacted at all sites by extremely low flows (Appendix I Table 8).

## Conclusion

Following a third year of clam shell additions, pH in the treated section of Richardson Brook was higher than in baseline years, as well as higher than the upstream control site. Increased pH persisted throughout the winter at the treated site. Sublethal stress from warm temperatures and low dissolved oxygen may be occurring during summer baseflow conditions at all sites. Acid neutralization capacity (ANC) was slightly higher in 2020 and 2021, suggesting increased buffering, but this cannot be attributed to the addition of shells as the increase was observed at all sites. Similarly, increases in dissolved organic carbon (DOC) were not due to shell additions. No change in any of the other monitored parameters has been observed following the addition of shells. Despite the increase in pH at the treatment site, sub-lethal stress due to low pH and aluminum toxicity is likely still occurring during episodic, precipitation-driven acidity events. The low pH events often coincide with the presence of the most sensitive salmon life stages (alevins and smolts), from March through June, however the hardier life stages (parr and adults) may also be impacted during the autumn rainy season. Clam shells will be added for an additional two years, accompanied by water quality monitoring, through 2023, to determine if the increase in pH is biologically significant for salmon.

## Works Cited

- Baker, J.P., Bernard, D.P., Christensen, S.W., Sale, M.J., Freda, J., Heltcher, K., Marmorek, D., Rowe, L., Scanlone, P., Suter, G., Warren-Hicks, W., and Welbourn, P. 1990. Biological effects of changes in surface water acid-base chemistry. NAPAP Report 13. In: National Acid Precipitation Assessment Program, Acidic Deposition: State of Science and Technology. Vol. II.
- Baker, J.P., Van Sickle, J., Gagen, C.J., DeWalle, D.R., Sharpe, W.E., Carline, R.F., Baldigo, B.P., Murdoch, P.S., Bath, D.W., Kretser, W.A., Simonin, H.A., Wigington, P.J., Jr. 1996. Episodic acidification of small streams in the northeastern United States: effects on fish populations. *Ecological Applications*. 422-437.
- Baldigo, B.P., and Murdoch, P.S. 2007. Effect of stream acidification and inorganic aluminum on mortality of brook trout (*Salvelinus fontinalis*) in the Catskill Mountains, New York. *Canadian Journal of Fisheries and Aquatic Science*. 54: 603-615.
- Brocksen, R.W., Marcus, M.D., and Olem, H. 1992. Practical guide to managing acidic surface waters and their fisheries. Lewis Publishers, Inc. Chelsea, Michigan. 190 p.
- Clair, T.A., and Hindar, A. 2005. Liming for the mitigation of acid rain effects in freshwaters: a review of recent results. *Environmental Reviews*. 13: 91-128.
- Cunjak, R.A., Roussel, J.-M., Gray, M.A., Dietrich, J.P., Cartwright, D.F., Munkittrick, K.R., and Jardine, T.D. 2005. Using stable isotope analysis with telemetry or mark-recapture data to identify fish movement and foraging. *Oecologia*. 144: 1-11.
- Dennis, I.F. and Clair, T.A. 2012. The distribution of dissolved aluminum in Atlantic salmon (*Salmo salar*) rivers in Atlantic Canada and its potential effect on aquatic populations. *Canadian Journal of Fisheries and Aquatic Science*. 69: 1174-1183.
- Driscoll, C.T., Lawrence, G.B., Bulger, A.J. Butler, T.J., Cronan, C.S., Eagar, C., Lambert, K.F., Likens, G.E., Stoddard, J.L., and Weathers, K.C. 2001. Acidic deposition in the Northeastern United States: sources and inputs, ecosystem effects, and management strategies. *BioScience*. 51.3: 180-198.
- Elliott, J.M., and Elliott, J.A. 2010. Temperature requirements of Atlantic salmon *Salmo salar*, brown trout *Salmo trutta* and Arctic charr *Salvelinus alpinus*: predicting the effects of climate change. *Journal of Fish Biology*. 77: 1793-1817.
- Frechette, D.M., Dugdale, S.J., Dodson, J.J., and Bergeron, N.E. 2018. Understanding summertime thermal refuge use by adult Atlantic salmon using remote sensing, river temperature monitoring, and acoustic telemetry. *Canadian Journal of Fisheries and Aquatic Sciences*. 75: 1999-2010.

- Garmo, Ø.A., Skjelkvåle, B.L., de Wit, H.A., Colombo L., Curtis, C., Fölster, J., Hoffmann, A., Hruška, J., Høggåsen, T., Jeffries, D.S., Keller, W.B., Krám, P., Majer, V., Monteith, D.T., Paterson, A.M., Rogora, M., Rzychon, D., Steingruber, S., Stoddard, J.L., Vuorenmaa, J., and Worsztynowicz, A. 2014. Trends in surface water chemistry in acidified areas in Europe and North America from 1990 to 2008. *Water, Air, and Soil Pollution*. 225: 1880.
- Haines, T.A., Norton, S.A., Kahl, J.S., Fay, C.W., Pauwels, S.J., and Jagoe, C.H. 1990. Intensive studies of stream fish populations in Maine. EPA/600/3-90/043.
- Halfyard, E. 2007. Initial results of an Atlantic salmon river acid mitigation program. MSc Thesis, Acadia University, 164 p.
- Henriksen, A., Skogheim, O.K., and Rosseland, B.O. 1984. Episodic changes in pH and aluminum-speciation kill fish in a Norwegian salmon river. *Vatten*. 40: 255-260.
- Hesthagen, T., Larsen, B.M., and Fiske, P. 2011. Liming restores Atlantic salmon (*Salmo salar*) populations in acidified Norwegian rivers. *Canadian Journal of Fisheries and Aquatic Sciences*. 68: 224-231.
- Izaak Walton League of America (IWLA). 2021. Biological monitoring instructions for stream monitors. URL <https://www.iwla.org/water/resources-for-monitors>.
- Jonsson, B., Forseth, T., Jensen, A.J., and Næsje, T.F. 2001. Thermal performance of juvenile Atlantic Salmon, *Salmo salar*. *Functional Ecology*. 15: 701-711.
- Kroglund, F., and Staurnes, M. 1999. Water quality requirements of smolting Atlantic salmon (*Salmo salar*) in limed acid rivers. *Canadian Journal of Fisheries and Aquatic Sciences*. 56: 2078-2086.
- Kroglund, F., Wright, R.F., and Burchart, C. 2002. Acidification and Atlantic salmon: critical limits for Norwegian rivers. Norwegian Institute for Water Research, Oslo. Report nr 111.
- Kroglund, F., Rosseland, B.O., Teien, H.-C., Salbu, B., Kristensen, T., and Finstad, B. 2008. Water quality limits for Atlantic salmon (*Salmo salar*) exposed to short term reductions in pH and increased aluminum simulating episodes. *Hydrology and Earth Systems Sciences*. 12: 491-507.
- Lacroix, G.L., and Knox, D. 2005. Acidification status of rivers in several regions of Nova Scotia and potential impacts on Atlantic salmon, Canadian Technical Report of Fisheries and Aquatic Sciences, 2573.
- Lawrence, G.B., Sutherland, J.W., Boylen, C.W., Nierzwicki-Bauer, S.W., Momen, B., Baldigo, B.P., and Simonin, H.A. 2007. Acid rain effects on aluminum mobilization clarified by inclusion of strong organic acids. *Environmental Science and Technology*. 41 (1): 93-98.
- Lien, L., Raddum, G.G., Fjellheim, A., Henriksen, A. 1996. A critical limit for acid neutralizing capacity in Norwegian surface waters, based on new analyses of fish and invertebrate responses. *The Science of the Total Environment*. 177: 173-193.
- Lund, S.G., Caissie, D., Cunjak, R.A., Vijayan, M.M., and Tufts, B.L. 2002. The effects of environmental heat stress on heat-shock mRNA and protein expression in Miramichi Atlantic salmon (*Salmo salar*) parr. *Canadian Journal of Fisheries and Aquatic Sciences*. 59: 1553-1562.
- MacAvoy, S.E., and Bulger, A.J. 1995. Survival of brook trout (*Salvelinus fontinalis*) embryos and fry in streams of different acid sensitivity in Shenandoah National Park, USA. *Water, Air, and Soil Pollution*. 85: 445-450.
- Magee, J.A., Obedzinski, M., McCormick, S.D., and Kocik, J.F. 2003. Effects of episodic acidification on Atlantic salmon (*Salmo salar*) smolts. *Canadian Journal of Fisheries and Aquaculture Science*. 60: 214-221.
- Maine Department of Environmental Protection Code of Maine Rules (MDEP CMR). Chapter 584: Surface Water Quality Criteria for Toxic Pollutants.
- Maine Department of Environmental Protection (MDEP). 2021. Quality Assurance Project Plan for the Salmon Habitat Monitoring Program. Effective date 2/22/2021.
- Maine Geological Survey (MGS). 1985. Bedrock\_500K\_Units. Augusta, ME, Maine Geological Survey. URL [https://services1.arcgis.com/RbMX0mRVOFNTdLzd/arcgis/rest/services/MGS\\_Bedrock\\_500K\\_Map\\_Data/FeatureServer](https://services1.arcgis.com/RbMX0mRVOFNTdLzd/arcgis/rest/services/MGS_Bedrock_500K_Map_Data/FeatureServer). Using: ArcGIS. Version 10.3.1. Redlands, CA: Environmental Systems Research Institute, Inc., 2010. Data accessed 2/1/2021.
- Maine Revised Statutes (M.R.S.). Title 38: Waters and navigation. Chapter 3: Protection and improvement of waters. Article 4-A: Water Classification Program. Sections 464 and 465.
- McCormick, S.D., Hansen, L.P., Quinn, T.P., and Saunders, R.L. 1998. Movement, migration, and smolting of Atlantic salmon (*Salmo salar*). *Canadian Journal of Fisheries and Aquatic Science*. 55 (Suppl. 1): 77-92.
- McCormick, S.D., Keyes, A., Nislow, K.H., and Monette, M.Y. 2009. Impacts of episodic acidification on in-stream survival and physiological impairment of Atlantic salmon (*Salmo salar*) smolts. *Canadian Journal of Fisheries and Aquatic Science*. 66: 394-403.

- Monette, M.Y., Björnsson, B.T., and McCormick, S.D. 2008. Effects of short-term acid and aluminum exposure on the parr-smolt transformation in Atlantic salmon (*Salmo salar*): disruption of seawater tolerance and endocrine status. *General and Comparative Endocrinology*. 158: 122-130.
- National Oceanic and Atmospheric Administration (NOAA). 2021. Gulf of Maine region quarterly climate impacts and outlook. March, June, and September 2021. URL <https://gulfofmaine.org/public/climate-network/climate-outlook/>.
- National Research Council (NRC). 2004. Atlantic Salmon in Maine. Washington, DC: The National Academies Press. URL <https://doi.org/10.17226/10892>.
- Potter, W. 1982. The effects of air pollution and acid rain on fish, wildlife, and their habitats – rivers and streams. U.S. Fish and Wildlife Service, Biological Services Program, Eastern Energy and Land Use Team, FWS/OBS-80/40.5. 52 pp.
- Project Share and U.S. Fish and Wildlife Service (USFWS). 2009. Restoring salmonid aquatic/riparian habitat: a strategic plan for the Downeast Maine DPS rivers.
- R Core Team. 2021. R: A language and environment for statistical computing. R Foundation for Statistical Computing, Vienna, Austria. URL <http://www.R-project.org/>.
- Stanley, J.G., and Trial, J.G. 1995. Habitat suitability index models: nonmigratory freshwater life stages of Atlantic salmon. U.S. Department of the Interior. Biological Science Report 3.
- Staurnes, M. Kroglund, F., and Rosseland, B.O. 1995. Water quality requirement of Atlantic salmon (*Salmo salar*) in water undergoing acidification or liming in Norway. *Water, Air, and Soil Pollution*. 85: 347-352.
- Tipping, E., Woof, C., and Hurley, M.A. 1991. Humic substances in acid surface waters; modelling aluminum binding, contribution to ionic charge-balance, and control of pH. *Water Resources*. 25(4): 425–435.
- United States Atlantic Salmon Assessment Committee (USASAC). 2020. Annual Report, no. 32 – 2019 activities.
- United State Drought Monitor. 2021. National Drought Mitigation Center, Lincoln, NE. URL <https://droughtmonitor.unl.edu/Maps/MapArchive.aspx>
- United States Environmental Protection Agency (USEPA). 1986. Quality Criteria for Water. EPA 440/5-86-001.
- United States Environmental Protection Agency. 2018. Final Aquatic Life Ambient Water Quality Criteria for Aluminum. EPA- 822-R-18-001.
- Weather Underground. 2021. Tom’s Backyard KMEALEXA2, Alexander Elementary School KMEBAILE9 and HeatherWood Gardens KMEBARIN2. URL <https://www.wunderground.com/>.
- Whiting, M.C. 2014. Final report for Project SHARE’s Clam Shell Pilot Project. Maine Department of Environmental Protection: Bangor, Maine.
- Whiting, M.C. and Otto, W. 2008. Spatial and temporal patterns in the water chemistry of the Narraguagus River: a summary of the available data from the Maine DEP Salmon Rivers Program. Maine Department of Environmental Protection: Bangor, Maine.
- Wickham, H. 2009. ggplot2: Elegant Graphics for Data Analysis. Springer-Verlag New York.
- Wilson Engineering, LLC. 2021. Water Resources Database (WRDB). St. Louis, Missouri. URL [wrdb.com](http://wrdb.com).
- Zimmermann, E. 2018. Reducing acidification in endangered Atlantic salmon habitat: baseline data. Maine Department of Environmental Protection: Augusta, ME.
- Zimmermann, E. 2019. Reducing acidification in endangered Atlantic salmon habitat: baseline data summary. Maine Department of Environmental Protection: Augusta, ME.
- Zimmermann, E. 2020. Reducing acidification in endangered Atlantic salmon habitat: first year of clam shells. Maine Department of Environmental Protection: Augusta, ME.
- Zimmermann, E. 2021. Reducing acidification in endangered Atlantic salmon habitat: second year of clam shells. Maine Department of Environmental Protection: Augusta, ME

## Appendix I – Summary Data Tables

**Table 1.** Analytical laboratories, methods, and certification.

Analysis Lab	Analyte	Method	DEP Certified?
UMO Sawyer Water Research Lab	ANC	E600/4-87/26 5.53	No. This is a research method with no state certification, is approved by the DEP biologist, and is used for continuity of data to compare with prior liming projects.
	Aluminum (speciation)	SW6010B	No. This is a research method with no state certification, is approved by the DEP biologist, and is used for continuity of data to compare with prior liming projects.
	pH (closed-cell)	E600/4-87/26 19.0	No. This is a research method with no state certification, is approved by the DEP biologist, and is used for continuity of data to compare with prior liming projects.
Maine Environmental	Calcium and other cations	E200.7	Yes
	DOC	SM5310B	Yes
	Anions (chloride and sulfate)	E300.0	Yes
Eastern Analytic, Inc.	Nitrate	E300.0 and E353.2	Yes

**Table 2.** Continuous Data Summary. Summary statistics (mean, standard deviation (SD), minimum and maximum) of measurements from Manta+ 20 and YSI 600 XLM sondes, May to Nov. 2021 (n ~ 10,000), and Onset Hobo U26 dissolved oxygen loggers, June to Nov. 2020 (n ~ 9,000). Dissolved oxygen data for Barney Brook are discrete measurements from a Eureka Manta2 Sub2 sonde (n = 12).

Stream Name	pH				Temperature (°C)				Specific Conductance (µS/cm)				Dissolved Oxygen (mg/L)			
	Mean	SD	Min	Max	Mean	SD	Min	Max	Mean	SD	Min	Max	Mean	SD	Min	Max
Barney Brook	6.27	0.32	5.19	6.8	12.28	4.96	0.24	21.43	33	7	15	54	10.88	1.96	9.15	15.23
Beaverdam Stream	6.05	0.38	5.22	7.04	14.95	6.00	-0.05	28.3	32	8	16	68	9.20	1.16	6.92	13.11
Creamer Brook	5.52	0.41	4.52	6.20	13.37	5.43	0.08	23.11	26	3	20	34	9.53	1.11	7.38	13.70
Richardson Brook – 09 (upstream control)	5.55	0.35	4.70	6.27	14.74	6.20	0.11	27.73	20	2	16	26	8.89	1.52	5.86	13.31
Richardson Brook – B (treatment)	5.97	0.44	4.75	6.95	14.44	6.01	0.01	26.37	19	4	12	29	9.72	1.79	6.56	15.44

**Table 3.** Discrete Data Summary. Summary statistics (mean, SD, minimum and maximum) from grab samples collected in 2017 (June 20, Aug. 1, and Oct. 11), 2018 (April 18, July 23, and Nov. 5), 2019 (April 1, July 31, and Nov. 19), 2020 (April 28, July 22, and Nov. 23), and 2021 (April 14, Aug. 17, Sept. 29, and Nov. 30). n = 16\*.

Stream Name	Calcium (mg/L)				Dissolved Organic Carbon (mg/L)				ANC (µeq/L)				pH (closed-cell)			
	Mean	SD	Min	Max	Mean	SD	Min	Max	Mean	SD	Min	Max	Mean	SD	Min	Max
Barney Brook	3.10	1.44	1.16	5.98	11.3	5.9	3.4	27	157.4	110.8	46.6	435.9	6.42	0.37	5.82	6.96
Beaverdam Stream	2.12	0.73	0.92	3.20	12.4	6.0	6.1	26	92.8	77.3	31.4	322.3	6.11	0.46	5.28	6.88
Creamer Brook	1.80	0.66	1.19	3.66	12.1	4.2	7.6	20	54.6	27.5	14.8	94.9	5.79	0.41	4.96	6.26
Richardson Brook - 09+ (upstream control)	1.40	0.41	0.72	2.10	12.6	4.7	5.6	21	54.1	28.1	13.3	104.0	5.66	0.45	4.80	6.25
Richardson Brook – B (treatment)	1.65	0.58	0.70	2.85	12.4	4.3	6.8	20	65.2	38.0	13.9	147.0	5.95	0.49	4.94	6.76

\* Creamer Brook was not sampled in April in 2018, 2019, 2020, or 2021 (n = 12). Beaverdam Stream was not sampled in 2017 (n = 13).

+ Rich09 includes samples collected from Rich-A (a site 360m downstream) in 2017, 2018, and April 2019.

**Table 4.** Treatment Summary. Mean values ( $\pm$  SD) before any change in pH was observed at the treated Richardson Brook site ('pre': May 30, 2017 – July 26, 2020) and after pH increased ('post': July 26, 2020 – Nov. 30, 2021).

Stream Name	pH		Calcium (mg/L)		Exchangeable Aluminum (µg/L)		Acid Neutralization Capacity (µEq/L)	
	Pre <i>n</i> ~ 30,000	Post <i>n</i> ~ 3,000	Pre <i>n</i> = 11*	Post <i>n</i> = 5*	Pre <i>n</i> = 11*	Post <i>n</i> = 5*	Pre <i>n</i> = 11*	Post <i>n</i> = 5*
Barney Brook	6.3 $\pm$ 0.3	6.4 $\pm$ 0.2	3.2 $\pm$ 1.6	2.9 $\pm$ 1.0	16.5 $\pm$ 15.6	8.2 $\pm$ 6.6	177 $\pm$ 128	146 $\pm$ 82
Beaverdam Stream	6.1 $\pm$ 0.5	6.5 $\pm$ 0.4	1.9 $\pm$ 0.8	2.2 $\pm$ 0.7	8.9 $\pm$ 6.9	9.0 $\pm$ 9.4	97 $\pm$ 100	76 $\pm$ 32
Creamer Brook	5.7 $\pm$ 0.4	5.7 $\pm$ 0.4	1.9 $\pm$ 0.8	1.6 $\pm$ 0.3	29.5 $\pm$ 18.0	29.5 $\pm$ 15.0	58 $\pm$ 30	48 $\pm$ 24
Richardson Brook – 09+ (upstream control)	5.6 $\pm$ 0.4	5.4 $\pm$ 0.5	1.3 $\pm$ 0.4	1.6 $\pm$ 0.5	15.8 $\pm$ 10.6	13.5 $\pm$ 9.9	51 $\pm$ 31	47 $\pm$ 23
Richardson Brook – B (treatment)	5.5 $\pm$ 0.5	6.1 $\pm$ 0.8	1.5 $\pm$ 0.6	1.9 $\pm$ 0.5	18.6 $\pm$ 9.4	19.6 $\pm$ 16.0	62 $\pm$ 45	66 $\pm$ 31

\* Creamer Brook was not sampled in April 2018-2021 (n = 8 pre, 4 post). Beaverdam Stream was not sampled in 2017 (n = 8 pre).

+ Rich09 includes samples collected from Rich-A (a site 360m downstream) in 2017, 2018, and April 2019 (pre).

**Table 5.** Aluminum Species Data Summary. Summary statistics (mean, SD, minimum and maximum) from grab samples collected in 2017 (June 20, Aug. 1, and Oct. 11), 2018 (April 18, July 23, and Nov. 5), 2019 (April 1, July 31, and Nov. 19), 2020 (April 28, July 22, and Nov. 23), and 2021 (April 14, Aug. 17, Sept. 29, and Nov. 30). n = 17\*.

Stream Name	Total Aluminum (µg/L)				Dissolved Aluminum (µg/L)				Exchangeable Aluminum (µg/L)			
	Mean	SD	Min	Max	Mean	SD	Min	Max	Mean	SD	Min	Max
Barney Brook	179	90	40	423	154	78	32	377	14	14	2	54
Beaverdam Stream	140	54	54	241	119	50	32	219	9	8	<1	25
Creamer Brook	237	93	94	424	216	89	92	399	30	16	9	53
Richardson Brook – 09 <sup>+</sup> (upstream control)	187	69	101	340	169	60	75	279	15	10	3	40
Richardson Brook – B (treatment)	180	60	88	293	166	57	79	278	19	11	2	44

\* Creamer Brook was not sampled in April 2018-2021 (n = 12). Beaverdam Stream was not sampled in 2017 (n = 13).

+ Rich09 includes samples collected from Rich-A (a site 360m downstream) in 2017, 2018, and April 2019.

**Table 6.** Exceedance Summary. Percentage of data observations that exceeded stress threshold values for sonde data (pH, temperature and DO) April-Nov. 2021. Grab sample data (calcium and exchangeable aluminum) combine all five years of the study 2017-2021.

Stream Name	Continuous Data					Grab Sample Data		
	pH (n ~ 10,000)		Temperature (n ~ 10,000)	Dissolved Oxygen (n ~ 9,000) <sup>^</sup>		Calcium (n = 16)*		Exchangeable Aluminum (n = 16)*
<i>Thresholds</i>	<5.5	<6.5	>20.0 °C	<5 mg/L	<7 mg/L	<2.0 mg/L	<4.0 mg/L	>15 µg/L
Barney Brook	2.0	67.8	2.1	0	0	29.4	76.5	25.0
Beaverdam Stream <sup>a</sup>	7.8	84.9	22.4	0	0.1	38.5	100	23.1
Creamer Brook	40.6	100	8.8	0	0	83.3	100	66.7
Richardson Brook – 09 <sup>+</sup> (upstream control)	40.5	100	19.9	0	0.9	93.8	100	56.3
Richardson Brook – B (treatment)	18.5	89.9	17.4	0	6.4	68.8	100	37.5

<sup>^</sup> DO data for Barney Brook are discrete measurements from a Eureka Manta2 Sub2 sonde (n = 12).

\* No grab samples were collected at Creamer Brook in April in 2018-2021 (n = 12)

<sup>a</sup> No grab samples were collected at Beaverdam Stream in 2017 (n = 12).

+ Rich09 includes samples collected from Rich-A (a site 360m downstream) in 2017, 2018, and April 2019.

**Table 7.** Base Cation Surplus (BCS). Summary statistics (mean and SD) from grab samples collected in 2019 (July 31 and Nov. 19) and 2020 (April 28, July 22 and Nov. 23), and 2021 (April 14, Aug. 17, Sept. 29, and Nov. 30). Cations include calcium, potassium, magnesium, and sodium. Anions include chloride, nitrate, sulfate, and strong organic anions (0.071\*DOC-2.1, from Lawrence et al. 2007). Data converted from mg/L. n = 9\*.

Stream Name	Cations (µEq/L)		Anions (µEq/L)		BCS (µEq/L)			
	Mean	SD	Mean	SD	Mean	SD	Min	Max
Barney Brook	309.3	92.1	163.1	46.5	146.2	83.3	57.8	301.3
Beaverdam Stream	297.2	75.7	238.7	58.9	58.5	34.5	26.2	131.8
Creamer Brook	218.7	26.9	199.4	38.3	19.3	31.1	-14.4	78.4
Richardson Brook – 09 (upstream control)	193.3	38.6	164.8	37.0	28.6	30.9	-5.6	85.7
Richardson Brook – B (treatment)	212.8	39.9	166.3	38.4	46.4	29.0	11.5	108.3

\* Creamer Brook was not sampled in April 2019-2021 (n = 7).

**Table 8.** Macroinvertebrate Summary. Mean values ( $\pm$  SD) before any change in pH was observed at the treated Richardson Brook site ('pre': 2017 – 2019, n =7 at Beaverdam Stream, n = 6 at Northern Stream, and n = 9 at Richardson Brook) and after pH increased ('post': 2020 – 2021, n = 6). Macroinvertebrates were identified to the family in the field by DSF. Water quality index values are based on the occurrence of different taxa groups (IWLA 2021).

Stream Name	Mean Abundance		Mean Family Richness		EPT Family Richness		Water Quality Index	
	Pre	Post	Pre	Post	Pre	Post	Pre	Post
Beaverdam Stream	69 $\pm$ 35	35 $\pm$ 17	11 $\pm$ 5	11 $\pm$ 2	9 $\pm$ 3	9 $\pm$ 2	13 $\pm$ 3	12 $\pm$ 1
Northern Stream	115 $\pm$ 25	55 $\pm$ 32	11 $\pm$ 5	14 $\pm$ 4	11 $\pm$ 1	10 $\pm$ 2	17 $\pm$ 3	15 $\pm$ 3
Richardson Brook (treatment)	62 $\pm$ 24	44 $\pm$ 18	10 $\pm$ 4	12 $\pm$ 2	9 $\pm$ 2	10 $\pm$ 2	14 $\pm$ 2	13 $\pm$ 3