Proceedings of The National Conference
On Undergraduate Research (NCUR) 2017
University of Memphis, TN
Memphis, Tennessee
April 6-8, 2017

# Effects of Atmospheric Acid Reduction on Water Chemistry of Three Virginia Trout Streams

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## Abstract

Acid deposition from fossil fuel combustion has adversely affected streams in North America for more than four decades by emitting SOx and NOx gases into the atmosphere. Free radical reactions occur to form sulfuric and nitric acid. That is released into the streams. The pH and ANC (acid neutralizing capacity) decrease, which leads to fauna and flora mortality. The U. S. Congress passed the Clean Air Act to reduce power plant emissions in 1990. SOx and NOx gases have decreased by 82 % and 76%, respectively between 1996 and 2015. We have intensively collected water chemistry data on three acid sensitive streams in the George Washington National Forest: Little Stony Creek, Mill Creek, and Mountain Run monthly since 1987. The purpose of this project is to ascertain if the reductions in acid deposition are being realized with a positive response within the streams. Little Stony, Mill Creek, and Mountain Run have shown a 25.6, 25.6, and 42.8% reduction in sulfate concentration, respectively. Resulting in an ANC increase of 19% (8.1 µeq/L to 13.6 µeq/L), 9.0% (–11.1µeq/L to –8.5 µeq/L), and 24.8% (–27.2 µeq/L to –20.4 µeq/L) for Little Stony, Mill Creek, and Mountain Run, respectively. All three streams show a positive response to the reductions in atmospheric acid but have not yet returned to pre-industrial age values.

**Key Words: Atmospheric Acid, Water Chemistry, Limestone Treatment**

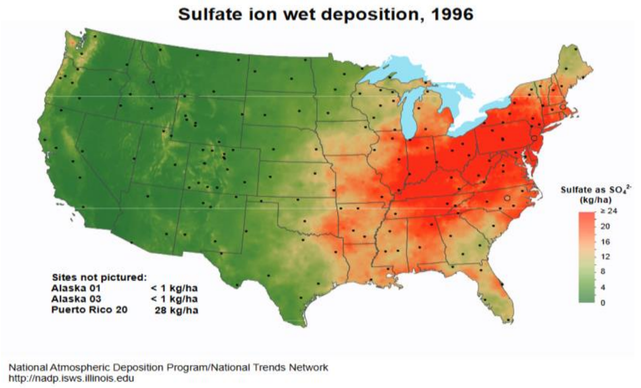
## 1. Introduction

Rainfall is naturally acidic (pH~5.6) due to dissolution of carbon dioxide in the atmosphere to form carbonic acid ($H_2CO_3$). Sulfur dioxide ($SO_2$) and nitrogen oxides ($NO_x$) from burning fossil fuels create sulfuric and nitric acids that lower rainfall to near pH 4.5. [Equations (1-5)]

$$SO_2\,(g) + OH^-\,(g) \rightarrow \bullet HSO_3\,(aq) \tag{1}$$
$$\bullet HSO_3\,(aq) + O_2\,(g) \rightarrow SO_3\,(g) + \bullet OOH \quad (g) \tag{2}$$
$$SO_3\,(g) + H_2O\,(l) \rightarrow H_2SO_4\,(aq) \tag{3}$$
$$NO\,(g) + O_3\,(g) \rightarrow NO_2\,(g) + O_2\,(g) \tag{4}$$
$$NO_2\,(g) + \bullet OH\,(g) \rightarrow HNO_3\,(aq) \tag{5}$$

The deposition of human origin acid compounds is called anthropogenic atmospheric acid deposition or simply “Acid Rain”. In the 1980s some streams and lakes experienced fauna and biota depressions. In response to this, the United States Congress founded the National Acid Precipitation Assessment Program (NAPAP) to study the effects of acid rain on natural and manmade resources in the United States, which revealed that headwater streams within the

Appalachian Mountains that originate in carbonate poor watersheds were adversely affected.[1] In the 1900s the extent of acid deposition was demonstrated by the amount of sulfate ion deposition depicted in Figure 1.


Sulfate ion wet deposition, 1996

National Atmospheric Deposition Program/National Trends Network
http://nadp.isws.illinois.edu

Figure 1. Isopleth map from National Acid Deposition Program (NADP) showing sulfate ion wet deposition (kg/ha) in 1996.

The effect of the acid deposition on streams was not only dependent on the amount of acid entering a watershed from rainfall but also on the type of geology. The predominate buffer in surface waters is bicarbonate ($HCO_3^-$) ion from the dissolution of minerals containing carbonate salts, $Ca^{2+}$ and $Mg^{2+}$ carbonate [Equation (6-7)]. Waters located within a region of low carbonate geology are prone to acidification when rainfall acid exceeds natural buffer capacity.

$$CaCO_3\ (s) + H_2O\ (l) + CO_2\ (g) \rightarrow Ca^{2+}\ (aq) + 2HCO_3^-\ (aq) \qquad (6)$$

$$HCO_3^-\ (aq) + H^+\ (aq) \rightarrow CO_2\ (g) + H_2O\ (l) \qquad (7)$$

As there is very little other bases present, the concentration of $HCO_3^-$ is termed the Acid Neutralizing Capacity (ANC). As ANC increases, pH also increases as seen in Figure 2. A stream with a higher concentration of bicarbonate is capable of buffering out more acid and being less susceptible to a pH depression.

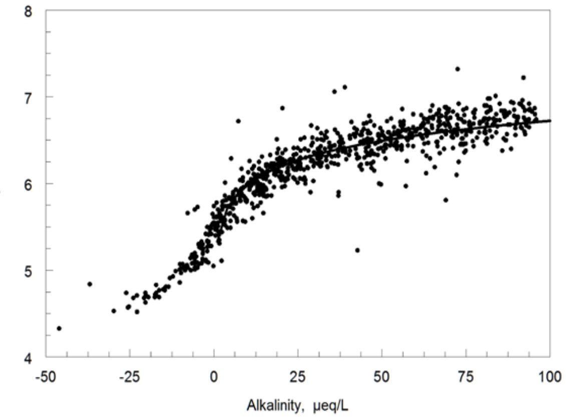


Figure 2. Relationship between pH and alkalinity (µeq/L) for 800 USFS streams monitored by our research group from 2002-2015.

Studies done in the 1980s to assess the effects of acid rain on the water chemistry of Virginia streams and aquatic life estimated that an average 29 µeq/L ANC was lost compared to preindustrial age water chemistry. In addition, 22 µeq/L sulfate ($SO_4^{-2}$) from natural atmospheric deposition had been added.

In the 1990s, The Clean Air Act (CAA) put restrictions on emissions of around 687 power plants and 19 hundred units within the continental United States.[2] One example is a 1.6 GW coal fired power generating station in West Virginia; Mount Storm. Due to the CAA the station was retrofitted with two limestone scrubbers in the late 1990s which significantly reduced emission, Figure 3.[2]

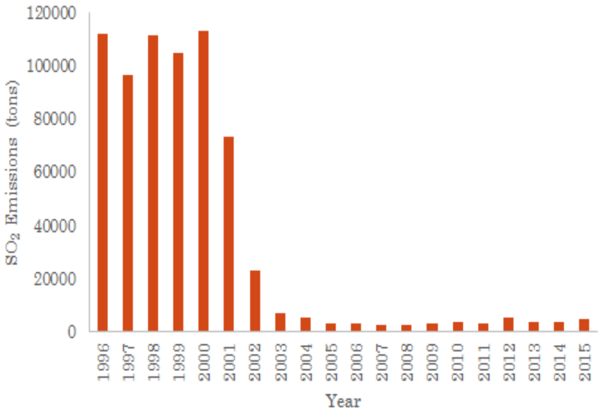


Figure 3. $SO_2$ emissions (Tons) from Mt. Storm Power Station from 1996-2015.

It was thought that the reductions in emissions and deposition might result in “improvements” in stream water chemistry for streams that have been adversely affected. The purpose of the present study was to examine the data from acid monitoring stations in our region and compare that with stream data we have collected. Three acid sensitive streams that we routinely sample were selected for detailed evaluation: Little Stony Creek (LS), Mill Creek (MC), and Mountain Run (MR) presented in Table 1.

Table 1. Location, watershed size and geology of the three streams being evaluated

| | **Little Stony Creek** | **Mill Creek** | **Mountain Run** |
|---|---|---|---|
| **Location** | Shenandoah County, Va | Shenandoah County, Va | Rockingham County, Va |
| **Watershed (Ha)** | 1015 | 388 | 155 |
| **Geology** | Sandstone and shale | Sandstone and shale | Sandstone |
| **Year Monitoring Began** | 1987 | 1987 | 1992 |
| **Frequency** | Monthly | Monthly | Monthly |

When a stream is acid sensitive, aquatic biota and fauna tend to struggle and show depressed biomass as atmospheric acid is introduced and is of concern to wildlife management agencies including the United States Forest Service (USFS) and the Virginia Department of Game and Inland Fisheries (VDGIF). By the late 1980s, it had been recommended that for certain streams artificial introductions of carbonate to the streams would enable temporary restoration of water chemistry to offset the acid loads.[1] The best available material is limestone and the process of addition is known as “liming.” Since then we have designed and monitored more than two dozen sites in western Virginia where liming is used to enhance stream water chemistry.

A number of parameters were evaluated so for effective and long term treatment including quality of limestone, particle size, streamflow regime, morphology, cost and stream access. Several models were created to predict the amount of limestone needed. The “lost” ANC model, based on the 1980s data, estimated the mass of limestone needed to restore the average 29 µeq/L. Another model, the target model estimated to add enough limestone to bring the stream to target values of pH > 6.5 and ANC > 25 µeq/L.[3] A deposition based model has been developed since more widespread deposition data have become available from NADP. This model is currently the preferred prediction approach for limestone dose in current liming projects. It estimates to add the amount of limestone to equal the annual deposition of acid. A fourth model is the sulfate equivalence model which gives the amount of limestone necessary to match the concentration of sulfate in the stream water. This model suffers where sulfate is present in the streams that have originated from geologic sources other than acid rain. Regardless of which model is used for ongoing liming projects it is important to assess the effect of acid reductions. Thus, the secondary purpose of this study was to ascertain whether or not the current liming projects can be modified or even terminate as a result of these changes. New estimates of limestone dose and frequency based on stream water chemistry changes resultant from the CAA could save cost and labor.

## 2. Methodology

This project took advantage of a large amount of data, both from our own research and governmental website databases. Resources included power plant emissions reduction, NADP data, and weather data.[2, 4, 5] Although our database of stream water chemistry data extends back about 30 years, we used data in the twenty year period 1996-2015 to match the timing of the CAA as well as the availability of government data. Atmospheric data were taken from the NADP website from three stations that are regionally located with the vicinity of the study streams: Va28 Big Meadows Monitoring station, Va00 Charlottesville Monitoring station, and W\V18 Parsons Monitoring station. Weather data were taken from Dale Enterprise in Rockingham County, Virginia and Wardensville in Hardy County, West Virginia. Our research group has collected water chemistry data by previously described methods.[1, 6]

## 3. Results

The ionic composition of stream water includes cations and anions present from other precipitation or weathering of geologic material as follow [Equation (8)]:

$$(H^+) + (Ca^{2+}) + (Mg^{2+}) + (Na^+) + (K^+) + (NH_4^+) = (Cl^-) + (NO_3^-) + (SO_4^{2-}) + (HCO_3 -) \qquad (8)$$

Sulfate ion increase in stream water results in an increase in acidity and replacement of bicarbonate (Galloway 1983). It is thought that emissions reductions would enable restoration of bicarbonate and increase pH of the stream. Data for $SO_4^{2-}$ concentration in rainwater from the NADP was available as annual averages expressed in µeq/L, Figure 4, for the three stations. Linear fitted values were plotted to reveal the trends listed in Table 2. Scatter is large and is the result of natural events, including high rainfall or low rainfall and other weather patterns, but it is apparent that the three sites have experienced dramatic reductions since CAA went into effect. In 1996 the average $SO_4^{2-}$ concentration was 41.4 µeq/L and by 2015 it was down to 12.5 µeq/L, a 70% reduction.

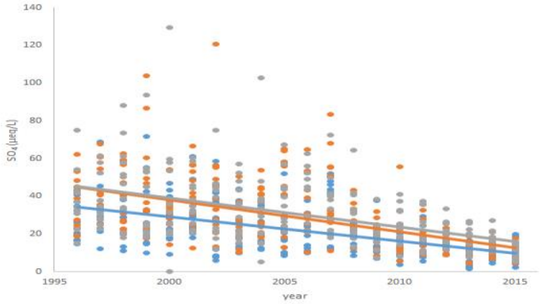


Figure 4. Sulfate concentration values for rainwater taken at three NADP monitoring stations: Va28 (blue), Va00 (orange), and Wv18 (gray) from 1996 to 2015 with fitted trend lines.

The reduction in sulfate was expected to produce a concurrent reduction in the sulfate concentration of the streams and a corresponding ANC increase. Annual averages of all three streams were calculated for these two parameters and trends evaluated. For Little Stony Creek the sulfate concentration decreased, while the ANC increased during the twenty year period of evaluation, evident in Figure 5. Similar trends were observed for the other streams, Mountain Run and Mill Creek.

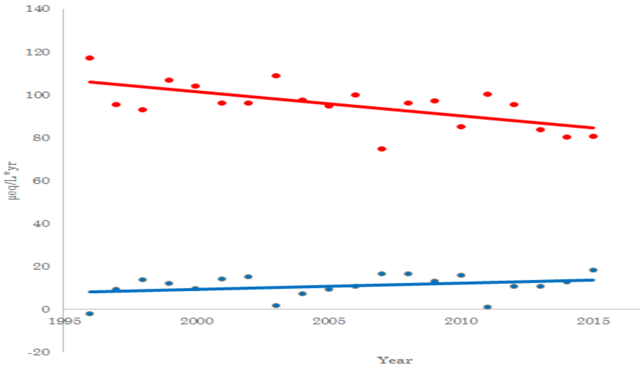


Figure 5. Sulfate (red) and ANC (blue) concentrations for Little Stony Creek with fitted linear lines 1996-2015.

Linear trend lines were generated for the three streams described in Table 2**.** Trend lines used to give annual average estimates of concentrations at the beginning and end of the twenty year period. Low correlation coefficients were observed but that was expected for natural samples subject to extreme changes in flow and precipitation. Sulfate and ANC concentration are inversely related in the charge balance equation [Equation (8)]. With a decrease in sulfate injection, more ANC was present in the stream. The slope for the ANC increase is less than that for the reduction of $SO_4^{2-}$ which supports one model of stream acidification, which predicted that stream water response to decreases in deposition would require time.[7]

Table 2. Calculated trend lines for ANC and $SO_4^{2-}$ concentration for the three streams are presented with the corresponding linear regression values.

| | **Little Stony** | | **Mill Creek** | | **Mountain Run** | |
|---|---|---|---|---|---|---|
| | Trend Line (y=) | $R^2$ | Trend Line (y=) | $R^2$ | Trend Line (y=) | $R^2$ |
| ANC | .29x–573.93 | 0.1012 | 0.14x–285.12 | 0.2656 | .35x–735.15 | 0.2224 |
| $SO_4^{2-}$ | 1.13x+2369.1 | 0.4167 | –1.30x+2722.6 | 0.6191 | 1.75x+3596.9 | 0.5697 |

Sulfate concentration values and annual average volumes of rainfall were used to determine the annual mass of deposited sulfate in the watershed, Table 3. In addition mass of sulfate discharged by the streams was calculated from the average annual concentration values and discharge data.

Table 3. Sulfate (tonnes/year) deposited and discharged in the three study watersheds 1996-2015 with difference values.

| | Tonnes/year | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| Year | LS input | LS output | LSΔ | MC input | MC output | MCΔ | MR input | MR output | MRΔ |
| 1996 | 23.5 | 38.6 | –15.2 | 9.0 | 15.8 | –6.8 | 4.0 | 8.8 | –4.9 |
| 1997 | 14.2 | 13.0 | –1.2 | 5.5 | 4.8 | 0.7 | 2.6 | 4.4 | –1.9 |
| 1998 | 16.6 | 19.8 | –3.2 | 6.4 | 9.4 | –3.0 | 2.9 | 4.6 | –1.8 |
| 1999 | 15.8 | 18.5 | –2.7 | 6.1 | 6.5 | –0.4 | 2.3 | 3.5 | –1.2 |
| 2000 | 12.5 | 19.4 | –6.9 | 4.8 | 8.3 | –3.5 | 2.2 | 3.2 | –1.0 |
| 2001 | 13.1 | 17.1 | –4.0 | 5.1 | 7.5 | –2.4 | 1.9 | 2.7 | –0.9 |
| 2002 | 19.4 | 25.3 | –5.8 | 7.5 | 10.5 | –3.0 | 2.5 | 3.7 | –1.2 |
| 2003 | 18.9 | 36.8 | –17.9 | 7.3 | 14.3 | –7.0 | 2.8 | 7.2 | –4.4 |
| 2004 | 14.9 | 23.2 | –8.3 | 5.8 | 9.6 | –3.8 | 2.3 | 4.4 | –2.1 |
| 2005 | 12.3 | 16.4 | –4.2 | 4.7 | 6.8 | –2.1 | 1.9 | 2.9 | –1.0 |
| 2006 | 14.9 | 23.4 | –8.5 | 5.7 | 9.7 | –4.0 | 2.5 | 4.5 | –2.0 |
| 2007 | 13.6 | 12.5 | 1.1 | 5.3 | 6.5 | –1.3 | 2.3 | 3.2 | –0.9 |
| 2008 | 11.0 | 21.3 | –10.3 | 4.3 | 9.2 | –5.0 | 1.6 | 3.6 | –2.0 |
| 2009 | 9.3 | 19.6 | –10.4 | 3.6 | 8.8 | –5.2 | 1.4 | 3.9 | –2.6 |
| 2010 | 7.5 | 14.9 | –7.4 | 2.9 | 6.2 | –3.4 | 1.0 | 2.6 | –1.6 |
| 2011 | 10.6 | 28.3 | –17.7 | 4.1 | 11.7 | –7.6 | 1.6 | 4.3 | –2.6 |
| 2012 | 6.2 | 17.5 | –11.3 | 2.4 | 6.9 | –4.5 | 1.1 | 2.9 | –1.8 |
| 2013 | 7.0 | 22.0 | –14.9 | 2.7 | 10.1 | –7.4 | 1.0 | 4.0 | –3.0 |
| 2014 | 4.7 | 13.2 | –8.5 | 1.8 | 5.7 | –3.9 | 1.1 | 3.7 | –2.6 |
| 2015 | 4.2 | 15.2 | –11.1 | 1.6 | 6.4 | –4.8 | 0.8 | 3.4 | –2.6 |

Sulfate deposition in 2015 for Little Stony Creek watershed was 4.2 tonnes, which was an 82% reduction from 1996. The decrease in sulfate deposition was generally accompanied by a decrease in sulfate being discharged. No consistent trend was observed in the difference of deposition and discharge. Similar trends were observed for both MC and MR, with 82% reduction and 85% reduction, respectively. Absolute values differed due to watershed size differences

As had been anticipated, in general the sulfate deposition reductions were accompanied by stream water ANC increases. The masses of ANC discharged annually were calculated and presented in Table 4, in a similar fashion to sulfate. There is no ANC deposited in the watershed due as it originates entirely from the weathering of geology. The calculated discharge of ANC for Little Stony Creek rose from –0.8 tonnes in 1996 to 4.6 tonnes in 2015, which is a 117% increase.

Table 4. ANC (tonnes/year) discharged from the three study streams 1996-2015.

| | Tonnes/year | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| Year | LS input | LS output | LSΔ | MC input | MC output | MCΔ | MR input | MR output | MRΔ |
| 996 | 0.0 | –0.8 | 0.8 | 0.0 | –1.8 | 1.8 | 0.0 | –2.6 | 2.6 |
| 997 | 0.0 | 2.1 | –2.1 | 0.0 | –0.7 | 0.7 | 0.0 | –1.2 | 1.2 |
| 998 | 0.0 | 3.4 | –3.4 | 0.0 | –1.0 | 1.0 | 0.0 | –1.6 | 1.6 |
| 999 | 0.0 | 2.2 | –2.2 | 0.0 | –0.8 | 0.8 | 0.0 | –1.0 | 1.0 |
| 000 | 0.0 | 2.1 | –2.1 | 0.0 | –0.8 | 0.8 | 0.0 | –0.8 | 0.8 |
| 001 | 0.0 | 2.4 | –2.4 | 0.0 | –0.6 | 0.6 | 0.0 | –0.8 | 0.8 |
| 002 | 0.0 | 3.5 | –3.5 | 0.0 | –0.8 | 0.8 | 0.0 | –1.0 | 1.0 |
| 003 | 0.0 | 0.6 | –0.6 | 0.0 | –1.5 | 1.5 | 0.0 | –1.6 | 1.6 |
| 004 | 0.0 | 1.8 | –1.8 | 0.0 | –0.8 | 0.8 | 0.0 | –1.3 | 1.3 |
| 005 | 0.0 | 1.7 | –1.7 | 0.0 | –0.6 | 0.6 | 0.0 | –0.8 | 0.8 |
| 006 | 0.0 | 2.9 | –2.9 | 0.0 | –1.0 | 1.0 | 0.0 | –1.5 | 1.5 |
| 007 | 0.0 | 3.5 | –3.5 | 0.0 | –0.7 | 0.7 | 0.0 | –1.0 | 1.0 |
| 008 | 0.0 | 3.5 | –3.5 | 0.0 | –0.7 | 0.7 | 0.0 | –0.9 | 0.9 |
| 009 | 0.0 | 3.2 | –3.2 | 0.0 | –0.9 | 0.9 | 0.0 | –1.0 | 1.0 |
| 010 | 0.0 | 2.8 | –2.8 | 0.0 | –0.7 | 0.7 | 0.0 | –0.5 | 0.5 |
| 011 | 0.0 | 0.3 | –0.3 | 0.0 | –1.1 | 1.1 | 0.0 | –1.4 | 1.4 |
| 012 | 0.0 | 2.4 | –2.4 | 0.0 | –0.7 | 0.7 | 0.0 | –1.0 | 1.0 |
| 013 | 0.0 | 2.9 | –2.9 | 0.0 | –1.1 | 1.1 | 0.0 | –1.3 | 1.3 |
| 014 | 0.0 | 3.4 | –3.4 | 0.0 | –0.6 | 0.6 | 0.0 | –1.2 | 1.2 |
| 015 | 0.0 | 4.6 | –4.6 | 0.0 | –0.6 | –0.6 | 0.0 | –1.3 | 1.3 |

During the study period LS, MC, and MR had ANC increased by 19, 9, and 24.8%, respectively. The differences of the increases are due to differences in geology of the watersheds. Even though all three streams experienced increases in ANC, they remain within the acid sensitive categories, and MC and MR currently have negative ANC values. The ANC increase was brought on by the reduction of $SO_4^{2-}$ concentration within each stream. LS, MC, and MR had reductions of $SO_4^{2-}$ of 25.6, 25.6, and 42.8%. These values do not match the increase in ANC which reflects that the three streams are have yet to return to pre-industrial age values. So liming projects will need to be continued for the time being. For best recommendations of liming amounts for management agencies, the summary data of water chemistry was done in Table 5.

Table 5. Summary of annual average ANC and Sulfate concentration values (µeq/L) trends for the three streams from 1996 and 2016 with difference as percent gain or loss.

| , | **Little Stony Creek** | | **Mill Creek** | | **Mountain Run** | |
|---|---|---|---|---|---|---|
| | 1996 | 2015 | 1996 | 2015 | 1996 | 2015 |
| ANC (µeq/L) | 8.1 | 13.6 | – 11.1 | – 8.5 | – 27.2 | –20.4 |
| % Δ | | + 19.0 | | + 9.0 | | + 24.8 |
| $SO_4^{-2}$ (µeq/L) | 106.0 | 84.5 | 118.8 | 94.0 | 99.7 | 66.4 |
| % Δ | | – 25.6 | | – 25.6 | | – 42.8 |

As mentioned above there are four methods we have used to estimate limestone dosage: “Lost” ANC, “Target” pH and ANC, Total Acid Titration, and Sulfate Equivalence. Using the fitted linear annual averages presented above, the amount of limestone for the respective methods were calculated in Table 6 as follows. In the “Lost” ANC model, LS had an ANC of 8.1 µeq/L in 1996; i.e., 21 µeq/L of “lost” ANC. By 2015 the ANC had increased to 13.6 µeq/L. The predicted annual limestone mass requirement in 1996 for this model was 106 tonnes, but had fallen to 85.5 tonnes in 2015. Similar trends were observed with MC and MR with respective reductions of 76 and 81% in limestone requirement amounts. In the Target model, the 1996 limestone dose needed to achieve an ANC of 25 µeq/L was 4.05 tonnes; raising the ANC by 16.9µeq/L. By 2015 the limestone needed reduced by 39.9% to 2.73 tonnes, raising the ANC by 11.4µeq/L. Similar reductions trends were seen in MC (74.7%) and MR (14.1%). In 1996, 13.50 tonnes of limestone was needed to mitigate acidification using the Titration model for LS. There has been a 91.7% reduction by 2015, only needing 5.01 tonnes to achieve the same effect. MC and MR experienced similar reductions of 91.6 and 91.8% respectively. In LS, the amount of limestone needed to fulfil the requirements of the sulfate model was 20.1 tonnes; by 2015 there was a 29.1% difference. MC and MR had similar reductions of 29.5 and 54.7%, respectively.

Table 6. Mass requirements of limestone +dose for each year in 1996 and 2015.

| | Little Stony Creek (Tonnes/Year) | | Mill Creek (Tonnes/Year) | | Mountain Run (Tonnes/Year) | |
|---|---|---|---|---|---|---|
| Model | 1996 | 2015 | 1996 | 2015 | 1996 | 2015 |
| “Lost” ANC Replacement | 6.95 | 6.32 | 2.55 | 1.14 | 1.43 | 0.60 |
| “Target” pH & ANC | 4.05 | 2.73 | 3.18 | 1.45 | 2.58 | 2.24 |
| Total Acid Titration | 13.50 | 5.01 | 4.95 | 1.84 | 2.78 | 1.03 |
| Sulfate Equivalence | 20.1 | 15.0 | 8.53 | 6.34 | 3.84 | 2.19 |

## 4. Conclusion

The three streams: LS, MC, and MR experienced pH and ANC depression from an increase of atmospheric acid brought on by the burning of fossil fuels. The Clean Air Act put restrictions on power plant emissions of $SO_2$ in order to reduce the amount of atmospheric acid by the late 1990s. The atmospheric condition and stream chemistry from 1996 to 2015 were evaluated to determine if the CAA reductions were being realized within the streams. Since the implementation of the Clean Air Act there has been an overall reduction of 82% in $SO_2$ emissions. The emission reduction has led to an 82% reduction in sulfate deposition in the study stream watersheds that resulted in a 25.6, 25.6, and 42.8% reduction of sulfate concentrations in LS, MC, and MR respectively. These reductions have yet to be fully realized in ANC with only a 19.0, 9.0, and 24.8% increases in the LS, MC, and MR, respectively. The mitigation dose estimates used to reduce the effects of acidification were evaluated and it was found that lesser amounts are now needed, but liming must continue to maintain water chemistry in treated streams at the present time.

## 5. Acknowledgements

The authors greatly appreciate financial support by The US Forest Service (George Washington and Jefferson National Forests), Virginia Department of Game and Inland Fisheries, The National Science Foundation and James Madison University for a variety of grants and cooperative agreements used to collect data for this study. Too many people have been involved to name everyone but we greatly appreciate the many staff scientists of the above agencies who have participated. We also appreciate the work of the many students of the Downey lab research group who have been involved in the research for the past three decades.

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