ATTACHMENT A

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The Impact of Coal Combustion Residue Effluent on Water Resources: A North Carolina Example

Laura Ruhl,[†,‡] Avner Vengosh,[*,†] Gary S. Dwyer,[†] Heileen Hsu-Kim,[§] Grace Schwartz,[§] Autumn Romanski,[#] and S. Daniel Smith[#]

[†]Division of Earth and Ocean Sciences, Nicholas School of the Environment, Duke University, Durham, North Carolina 27708, United States

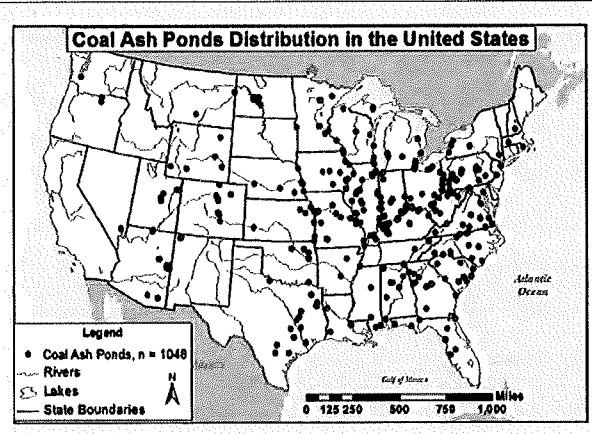
[‡]Department of Earth Science, University of Arkansas at Little Rock, Little Rock, Arkansas 72204, United States

[§]Civil and Environmental Engineering, Pratt School of Engineering, Duke University, Durham, North Carolina 27708, United States

[#]Surface Water Protection Section, Division of Water Quality, North Carolina Department of Environment and Natural Resources, Raleigh, North Carolina 27609, United States

Supporting Information

ABSTRACT: The combustion of coal to generate electricity produces about 130 million tons of coal combustion residues (CCRs) each year in the United States; yet their environmental implications are not well constrained. This study systematically documents the quality of effluents discharged from CCR settling ponds or cooling water at ten sites and the impact on associated waterways in North Carolina, compared to a reference lake. We measured the concentrations of major and trace elements in over 300 samples from CCR effluents, surface water from lakes and rivers at different downstream and upstream points, and pore water extracted from lake sediments. The data show that CCR effluents contain high levels of contaminants that in several cases exceed the U.S. EPA guidelines for drinking water and ecological effects. This investigation demonstrates the quality of receiving waters in North Carolina depends on (1) the ratio between effluent flux and freshwater resource volumes and (2) recycling of trace elements through adsorption on suspended particles and release to deep surface water or pore water in bottom sediments during periods of thermal water stratification and anoxic conditions. The impact of CCRs is long-term, which influences contaminant accumulation and the health of aquatic life in water associated with coal-fired power plants.



INTRODUCTION

Numerous studies have shown that effluents generated from leaching of coal combustion residues (CCRs) typically have high concentrations of toxic elements.[1,2] Yet, the overall impact of disposed CCR wastes on the quality of water resources in the U.S. is largely unexplored, except in a few specific case-studies, such as CCR spills.[3,4] In the U.S., approximately six hundred power plants[5] generate 130 million tons of CCRs annually,[6] of which 56% is stored in surface impoundments and landfills, while the remaining are reused for concrete, cement, and construction industries.[7] CCRs, encompassing fly ash, bottom ash, and flue gas desulfurization (FGD) material, represent one the largest industrial waste streams in the U.S. and are not classified as hazardous waste.[8] Despite the large volume of CCR effluents generated annually and their disposal into hundreds of surface water bodies, the environmental risks associated with these disposal practices are not well-known. Moreover, because of the lack of CCR waste data,[9] the effluents that are discharged from coal fired power plants and permitted by the national and state regulatory bodies lack consistent monitoring and limit requirements that are relevant to composition of CCR effluents.

Water in coal-fired power plants is used in steam production and cooling, as well as the transport of CCRs from the plant to holding ponds. In spite of some losses,[10] the residual effluent water is discharged to the environment and is permitted through the National Pollution Discharge Elimination System (NPDES) Program. The NPDES Program as established by the Clean Water Act requires the control and permitting of point source discharges of wastewater.[11] Although the NPDES regulations for CCR effluents disposal vary between states, in most cases they consist of only limited factors. For example, NC regulations follow the federal guiding permit limits for effluent discharge that include only total suspended solids and

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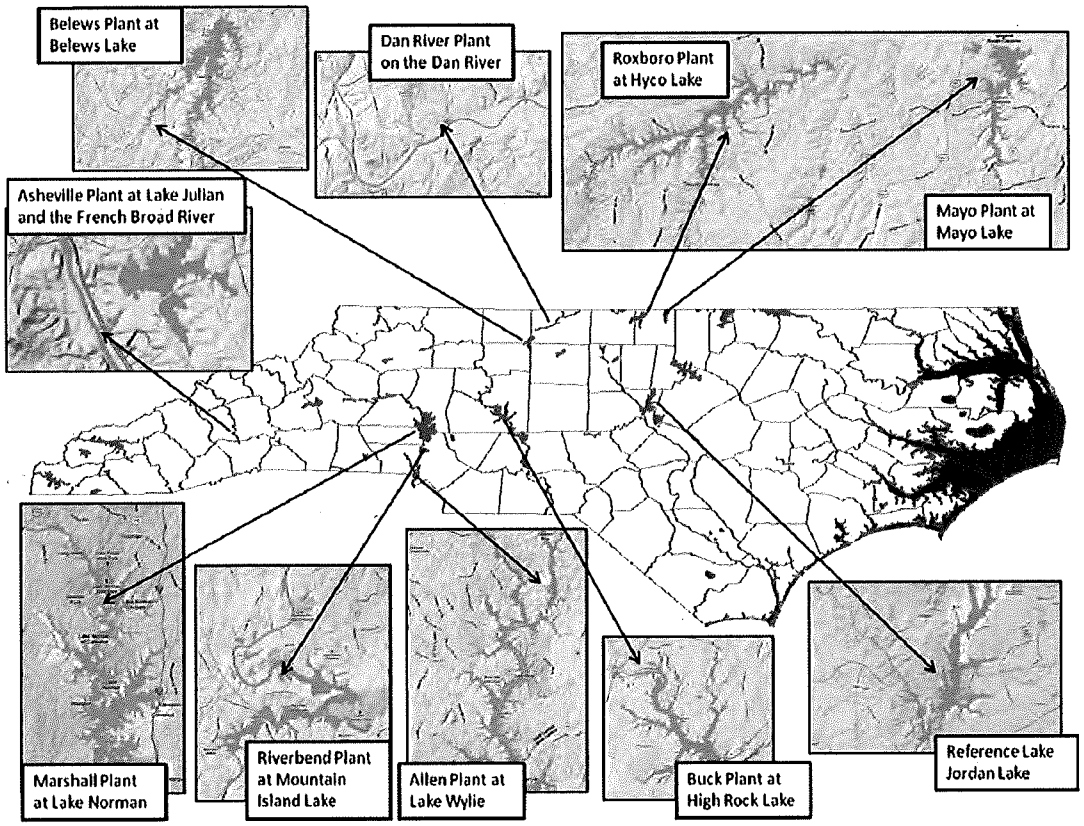

Figure 1. Map of coal-fired power plants and CCR disposal sites to waterways in North Carolina that were investigated in this study. Also included is a reference lake (Jordan Lake).

Table 1. Background Information on the Investigated Coal-Fired Electrical Power Plants, CCR Effluent Discharge through NPDES Outfalls, and Associated Waterways in North Carolina[a]

coal-fired power plant site	owner	size (MW)	town location	av. ash pond discharge flow (MGD)	av. once-through cooling H_2O flow (MGD)	water body	basin	scrubber system	effluent sampling
Roxboro Steam Station	Progress Energy	2558	Semora	11	1007	Hyco Lake	Roanoke	Wet FGD System	indirect
Mayo Stream Station	Progress Energy	745	Roxboro	7	unkown	Mayo Reservior	Roanoke	Wet FGD System	indirect
Allen Steam Station	Duke	1140	Belmont	15	5	Calawba River/Lake Wylie	Calawba	Wet FGD System	indirect
Marshall Steam Station	Duke	2090	Terrell	8	1463	Calawba River/Lake Norman	Calawba	Wet FGD System	indirect
Belews Greek Steam Station	Duke	2240	Wallnut Cove	9	1256	Dan River/Belews Lake	Roanoke	Wet FGD System	
Asheville Steam Station	Progress Energy	376	Arden	3	251	French Broad River/ Lake Julian	French Broad	Wet FGD System	direct
Riverbend Steam Station	Duke	454	Mount Holly	4	375	Calawba River/ML Island Lake	Catawba	None	direct
Buck Steam Station	Duke	369	Salisbury	4	258	High Rock Lake	Yadkin-PeeDee	None	indirect
Dan River Steam Station	Duke	276	Eden	1	201	Dan River	Roanoke	None	direct
Reference Lake	No Power Plant	N/A	Apex	N/A	N/A	B.Everell Jordan Lake	Cape Fear	None	N/A

[a]The size (in megawatts) of the plants as well as the amount of water discharged (in million gallons per day) in each plant are reported. Also listed is the reference site, Jordan Lake.

oil and grease[12] but do not include other constituent limits that could be relevant to CCR effluents, unless written in specifically by the permitting body.

In recent years, air regulations have become more stringent (e.g., Interstate Clean Air Rule and Clean Air Act), requiring the capture of potential atmospheric pollutants, like sulfur

oxides (SO_x). The FGD process effectively removes many of the volatile elements associated with the SO_x. Most coal-fired power plants in the U.S. do not have FGD scrubbers, but those with FGD capabilities produced 58% of the electricity generated from coal in the U.S. in 2010.[13] Of those with FGD scrubbers, most (up to 88% in 2010) use a wet FGD disposal system.[1] This process results in cleaner air emissions, but the trade-off is significant enrichments of contaminants in solid wastes and wastewater discharged from power plants. Several studies have shown that groundwater near these CCR disposal facilities was contaminated by CCR leachates,[14] and wildlife poisoning and environmental damages from CCR impoundments.[15] Yet, the long-term impact of CCR effluents is poorly studied in surface water surrounding coal-fired power plants.

This study aims to investigate the impact of CCR disposal on surface water surrounding coal-fired power plants in North Carolina. We systematically document the quality of discharged effluents from ten CCR effluent and cooling water discharge sites and the impact on associated waterways (lakes, rivers), in addition to a reference (control) lake (Figure 1; Table 1). We measured the concentrations of major and trace elements in 76 CCR effluent samples, 129 surface water samples from lakes and rivers from different downstream and upstream (background) sites, and 98 pore water samples extracted from the lake sediments. The study is based on an investigation of monthly sampling over one year at two lakes (Hyco and Mayo) and a single sampling for ten other waterways (Table 1, Figure 1).

METHODS

During August 2010 to February 2012, a total of thirty-six field trips were made to the research sites in North Carolina (Figure 1, Supporting Information Table S1) with over 300 surface and pore water samples were collected. Samples were collected monthly from Hyco and Mayo Lakes from August 2010 through August 2011. The other investigated water resources were Lake Norman, Mountain Island Lake, Lake Wylie, High Rock Lake, Belews Lake, Dan River, French Broad River, Lake Julian, and Jordan Lake as a reference lake (Figure 1). These bodies of water were sampled during the summer of 2011, with the exception of Mountain Island Lake, which was sampled both during the summers of 2010 and 2011. Water sampling procedure strictly followed USGS protocols.[16] Water samples were taken at various depths with a Wildco Beta water sampler (for trace metals) to capture variations in the water column induced by the epilimnion and hypolimnion during lake water stratification. Cations and trace metals were measured in both dissolved and total forms. After filtration of samples in the field (0.45 μm syringe filters), trace elements were measured by inductively coupled plasma mass spectrometry (ICP-MS), major elements by direct current plasma optical emission spectrometry (DCP-OES), and anions by ion chromatography (IC) at Duke University. Pore water was extracted from lake bottom sediments obtained using a Wildco box core (up to 25 cm depth), then vacuum filtration or centrifugation to extract the pore water. Inorganic arsenic species were measured using the Bednar method,[17] in which the uncharged arsenic species As^{III} was separated from pore water through an anion exchange resin cartridge and preserved in the field. Trace elements were measured with a VG PlasmaQuad-3 (Thermo Fisher Scientific Inc.) inductively coupled plasma mass spectrometry (ICP-MS) and major elements with an ARL SpectraSpan 7 (Thermo Fisher Scientific Inc.) direct current plasma optical emission spectrometry (DCP-OES). Both instruments were calibrated to the National Institute of Standards and Technology 1643e standard, which was used at varying concentrations before, after, and throughout sample runs. Internal standards of In, Th, and Bi were spiked into all samples prior to measurement on the ICP-MS. The detection limit of the ICP-MS of each element was determined by dividing three times the standard deviation of repeated blank measurements by the slope of the external standard. The resulting values were then averaged (n = 4) and are reported for trace elements measured on the ICP-MS in Supporting Information Table S2. Analytical precision was calculated as the relative percent difference (RPD) of the results of duplicate sample measurements and is also reported in Supporting Information Table S2.

RESULTS AND DISCUSSION

Quality of Discharged Effluent. This study documented elevated contaminant concentrations in CCR effluents discharged from coal-fired power plants into receiving waters in NC (Figures 2 and 3; Supporting Information Table S2). For example, during the Summer 2011 sampling, the Asheville and Riverbend Plant outfalls contained arsenic concentrations above the EPA drinking water standard of 10 μg/L with concentrations reaching 44.5 μg/L and 92 μg/L, respectively. Mayo NPDES discharge yearlong average selenium concentration (5.6 ± 5.4 μg/L) exceeded the 5 μg/L EPA Chronic Criterion Concentration (CCC) for aquatic life. Several of the individual monthly sampling events at the Mayo NPDES outfall showed Se concentrations almost 4 times the CCC limit, as high as 19 μg/L (Figure 3). The summer sampling event at the Asheville Plant revealed selenium concentrations over 17 times the CCC (87.2 μg/L). The NPDES outfall for the Asheville plant also exceeded other human and aquatic life benchmarks, including antimony above the EPA's MCL (6 μg/L) at 10.9 μg/L, cadmium exceeded the fresh water aquatic life (EPA CCC) standard (0.25 μg/L) at 0.8 μg/L, and thallium concentrations were greater than the 2 μg/L EPA MCL at 2.9 μg/L. The outfalls were sampled directly from the outfall in some sites, but in others from the water near the outfall, where direct sampling was not accessible (Table 1). Thus, the data at some of the outfall sites (Roxboro, Mayo, Marshall, and Allen)

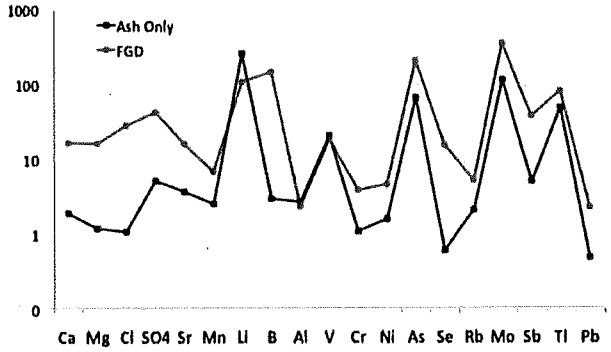

Figure 2. Mean values of enrichment factors of dissolved constituents in CCR effluents disposed from plants with an FGD system (red) and without an FGD system (blue). The enrichment factors were calculated by the ratio of different elements concentrations in directly sampled CCR effluents to the concentrations in the upstream water that feeds each plant.

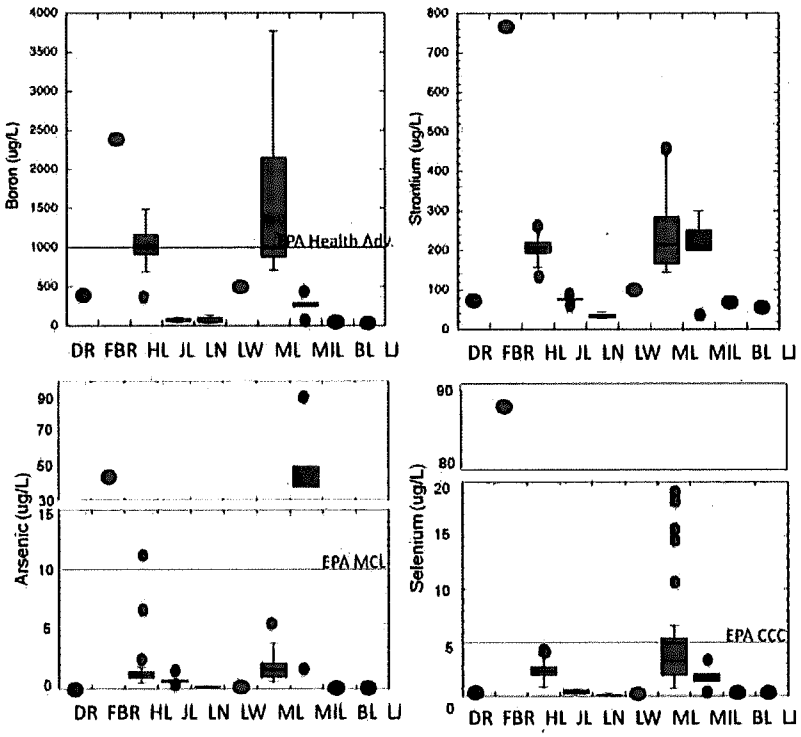

Figure 3. Concentration ranges of selected contaminants in CCR effluents from coal plants in NC. Red symbols correspond to plants with combined coal ash and FGD systems, blue symbols for only coal ash (without FGD), green for the reference lake (Jordan Lake), and black for cooling water separated from CCR effluents. The EPA drinking water (MCL) and ecological (CCC) benchmarks are referenced. Sites include the Dan River (DR), French Broad River (FBR), Hyco Lake (HL), Jordan Lake (JL), Lake Norman (LN), Lake Wylie (LW), Mayo Lake (ML), Mtn. Island Lake (MIL), Belews Lake (BL), and Lake Julian (LJ) that are shown in Figure 1.

underestimate the full extent of the CCR waste stream contaminant level of the discharge. In spite of efforts to reduce the levels of contaminants discharged through the NPDES outfall by using settling ponds, clarifier, bioreactor, or wetland at some sites[1,18] our data clearly show high contaminant levels that suggest the need for enhanced removal/wastewater treatment.

Many of the outfalls sampled consisted of wastewater from the FGD process that was subsequently diluted with the ash pond water (or other process water), and at some locations, also mixed with the cooling water (e.g., Roxboro plant at Hyco Lake; Supporting Information Table S3) prior to discharge at the outfall. Therefore for plants with an FGD system, the effluent concentrations represent some dilution of the original FGD wastewaters.[19,20] The data show that outfalls sampled from coal fired power plants with an FGD system ($n = 69$) had significantly higher concentrations of major ions (Ca, Mg, and Cl; $p < 0.01$) and minor constituents such as B ($p < 0.01$), Br ($p < 0.01$), and Cr ($p < 0.05$) relative to outfalls with only ash pond water or cooling water disposal ($n = 5$ and $n = 7$ respectively; Figure 3). The plants with no FGD system, but with wet ash disposal systems and subsequent discharges ($n = 5$), had higher concentrations of several constituents including As, V, Sb, Li, Tl, and Mo ($p < 0.01$) relative to effluents from FGD systems (Supporting Information Table S2). Selenium concentrations were also higher at FGD outfalls with several plants exceeding the EPA's CCC of 5 μg/L (Mayo at 19 μg/L and Asheville at 82 μg/L compared to Riverbend at <3.5 μg/L and Dan River at <DL of selenium). Overall, the CCR outfalls were enriched in many constituents compared to the upstream waters that feed them, and the FGD effluents had larger enrichments in many ions compared to the ash discharge only outfalls (Figure 2).

Annual fluxes of dissolved trace elements through CCR-effluent discharge into NC waterways show large variations (Supporting Information Table S4). The magnitude of the arsenic flux from CCR effluents exceeded the natural flux of the associated water system in some cases (Roxboro, Ashville, Mayo) and was lower in others (Dan River, Allen, Riverbend). The anthropogenic fluxes exceed the natural fluxes even in sites where the CCR discharge flow rate consisted of less than a percent of the natural water flow. The flux measurements reported in this study were also compared to the Toxic Release Inventory (TRI)[21] and show both consistent and inconsistent results (Supporting Information Table S4). The overall CCR fluxes of contaminants into NC waterways, such as B, As, and Se were 278, 0.7, and 0.8 (metric) tons per year, respectively. Yet the magnitude of As, Se, and Sb fluxes were significantly lower than flux values reported previously for CCR discharge to the Chattahoochee River, Georgia.[22]

In contrast to the CCR outfalls, separated cooling water effluents that were sampled in this study had much lower contaminant concentrations, which did not exceed any of the human or aquatic life benchmarks (Supporting Information Table S2) and were not enriched in any constituents compared to their respective upstream waters and reference lake (Jordan Lake) (Supporting Information Table S2). Consequently, in outfall sites where CCR effluents and cooling water were blended, the contaminant level was significantly reduced. For example, in Hyco lake, where the cooling water constitutes

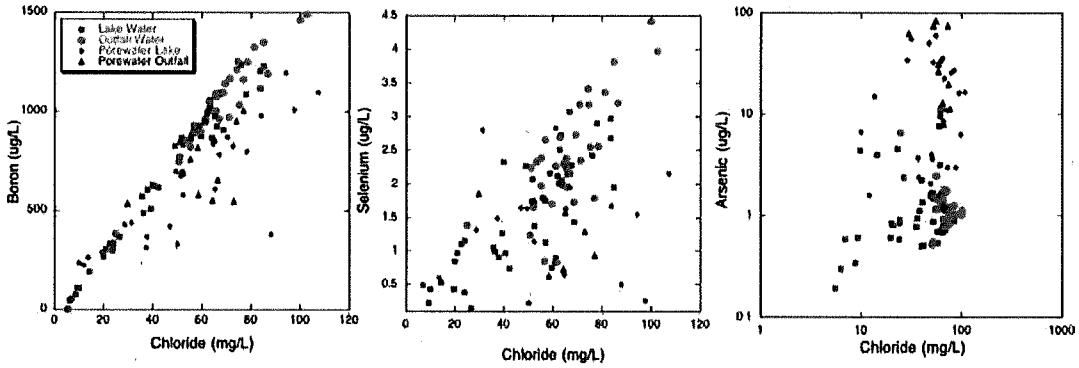

Figure 4. Boron, selenium, and arsenic versus chloride concentrations in Hyco Lake. The CCR effluent concentrations are marked with red circles, surface lake water by blue squares, porewater from outfall areas by black triangles, and porewater from downstream areas by green diamonds. Note the high correlation of boron with chloride in the lake water (i.e., a conservative behavior) relative to the low correlations of arsenic and selenium. The data show differential depletion of boron and selenium and enrichment of arsenic in pore water relative to lake water.

>98% of the effluent volume (Supporting Information Table S3) the contaminant levels of the NPDES outfall would be significantly higher if the cooling component was reduced or restricted (e.g., recirculating cooling water at Mayo Lake). The direct effluents from the FGD process and ash ponds at Roxboro were reported to have concentrations of As ranging between 1.6 and 394 μg/L and Se ranging 4.3–238 μg/L during the yearlong sampling (Supporting Information Table S3).[23,24] Therefore, cooling water has an important mitigating effect on the quality of NPDES outfalls in NC.

Impact on the Aquatic Systems. We further analyzed the impact of CCR effluents on the quality of receiving waters by systematically comparing the chemical composition in waters downstream of the disposal sites relative to upstream waters from the same river/lake and a reference lake that has no connection to coal plant discharge (Jordan Lake; Figure 1). The data show elevated concentrations, particularly for Ca, Mg, Sr, Li, B, V, Cr, Se, Mo, F, Cl, Br, SO_4^{2-} ($p < 0.01$), as well as for As and Tl ($p < 0.05$), in downstream water relative to upstream water. Likewise, the concentrations of Ca, Sr, Li, B ($p < 0.01$), as well as V, Se, and Mo ($p < 0.05$) were elevated in sites downstream of the outfalls relative to concentrations in the reference lake (Supporting Information Table S2).

In spite of the large dilution of effluent discharge, which plays a key role in reducing the dissolved constituents released to surface waters, we observed significant variations and differential impacts of various constituents after CCR release into the receiving waters. We grouped the major and minor elements according to their chemical behavior as monitored in Hyco and Mayo Lakes (Supporting Information Table S5). In Group 1, the concentrations of boron ($R^2 = 0.88$; Figure 4), calcium ($R^2 = 0.96$), strontium ($R^2 = 0.95$), bromide ($R^2 = 0.91$), and sulfate ($R^2 = 0.86$) in filtered water (0.45 μm) show linear correlations with chloride during the yearlong sampling (Figure 4, Supporting Information Figure S1, and Supporting Information Table S5), reflecting their conservative (i.e., nonreactive) behavior in the lake system. Thus dilution seems to be the key factor determining their concentrations in the affected rivers/lakes. The concentration of other elements (Se, Mg, Cr, V, and Ba defined as Group 2) in filtered water show a nonlinear correlation with Cl ($0.3 < R^2 > 0.6$) that suggests some attenuation in the lakes (e.g., sorption to particles). In contrast, As, Fe, and Mn that defines Group 3 show low or no correlation with chloride ($R^2 = 0.01$, 0.07, and 0.001, respectively; Supporting Information Table S3), indicating strong association with suspended particles in the water column. Higher dissolved concentrations of these constituents were observed at the bottom of the lake during periods of thermal stratification in the summer and low dissolved oxygen content (Supporting Information Figure S3). Seasonal stratification leads to the depletion of oxygen in bottom water during summer months and an overturning of the water column during the fall.[25] We hypothesize that under oxygenated water conditions, As oxyanions would be adsorbed onto Fe oxyhydroxides particles in the water column and bottom sediments.[26,27] During the stratification periods, when the bottom waters become anoxic, reductive dissolution of Fe (and Mn) oxyhydroxides results in release of dissolved As, Fe, and Mn to the bottom water. The reducing conditions would also convert arsenate (As(V)) into arsenite (As(III)), a neutrally charged form of arsenic at pH 7 (i.e., $H_3AsO_3^0$) that is less reactive toward sorption on oxyhydroxides[28,29] and also more toxic to wildlife.[30] The covariance of As with other redox sensitive elements like Fe and Mn during thermal stratification in Hyco Lake (Supporting Information Figure S2) supports this model.

In contrast, selenium does not increase with decreasing dissolved oxygen (and depth) in Hyco and Mayo Lakes, but rather shows a linear relationship with chloride, although with a relatively weak correlation ($R^2 = 0.65$; Figure 4 and Supporting Information Table S5), reflecting both dilution and retention effects. This is consistent with the selenium species geochemistry: under oxic conditions the oxidized species selenate (Se(VI)) would be less reactive toward sorption with oxyhydroxides and thus behave conservatively in the water column. In contrast, under anoxic conditions the partially reduced Se species selenite (Se(IV)) would have a strong sorption affinity for both oxyhydroxides[31] and clay minerals.[32,33] The most reduced forms of selenium (e.g., elemental Se^0 and FeSe) tend to persist as sparingly soluble minerals. Overall, a transition to anoxic conditions in the lake hypoliminion would result in lower dissolved Se concentrations.[34,35]

Bottom Lake Sediments and Pore Water. In addition to differential distribution of contaminants in the surface waters, this study revealed elevated levels of CCR contaminants (Supporting Information Table S6; Fe, Mn, Sr, As, Mo, Sb, Ni, V, and Br ($p < 0.01$), as well as Mg and F ($p < 0.05$)) in shallow

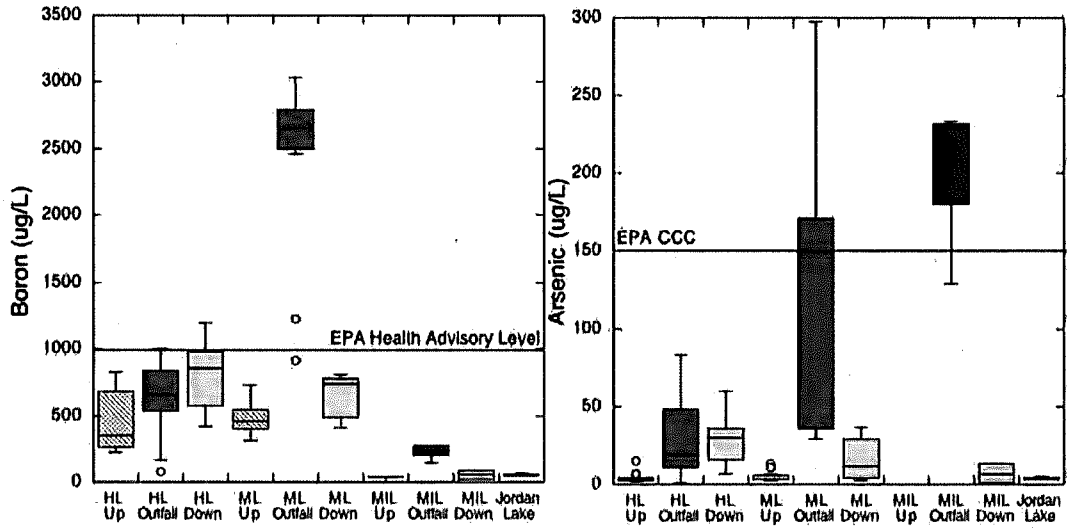

Figure 5. Boron and arsenic concentrations in porewater collected from upstream, outfall, and downstream sites of Hyco Lake (HL), Mayo Lake (ML), Mountain Island Lake (MIL), and Jordan Lake. Red symbols correspond to plants with combined coal ash and FGD systems, blue for only coal ash, and green for the reference lake (Jordan Lake). The EPA boron health advisory level is indicated, as well as the EPA CCC freshwater aquatic regulatory level.

pore water extracted from the lake bottom sediments that were significantly higher than those of the overlying bottom water. For example, As concentrations in pore water from Hyco, Mayo, and Mtn. Island lakes were as high as 83, 297, and 240 μg/L, respectively, exceeding the EPA's MCL (10 μg/L) and CCC (150 μg/L) standards (Figure 5). For comparison, the concentrations of trace elements (B and As, $p < 0.1$) in pore water from the reference lake were significantly lower than the CCR impacted lakes (Figure 5). We hypothesize that retention of CCR contaminants from the lake water via adsorption onto suspended matter in the water column results in accumulation of these contaminants in the sediments that are deposited on the lake bottom. Recent reports of higher concentrations of As and Se in lake bottom sediments at the outfall at Hyco and Mayo (As, 23 μg/g and 97 μg/g; Se, 8 μg/g and 10 μg/g dry weight, respectively) relative to the upstream branch of the lake (As, 6 μg/g and 12 μg/g; Se, 2 μg/g and 1.6 μg/g dry weight, respectively)[24,36] confirm that both As and Se are recycled through adsorption and desorption due to changes in the lake water chemistry, apparently induced from thermal stratification during the summer. Changes in the ambient conditions (pH, redox state) in the lake sediments would release these metalloids to the pore water.[4] We documented high levels of As in pore water and other redox-sensitive elements (Mn, Fe; Figure 5) that confirm this model. Additionally, direct arsenic speciation measurements show that over 82% of arsenic in the pore water collected at Hyco and Mayo Lakes were composed of the reduced and more mobile species arsenite (Supporting Information Table S7). In contrast, the Se concentrations were significantly higher in the CCR effluents and lake water relative to the pore water ($p < 0.01$) (Figures 3 and 5). This indicates that Se from CCR effluent can become associated with the sediment, but that Se species become immobilized by forming elemental selenium and metal–selenium complexes in the sediment, and therefore are not incorporated into the pore water.

Ecological and Environmental Implications. The accumulative nature of arsenic, selenium, and other CCR contaminants in lake systems could have ecological implications, particularly for benthic organisms and therefore the rest of the food chain. Indeed, elevated As and Se levels were reported in fish tissues from Hyco and Mayo Lakes especially near the NPDES outfall.[24,36] Furthermore, the 2010 Mayo Lake Environmental Report[36] showed deformities in some fish, including an extended lower jaw and spinal curvature, both of which are indicators of ingestion of high levels of Se.[37] If the base of the food chain is exposed to high levels of contaminants through the sediment and pore water, other organisms could be at risk if they feed on those organisms that live in the contaminated sediments and pore water.[38,39]

The impact of the effluent discharged from the NPDES outfalls on water quality in the downstream waterways is dependent on the flow rate to the river/lake (i.e., dilution effect; Supporting Information Table S4), residence time in the water body, as well as the mobilization (e.g., adsorption/desorption to sediment) properties of specific contaminants in the water. For example, the outfall on the French Broad River from the Asheville power plant had effluents with high contaminant concentrations (Supporting Information Table S2), but because of high river discharge flow, the downstream water was significantly diluted (although still detectable). A mass-balance calculation, using boron as a conservative tracer in surface water, show a contribution of 4.5% of CCR effluent into the downstream river with boron concentrations of 115 μg/L. It is important to note however, that these hydrologic systems could vary and be greatly affected by droughts. During the severe drought of 2007–2008 in North Carolina, the discharge of the French Broad River decreased drastically to just over 5 $m^3\ s^{-1}$, approximately 5 times lower than the river flow rate during the time of our sampling (25 $m^3\ s^{-1}$).[40] Using mass-balance calculation for conservative constituents, a 5-fold reduction in water flow would increase the CCR contribution up to 22% and would significantly increase the concentrations of such contaminants in the downstream river (e.g., boron up to 530 μg/L).

Our data also show CCR discharge into smaller lakes appears to have a greater impact relative to the larger lakes (e.g., Mayo Lake versus Lake Norman). This impact is therefore a combination of the volume of released CCR effluents, the lake inflows, plant water usage (removing from the lake system), and residence time. All of these factors can play a major role in the lake's water quality. For instance, Hyco and Mayo Lakes have boron concentrations of 958 μg/L and 703 μg/L, respectively compared to an upstream creek with boron concentrations of <3 μg/L and <7 μg/L, respectively (Supporting Information Table S2). This is a 300- and 100-fold enrichment in the boron content in the lakes. Conversely, Lake Norman, the largest lake in NC, 13−14 times the size of Hyco and Mayo Lakes, had only minor difference (12 μg/L) between its upstream the downstream boron concentrations. We conclude the smaller lakes and hydrological systems are more sensitive to CCR effluent contamination, particularly during drought periods when the dilution factor in the receiving water would be reduced. Moreover, as water regulatory agencies encourage power plants to install recycled cooling water systems rather than once-through cooling water as a way to conserve water, a potential unintended consequence of this policy is the discharge of CCR effluents with greater concentrations of CCR contaminants.

This study shows that coal-fired power plants that discharge their coal ash and FGD wastewaters had a significant effect on water quality of receiving waters of North Carolina. We show that even low concentrations of some contaminants, such as As with concentrations below health benchmarks at the NPDES outfall, could become problematic as As is retained in suspended sediments and remobilized with environmental changes in reduced bottom and pore waters. The results of this study have significant implications for hundreds of similar sites across the US given that CCR storage facilities continuously generate contaminants via leaching and transport to nearby hydrological systems. While this study focused on surface waters near CCR facilities, groundwater may have similar effects. Many CCR disposal ponds and landfills are not lined and, in many instances, are neither adequately monitored, nor regulated with respect to their effects on groundwater and surface waters. This study highlights the need for rigorous monitoring and clear regulations for limiting the CCR contaminants that are being discharged into U.S. waterways.

■ ASSOCIATED CONTENT

Supporting Information

Description of analytical techniques, additional quality control information, geo-references for sampling sites, along with supplementary graphs and figures. This material is available free of charge via the Internet at http://pubs.acs.org.

■ AUTHOR INFORMATION

Corresponding Author

*Tel: (919) 681-8050. Fax: (919) 684-5833. E-mail: vengosh@duke.edu.

Notes

The authors declare no competing financial interest.

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