nature sustainability

ARTICLES

https://doi.org/10.1038/s41893-021-00713-7

Addressing the contribution of indirect potable reuse to inland freshwater salinization

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Inland freshwater salinity is rising worldwide, a phenomenon called the freshwater salinization syndrome (FSS). We investigate a potential conflict between managing the FSS and indirect potable reuse, the practice of augmenting water supplies through the addition of highly treated wastewater (reclaimed water) to surface waters and groundwaters. From time-series data collected over 25 years, we quantify the contributions of three salinity sources—a water reclamation facility and two rapidly urbanizing watersheds—to the rising concentration of sodium (a major ion associated with the FSS) in a regionally important drinking-water reservoir in the Mid-Atlantic United States. Sodium mass loading to the reservoir is primarily from watershed runoff during wet weather and reclaimed water during dry weather. Across all timescales evaluated, sodium concentration in the reclaimed water is higher than in outflow from the two watersheds. Sodium in reclaimed water originates from chemicals added during wastewater treatment, industrial and commercial discharges, human excretion and down-drain disposal of drinking water and sodium-rich household products. Thus, numerous opportunities exist to reduce the contribution of indirect potable reuse to sodium pollution at this site, and the FSS more generally. These efforts will require deliberative engagement with a diverse community of watershed stakeholders and careful consideration of the local political, social and environmental context.

While historically a problem only in areas with arid and semi-arid climates, poor agricultural drainage practices, sodic soils and saline shallow groundwater[1,2], inland freshwater salinization is on the rise across many cold and temperate regions of the United States[3–6]. The trend is particularly notable in the densely populated Northeast and Mid-Atlantic[7–9] and agricultural Midwest[10–12] regions of the country. Globally, inland freshwater salinization has been reported in Canada, Finland, France, Greece, Italy, Iran and Russia[13]. The ions driving inland freshwater salinization vary by location and source but generally include a subset of the so-called major ions (defined here as Na^+, Ca^{2+}, Mg^{2+}, K^+, Cl^- and SO_4^{-2})[4]. Freshwater salinization is part of a broader change in the chemistry of many of Earth's inland freshwaters—including rising pH, alkalinity and base cation concentration—known as the 'freshwater salinization syndrome' (FSS)[6]. Human drivers include the use of deicers on roads and parking lots[7,14–19], water softener use[10,20], wastewater and industrial discharges[10,20,21], the weathering of concrete[7,22–25] and the accelerated weathering of geologic materials from the release of strong acids and human excavation of rock, which currently exceeds natural denudation processes by an order of magnitude[26,27]. In a recent modelling study, Olson[11] predicted that specific conductance (one measure of salinity) will increase >50% in more than half of US streams by 2100.

The FSS threatens freshwater ecosystem health and human water security. Chloride enrichment of streams is associated with declines in pollution-intolerant benthic invertebrates and loss of critical freshwater habitat[28]. Stream-borne salts can mobilize, through biogeochemical processes, previously sequestered contaminants (for example, nutrients and heavy metals) into sensitive ecosystems and drinking-water supplies[13,15,29,30], potentially reversing hard-won pollution reductions. Salinization of drinking-water supplies can mobilize lead, copper and other heavy metals from ageing drinking-water infrastructure through cation exchange and corrosion[31–33]. It can also alter the perception of potability—at high enough concentrations, sodium and other salts degrade the taste of drinking water[34]. The World Health Organization and the US Environmental Protection Agency (EPA) have set taste thresholds for the concentration of sodium in drinking water of 200 mg l^{-1} NaCl (about 78.6 mg l^{-1} Na) and between 30 and 60 mg l^{-1} Na, respectively[35,36]. An EPA drinking-water health advisory of 20 mg l^{-1} Na applies to individuals on sodium-restricted diets[34,36].

In this study, we explore a potential conflict between two important sustainability goals: (1) minimizing or reversing the FSS and (2) augmenting water supplies through the addition of highly treated wastewater to reservoirs and groundwaters, a practice referred to as 'indirect potable reuse' (IPR)[37]. While the number of IPR facilities is modest at present[38,39], the EPA recently released a draft national Water Reuse Action Plan[40,41] that promotes IPR and other forms of water reuse and recycling to address, where appropriate, expected water supply shortfalls over the next ten years in 40 of 50 US states[42]. More common is unplanned water reuse, which occurs, for example, when treated wastewater is discharged to surface waters upstream of a drinking-water intake[37]. Rice and Westerhoff[43] estimated that wastewater contributes >50% of the flow in 900 streams

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across the contiguous United States. Even in water-rich areas of the country, such as Indiana, unplanned water reuse constitutes a sizeable fraction of the water supply (3–134%, with the larger end of the range referring to circulation of wastewater through multiple water systems as it flows downstream)[44].

Human health and ecological concerns associated with IPR and unplanned water reuse typically focus on the impacts of discharged pathogens, nutrients, micropollutants and endocrine disruptors on receiving water quality[37,43,45,46]. These water reuse practices also have the potential to exacerbate the FSS. This is because salt entering a sewage collection system, or added during the treatment process, is not removed by conventional wastewater treatment processes. However, according to the literature, the contribution of treated wastewater to the FSS appears to be strongly context dependent. For example, in a study of salt retention in a rural watershed in New York State, "salt used for deicing accounted for 91% of the sodium chloride input to the watershed, while sewage and water softeners accounted for less than 10% of the input"[9]. By contrast, a study of sodium and chloride surface-water exports from the Dallas/Fort Worth region of Texas found that "the single largest contributor was wastewater effluent"[21]. A reasonable inference from these and other studies is that treated wastewater is a dominant source of freshwater salinity in warmer climates while deicers drive freshwater salinization in colder climates that receive snowfall[16,20,47]. This conclusion is supported by the strong south-to-north increasing trend in stream-specific conductance along the US east coast[14]. However, untreated wastewater drives the FSS across all climates, for example, as documented by the contribution of ageing sanitary infrastructure to stream chloride concentrations in Baltimore and Puerto Rico[48,49].

We hypothesize that two common methodological shortcomings in the literature may obscure the contribution of IPR and unplanned water reuse to the FSS in colder climates: (1) the focus is often on characterizing salt mass loads (salt mass per time) discharged from wastewater treatment plants, whereas many endpoints of human and ecological concern are concentration based (for example, EPA acute and chronic criteria for in-stream chloride concentrations[50] and the taste thresholds and health advisory for sodium concentrations in drinking water[34–36]); and (2) salt mass loads discharged from wastewater treatment plants are typically aggregated to monthly or longer period averages, thereby removing higher frequency processes (for example, day-to-day stream-flow variability) that can strongly influence the dilution of wastewater flows in inland freshwaters[37].

We test this hypothesis by analysing a >25-year time series of flow and sodium concentration measurements in the tributaries and highly treated wastewater (reclaimed water) that collectively drain to a regionally important drinking-water reservoir in Northern Virginia. Using regression and a copula-based conditional probability analysis[51] we demonstrate that, of the three sources evaluated here, reclaimed water dominates sodium mass loading to the reservoir during dry weather periods and has the highest sodium concentration year-round. To minimize the potential conflict raised earlier—between managing the FSS and augmenting water supplies through IPR—we suggest a set of locally tailored interventions that collectively increase a region's salt productivity, defined here as the goods and services produced per unit of salt discharged to inland freshwaters.

Field site

The Occoquan Reservoir, located approximately 30 km southwest of Washington DC in Northern Virginia, is one of two primary sources of water supply for nearly 2 million people in Fairfax County, Virginia, and surrounding communities (Fig. 1a). Sodium concentration in the reservoir began increasing around 1995 (purple curve in Fig. 1b) and now frequently exceeds the EPA's lower taste and health advisory thresholds (horizontal black solid and dashed lines). This trend prompted the local water purveyor, Fairfax Water, to explore planning-level options to address the rising sodium concentration in the reservoir, including the possible construction of a reverse osmosis treatment upgrade. The irony of desalinating freshwater and the estimated cost (US$1 billion, not including operating and maintenance costs and a vastly higher carbon footprint (S. Edgemon, personal communication) makes identifying, and ideally mitigating, sources of sodium in the reservoir a top regional priority.

On an annual basis, approximately 95% of the water flowing into the reservoir comes from its Occoquan River and Bull Run tributaries. Water from Bull Run includes baseflow and stormwater runoff from the Bull Run watershed ($1.94\times10^8\,\mathrm{m^3\,yr^{-1}}$) together with highly treated wastewater discharged from a water reclamation facility (Upper Occoquan Service Authority, UOSA) ($3.28\times10^7\,\mathrm{m^3\,yr^{-1}}$) located approximately 1.5 km upstream of Bull Run's confluence with the reservoir (red star in Fig. 1a). One of UOSA's missions is to improve drinking-water security in the region by augmenting stream flow into the Occoquan Reservoir with a high-quality and drought-proof source of water. Conceived and built in the 1970s, UOSA was the United States' first planned application of IPR for surface-water augmentation and a model for the design and construction of similar reclamation facilities around the world[37]. Water discharged from the Occoquan River comes primarily from baseflow and stormwater runoff from the Occoquan River watershed ($3.43\times10^8\,\mathrm{m^3\,yr^{-1}}$). Thus, possible sources of rising sodium concentration in the reservoir include deicer use and other land-based anthropogenic sodium sources in the rapidly urbanizing Occoquan River and Bull Run watersheds, which have experienced population increases of around 200,000 and 220,000 residents, respectively, over the past 20 years, and salt added to UOSA's sewershed from its >350,000 residential and commercial connections[52]. Possible sources of sodium within UOSA's sewershed include the down-drain disposal of sodium-containing drinking water and sodium-containing household products[53], use of water softeners in commercial and residential locations[10,20], and permitted and non-permitted sodium discharges from industrial and commercial customers. The sodium concentration in UOSA's effluent may also be elevated due to structural and non-structural water conservation measures that concentrate salts in wastewater[54]. Indeed, sodium concentration measured in daily flow-weighted composite samples of UOSA's reclaimed water are consistently higher than sodium concentrations measured in grab samples collected downstream on the Bull Run at station ST45 and on the Occoquan River at station ST10 (Fig. 1b).

Results

MLR models for sodium concentration. Multiple linear regression (MLR) models of sodium concentration generated for each monitoring station (ST10, ST45 and UOSA) were ranked by Bayesian information criterion (BIC) and then validated, depending on the length of the data record, using either leave-one-out cross validation (LOOCV) or the hold-out method (see Methods and Supplementary Information for details). The top-ranked MLR models (Supplementary Table 1) are significant ($P<0.001$) and capture between 31% and 87% of the measured variance in log-transformed sodium concentration (adjusted R^2 values and other model statistics are reported in Supplementary Table 1). The top-ranked MLR model for sodium concentration at ST45 captures the most variance ($R^2=87\%$, hold-out $R^2=81\%$), and its predictor variables include in situ specific conductance (positive correlation), maximum snow depth over the previous two weeks (positive correlation), log-transformed flow (negative correlation) and season (higher sodium concentration during the winter season). The top-ranked MLR model for sodium concentration in UOSA's reclaimed water captures the second-most variance ($R^2=54\%$, LOOCV-$R^2=51.6\%$) and has as its only predictor variable specific conductance measured on flow-weighted composite reclaimed water samples (positive

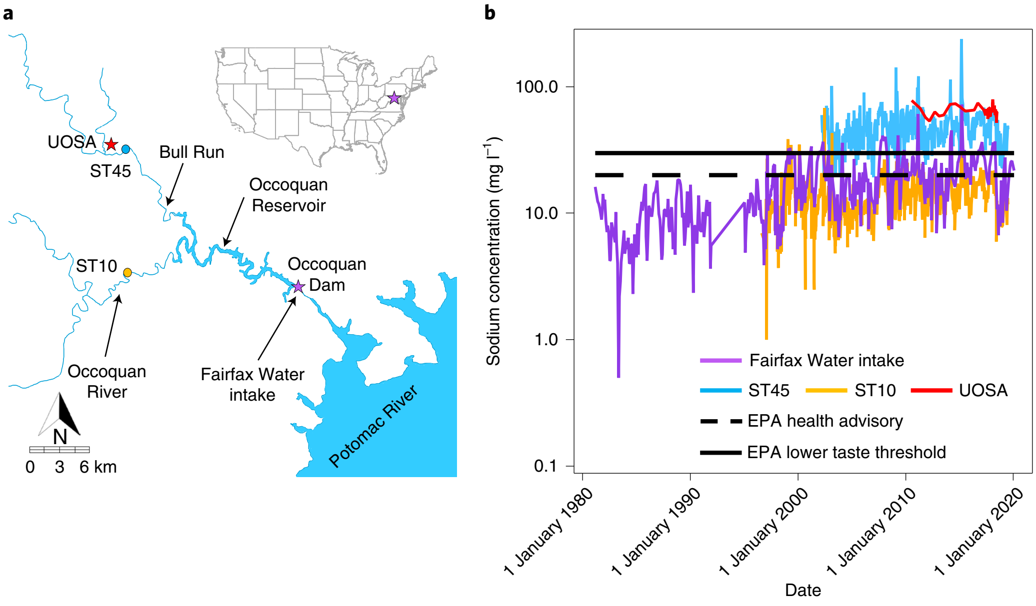

Fig. 1 | Both IPR and human activities in the Bull Run and Occoquan River watersheds contribute to salinization of the Occoquan Reservoir in Northern Virginia, USA. a, More than 95% of freshwater inflow to the reservoir is from the Occoquan River and Bull Run, which drain mixed undeveloped, agriculture, ex urban and urban landscapes. Shown are key geographical features, including the Occoquan Dam (where Fairfax Water sources its raw water), ion- and flow-monitoring sampling sites on the Occoquan River and Bull Run (monitoring stations ST10 and ST45), and the location on Bull Run where reclaimed water is discharged from the UOSA. Water from the Occoquan Reservoir is treated by Fairfax Water, the water wholesaler, and from there passes to various water distributors. **b**, Forty years of sodium concentration measurements at the Fairfax Water intake and upstream stations (ST10, ST45) and the final reclaimed water discharged by UOSA. Also shown are the EPA health advisory and lower taste threshold for sodium.

correlation). The top-ranked MLR model for sodium concentration at ST10 explains the least variance ($R^2 = 31\%$, hold-out $R^2 = 15\%$), presumably because in situ specific conductance measurements were not available at this station. Predictor variables for sodium concentration at ST10 include log-transformed flow (negative correlation), maximum snow depth over the previous two weeks (positive correlation) and number of days below freezing in the previous two weeks (positive correlation). In summary, sodium concentration at these three stations is (1) positively correlated with specific conductance measured either in situ (ST45) or on flow-weighted composites of the reclaimed water (UOSA); (2) positively correlated with environmental variables (antecedent snow, freezing weather and winter season) likely to be associated with deicer use (ST10 and ST45); and (3) negatively correlated with flow (ST10 and ST45), implying that stormwater tends to dilute in-stream sodium concentration.

Daily time series of sodium mass load and concentration. Synthetic time series of sodium concentration (generated using the top-ranked and validated MLR models described in the preceding) were combined with daily flow measurements at ST10, ST45 and UOSA to generate daily predictions (from 2010 through 2018) of sodium mass load and concentration in flows from the three putative sources evaluated in this study—Occoquan River watershed, Bull Run watershed and UOSA water reclamation facility (Methods). When these daily predictions are aggregated to annual averages, the results are in line with previous reports for regions that experience seasonal snowfall; namely, annual mass loading of sodium to the Occoquan Reservoir is dominated by the two watershed sources, not by UOSA (Fig. 2a). Consistent with Fig. 1b, however, the annualized sodium concentration in UOSA's reclaimed water ranges between 60 and 70 mg l^{-1}, well above EPA's lower threshold for taste (30 mg l^{-1}) and >1.5 and >4.5 times above the annualized sodium concentration in flow from the Bull Run and the Occoquan River watersheds, respectively (Fig. 2b).

These annualized results could be interpreted to imply that UOSA's reclaimed water contributes a relatively minor portion of sodium mass loading to the Occoquan Reservoir. However, the story is more nuanced when evaluated on a day-by-day basis (Fig. 3). During extended periods of reduced precipitation, sodium mass load from UOSA's reclaimed water frequently exceeds mass loads from either the Occoquan River or Bull Run watershed (see four vertical grey stripes, Fig. 3b). During wet weather, however, sodium mass loads from the two watersheds consistently exceed those from UOSA, often by >200-fold (note that the sodium mass load axis in Fig. 3b is logarithmic). Spikes in wet weather sodium mass loading from the two watersheds dominate the annual load estimates, giving the potentially misleading impression that UOSA's reclaimed water is a minor contributor to sodium in the reservoir (compare with Fig. 2a). These daily and annual sodium mass load estimates should be relatively robust to uncertainty in the MLR-generated synthetic sodium concentration time series because most of the variance in the daily mass load predictions ($R^2 = 66\%$, 91% and 82% for Occoquan River watershed, Bull Run watershed and UOSA, respectively) is attributable to measured daily average flow at each station.

Consistent with the annualized results (Fig. 2b), on a day-to-day basis the sodium concentration in UOSA's reclaimed water is nearly always higher than the sodium concentration in outflows from the Occoquan River and Bull Run watersheds (Fig. 3d). Sodium concentration in outflow from the Bull Run watershed is generally higher than in outflow from the Occoquan River watershed, consistent with the latter's greater impervious surface fraction (Supplementary Table 2).

Influence of weather on sodium mass loading. Application of a copula-based conditional probability analysis to daily predictions of sodium mass load for the period 2010–2018 (Methods) confirms that UOSA's reclaimed water dominates the sodium mass load entering the reservoir from the Occoquan River and Bull Run during dry and median weather conditions (Fig. 4). UOSA's percentage

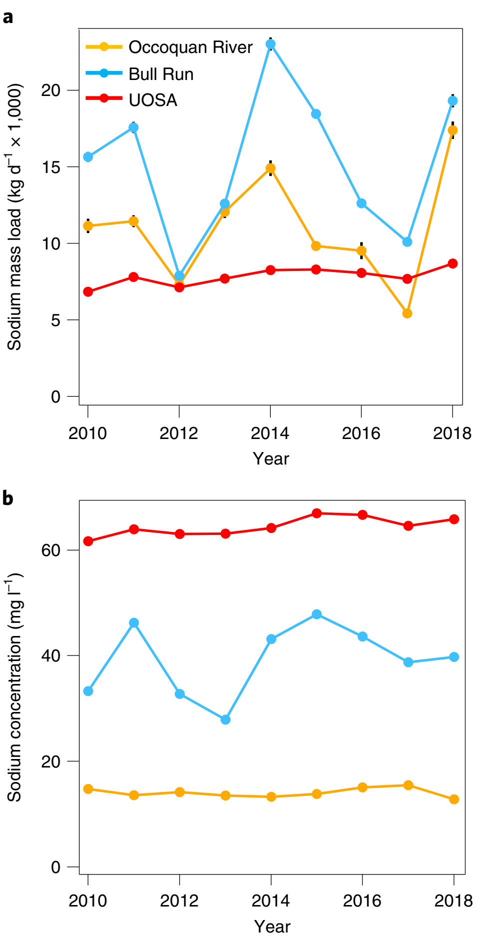

Fig. 2 | Annualized sodium load and concentration in outflow from the Occoquan River, Bull Run and UOSA water reclamation facility. a, Sodium mass loads. **b**, Sodium concentration. Error bars represent 95% prediction intervals (in some cases, too small to be visible).

contribution to sodium mass loading varies from 60% to 80% during dry conditions (corresponding to cumulative flow from the Occoquan River and Bull Run of $\langle Q_{\mathrm{Total}}\rangle = 2.55\,\mathrm{m^3\,s^{-1}}$), 30% to 50% during median conditions ($\langle Q_{\mathrm{Total}}\rangle = 6.91\,\mathrm{m^3\,s^{-1}}$) and 5% to 25% during wet conditions ($\langle Q_{\mathrm{Total}}\rangle = 31.0\,\mathrm{m^3\,s^{-1}}$). The Occoquan River and Bull Run watersheds exhibit the opposite pattern, contributing a greater percentage of the overall sodium load during wet weather periods. During wet weather, sodium mass loading from the Bull Run watershed is, on average, higher than sodium mass loading from the Occoquan River watershed, consistent with the land-use data in Supplementary Table 2.

Sources of wastewater salts. The results presented in the preceding support our hypothesis that, when evaluated on a day-to-day basis, discharge from water reclamation facilities can be an important component of the freshwater sodium budget even in colder climates, such as the Mid-Atlantic US, where deicers are a well-documented cause of inland freshwater salinization[7,8,14–19]. Where is the sodium in UOSA's reclaimed water coming from? UOSA water reclamation facility serves as a conduit through which sodium from myriad sources (watershed deicers, water treatment processes, household products, commercial and industrial discharges, drinking water treatment, and wastewater treatment) are focused into a single point source discharge (Fig. 5a). On the basis of data provided by the utility, we estimate that, on an annual average, 36% of the daily sodium mass load in UOSA's reclaimed water (7,600 ± 590 kg d^{-1}) is partitioned between chemicals used in water and wastewater treatment (for pH adjustment, chlorination, dechlorination and odour control), a single permitted discharge from a microfabrication facility and human excretion (human excretion was estimated by multiplying UOSA's service population (351,906)[52] by a mean per-person urine excretion rate of 3.608 g Na d^{-1} (ref. [55]) (Fig. 5b)). The source of the remaining 64% is unknown but presumably includes contributions from the down-drain disposal of sodium-containing drinking water (~2.5 mg l^{-1} Na) from Lake Manassas, the Potomac River and the Occoquan Reservoir, as well as sodium-containing household products that eventually end up in the sanitary sewer system.

Discussion

Given these results for the Occoquan Reservoir, how can the potential conflict between (1) minimizing freshwater salinization; and (2) promoting water security through IPR be addressed? One possible conceptual framework, borrowed from soft-path approaches for enhancing human water security[56,57], focuses on a variety of behavioural and technological interventions, applied at various scales, for increasing the goods and services produced per unit of salt discharged to inland freshwater; that is, improving salt productivity. As applied to sodium, we envision at least four ways in which salt productivity can be improved: (1) reduce watershed sources of sodium that enter the water supply (such as from deicer use); (2) enforce more-stringent pre-treatment requirements on industrial and commercial dischargers; (3) switch to low-sodium water and wastewater treatment methods; and (4) encourage households in the sewershed to adopt low-sodium products. These are considered in turn.

Because potable water supply and sewage collection systems are inextricably linked (Fig. 5a), factors that contribute salt to the former ultimately contribute salt to the latter as well. As mentioned earlier, many different sources (apart from treated wastewater) contribute salt to inland freshwaters, most notably deicer use in northern climates but also untreated sewage (such as from failing sanitary sewer systems[48]) and erosion of civil infrastructure (such as from concrete drainages[23]). With respect to deicers, their use on roadways can be curtailed without a reduction in public safety (for example, through the development of advanced pavement materials[58]). However, interventions at the watershed scale raise many questions across various domains, including human behaviour (how do we induce residents to be more conservative about their use of deicers on parking lots and driveways, and what is the 'right amount' of deicer they should be using?); hydrology (what are the hydrologic pathways by which salt moves through watersheds, and what are their timescales?); ecology (how do the changing concentrations and compositions of salinized waters alter biological communities and ecosystem processes?); and engineering design (are we unintentionally creating legacy salt pollution by adopting stormwater best management practices that transfer road salts to groundwater?). In such complex socio-hydro-ecological systems, well-intended interventions can have adverse consequences and so-called aggregation effects in which "desirable outcomes at a larger scale conceal inequalities and, as such, distributional injustices at the local scale"[59]. For example, deicer use might be reduced by lowering expectations for clean roads and public transportation during winter storms, but such actions could also limit access to free and subsidized school breakfast and lunch programmes for low-income children and thereby exacerbate child hunger[60].

Alternatively, more-stringent pre-treatment requirements can be imposed on commercial and industrial activities that discharge to the sewershed[45], although this will inevitably raise questions about potential economic trade-offs. For example, nearly 14% of the annual sodium load discharged by UOSA can be traced to a single chip fabrication facility (Fig. 5b). While imposing more-stringent sodium discharge limits on the facility would certainly reduce sodium loading

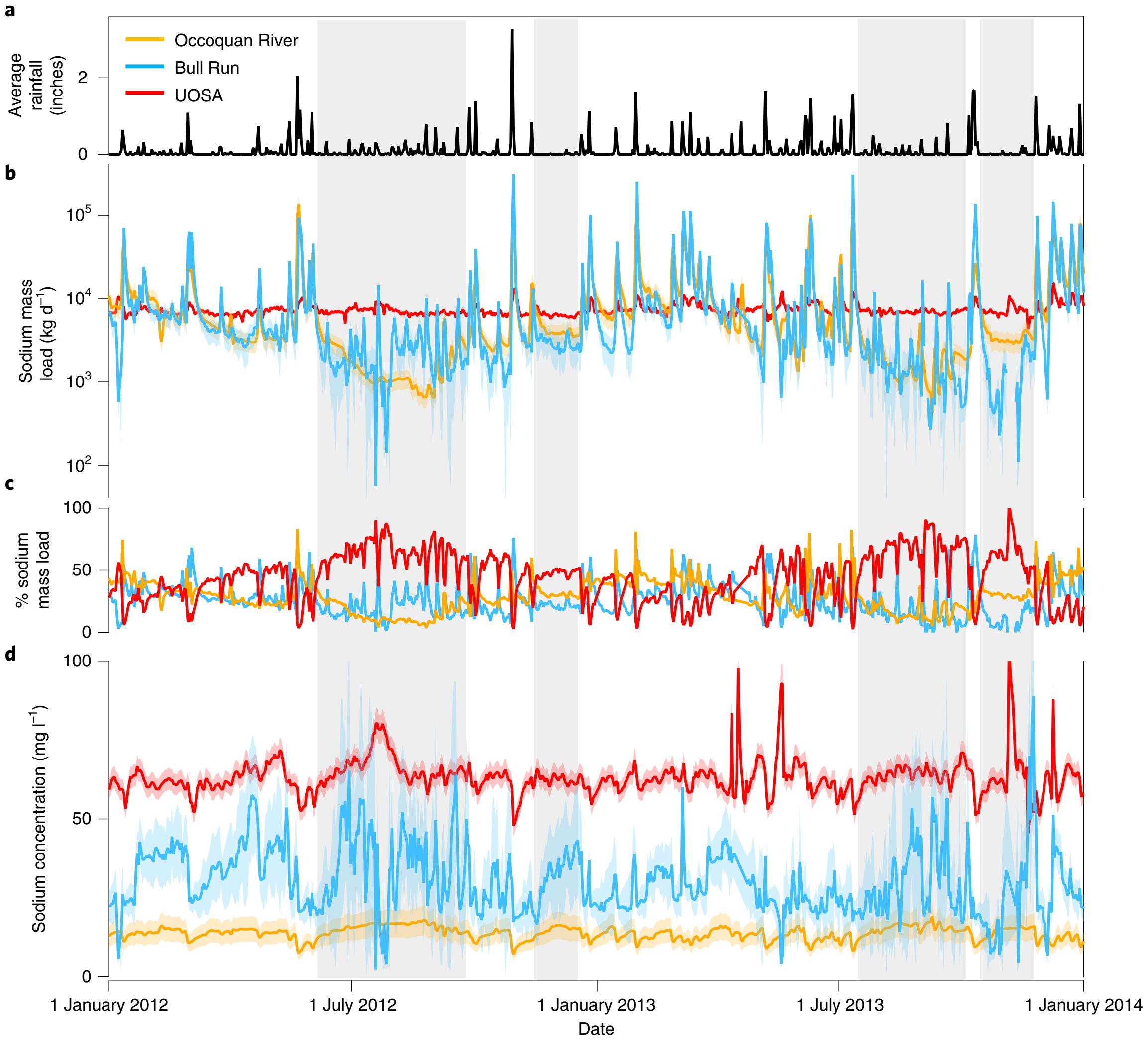

Fig. 3 | Daily sodium load and concentration in outflow from the Occoquan River, Bull Run and UOSA water reclamation facility for an illustrative two-year period (2012–2013). a, Daily average rainfall in the watershed calculated using the Thiessen polygon method. **b**, Daily sodium mass load entering the reservoir from each source (Occoquan River, Bull Run and UOSA). **c**, Percentage of daily sodium mass load entering the reservoir from each source. **d**, Daily sodium concentration in each source. Grey vertical stripes indicate extended periods of reduced precipitation. Coloured ribbons represent 95% prediction intervals.

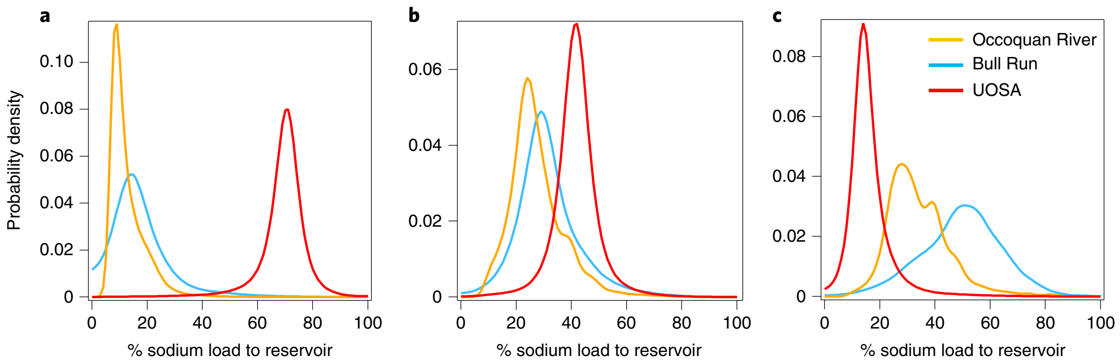

Fig. 4 | Probability density functions of the percentage sodium mass load entering the Occoquan Reservoir from the Occoquan River, Bull Run and UOSA conditioned on rate of flow into the reservoir. a, Low flow (2.55 m^3 s^{-1}). **b**, Medium flow (6.91 m^3 s^{-1}). **c**, High flow (31.0 m^3 s^{-1}). The salient feature of each curve is the range of values on the horizontal axis for which there is non-zero probability density. The peak height of each curve is determined by the unit area of each probability density function.

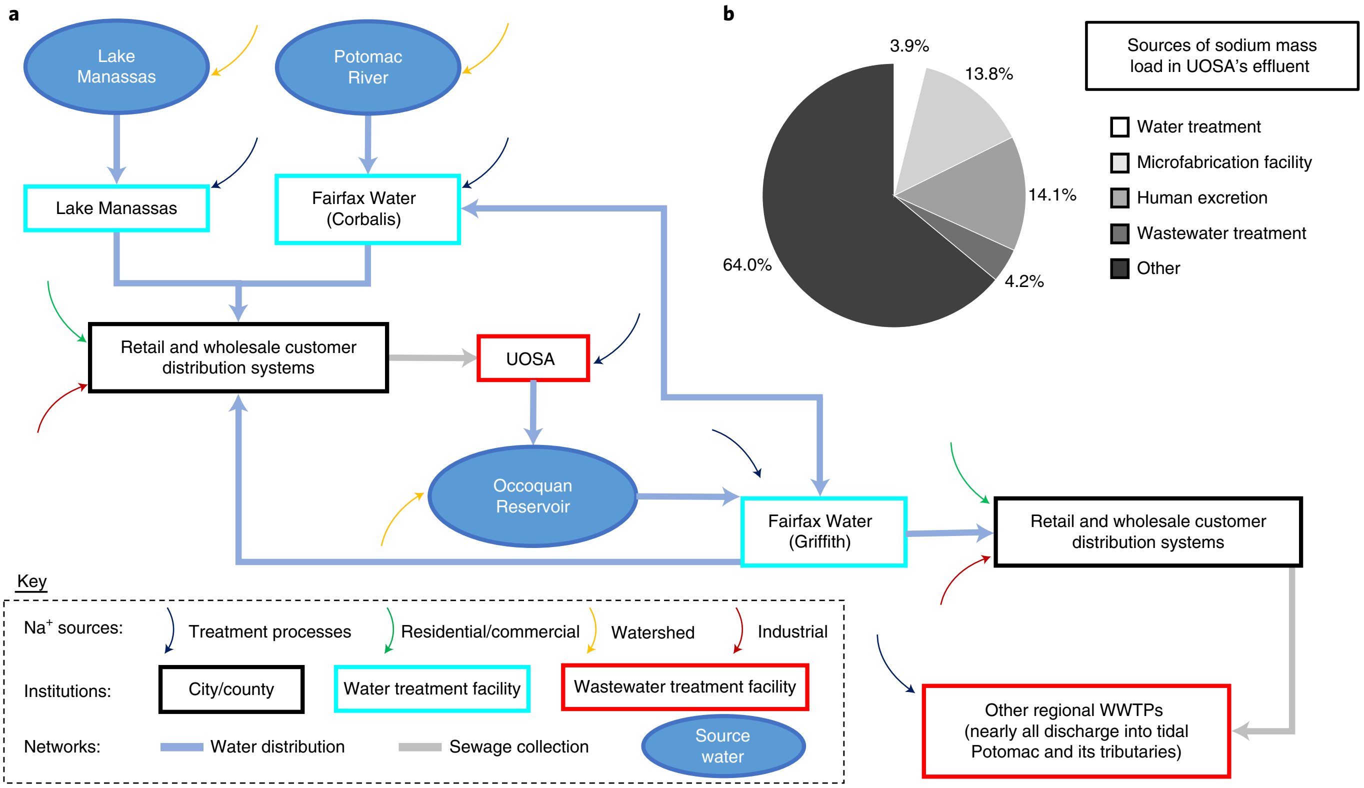

Fig. 5 | Sources of sodium discharged by the UOSA water reclamation facility. a, Schematic representation of the interdependent drinking-water distribution and sewage collection networks in the Occoquan watershed and surrounding area. Under normal conditions, the portion of the sewage network draining to UOSA receives water from the Fairfax Water Corbalis and the Lake Manassas water treatment plants, although some water from the Fairfax Water Griffith water treatment plant may also contribute to UOSA's inflow (forming a system-scale semi-closed loop for the circulation of sodium through the Occoquan Reservoir). **b**, Source breakdown for the annual sodium mass load in UOSA's reclaimed water. WWTP, wastewater treatment plant.

to the reservoir, it might also curtail plans to expand the facility and add up to 1,000 high-tech jobs to the local economy[61].

Changes in centralized water and wastewater treatment practices are also possible. Chlorine is a cost-effective and well-established method for destroying viruses, bacteria and protozoa, including those responsible for waterborne human disease[37]. Wastewater treatment plants that use chlorine for disinfection must also dechlorinate to prevent harm to downstream aquatic life. Dechlorination is typically achieved through the addition of sulfur dioxide or sulfite salts, including sodium sulfite, sodium bisulfite and sodium metabisulfite, thereby increasing the sodium content of the water[62]. Dechlorination dosages depend on the compound used; for example, sodium sulfite, sodium bisulfite and sodium metabisulfite require 1.8–2.0, 1.5–1.7 and 1.4–1.6 $\mathrm{mg\,l^{-1}}$ of chlorine residual, respectively[63]. Therefore, judicious choice of a dechlorinating agent or the use of alternative disinfectants (for example, ultraviolet light) can help reduce sodium mass loading from wastewater treatment. Interestingly, the use of ultraviolet light for disinfection might also reduce micropollutant concentrations in the reclaimed water[64].

Likewise, there are multiple steps in the drinking-water treatment process where sodium can be introduced. Drinking-water facilities should identify which of their processes contribute sodium and what alternative chemicals or processes might be adopted (see Supplementary Table 3), while being mindful of potential unintended consequences. As an example of a potential unintended consequence, adoption of the coagulant ferrous sulfate for drinking-water treatment, while potentially minimizing the addition of sodium, could accelerate the corrosion of downstream sewer infrastructure[65]. As with the chip fabrication facility example, economic constraints, as well as a risk-averse public service culture[66], may limit what can be achieved in practice.

Finally, improvements in salt productivity are possible at the household scale. Most research on household product ionic composition has been conducted in countries interested in greywater recycling as a water conservation strategy. For example, in 2008 a comprehensive study of sodium mass loads from household products in Melbourne, Australia, reported that[53] (1) laundry and dishwashing products contribute orders of magnitude more to sodium mass loads than do other household products; (2) median sodium mass loads from household products are 58–300% higher than those from human excretion; (3) mass loads of sodium can vary across product brands, which leads to high variability in the salinity of household sewage; and (4) product switching has the potential to reduce sodium mass loading to the sewershed. Assuming human excretion accounts for about 14% of the UOSA sodium mass loads (Fig. 5b), these Australian results suggest that household products could account for another 10–51%; notably, the upper limit would nearly close UOSA's annual sodium mass balance. Educational and social marketing campaigns aimed at informing consumers and manufacturers about the FSS, with the goal of fostering product and behavioural changes, could ultimately reduce salt loading from common household products such as detergents[67].

Methods

Historical monitoring data. To characterize the relative sodium contributions of the Bull Run watershed, the Occoquan River watershed and UOSA's reclaimed water to the Occoquan Reservoir, we utilized data from a long-term (>25 years) sampling programme that was originally established to monitor the effects of UOSA's water reclamation activities on water quality in the reservoir. We focused specifically on a 12-year period, 2006–2018, during which discrete surface-water samples were collected weekly or semi-weekly from the Occoquan River and the Bull Run monitoring stations (ST10 and ST45, $N=395$ and 338, yellow and blue circles, Fig. 1a) and analysed for a suite of water-quality parameters, including sodium concentration. Continuous measurements ($f=1\,\mathrm{hr}^{-1}$) of specific conductance ($N=106{,}708$ at ST45) and flow ($N=160{,}446$ and 170,179 at ST10 and ST45, respectively) were also available during this time frame. Daily average measurements of discharge from UOSA were provided by the utility for the period 2010–2018 ($N=2{,}941$), along with measurements of specific conductance ($N=2{,}943$) and sporadic measurements of sodium concentration ($N=68$) on daily flow-weighted composite samples of their reclaimed water.

Daily average time series of sodium concentration and mass loads. From the monitoring data described in the preceding, we set out to evaluate the relative contributions of three key sources—the Occoquan River watershed, the Bull Run watershed and UOSA—to sodium mass load (mass per time) and concentration (mass per volume) entering the Occoquan Reservoir under various weather and environmental conditions. Several limitations with the monitoring data had to be overcome (cf. ref. [44]): (1) flow and sodium concentration measurements at ST45 reflect the combined inputs from the Bull Run watershed and the UOSA water reclamation facility; (2) at ST10 and ST45, sodium concentrations were measured on grab samples, whereas sodium concentrations reported by UOSA were measured on daily flow-weighted composites of their final product; (3) the sampling schedules at ST10 and ST45 were asynchronous (grab samples were collected at different times on any given day, or on different days); and (4) while sodium measurements at ST10 and ST45 were collected every other week for the entirety of the study period (2010–2018), sodium measurements on UOSA's composite samples were sporadic and infrequent (Supplementary Fig. 1).

To address these challenges, for the period 2010–2018 (for which all of the required data resources were available), we constructed synthetic daily time series of average sodium mass load and concentration at the three monitoring locations as follows: (Step 1) at each monitoring station, an MLR model of log-transformed sodium concentration (dependent variable) was prepared (glmulti package[68] in R Statistical Software, R Core Team) by adopting, on the basis of stakeholder recommendations, the following set of potential environmental covariates (independent variables): (1) hourly stream flow (ST45 and ST10) or daily average reclaimed water discharged to Bull Run (UOSA), (2) maximum daily rainfall in the preceding two weeks, (3) maximum daily snow depth in the preceding two weeks, (4) number of days below freezing in the preceding two weeks, (5) season (as represented by sine and cosine functions with annual periodicity), and (6) either hourly in situ measurements of specific conductance (ST45) or measurements of specific conductance on daily flow-weighted composites of the reclaimed water (UOSA). For model validation we used the hold-out method at ST10 and ST45 and LOOCV at the UOSA station (see Supplementary Information for details); (Step 2) the populations of MLR models generated for each monitoring station in Step 1 were ranked according to BIC to identify the most parsimonious model, accounting for the trade-off between model fit and model complexity[69]. If the top-ranked models for a given station were within two BIC units, they were further ranked by LOOCV root mean squared error; (Step 3) the final top-ranked MLR model for each station from Step 2 was then used to generate an eight-year (2010–2018) synthetic time series of hourly (ST10 and ST45) or daily (UOSA) sodium concentration; and (Step 4) the synthetic sodium concentration time series from Step 3 were combined with hourly (ST10 and ST45) or daily (UOSA) flow measurements at each station and then aggregated to daily and annual sodium concentration and mass load using the aggregateSolute command in the USGS software package Loadflex (for error propagation we adopted the default data correlation structure, which assumes a unit correlation if two samples are collected on the same calendar date and zero correlation otherwise; cf. ref. [70]). The result was three fully aligned eight-year synthetic time series of daily and annual average sodium mass load and concentration (denoted here by the symbols $\langle L\rangle$ and $\langle C\rangle$, respectively) and associated prediction intervals at each of the three monitoring stations. As noted, ST45 receives water and sodium from both the Bull Run watershed and the UOSA water reclamation facility. The contribution of the Bull Run watershed to daily average sodium concentration and mass load was therefore isolated by mass balance where $\langle Q\rangle$ denotes daily average flow measurements and the subscript 'BR' refers to the Bull Run watershed:

$$\langle C_{\mathrm{BR}}\rangle = \frac{\langle L_{\mathrm{ST45}}\rangle - \langle L_{\mathrm{UOSA}}\rangle}{\langle Q_{\mathrm{ST45}}\rangle - \langle Q_{\mathrm{UOSA}}\rangle} \tag{1a}$$

$$\langle L_{\mathrm{BR}}\rangle = \langle L_{\mathrm{ST45}}\rangle - \langle L_{\mathrm{UOSA}}\rangle \tag{1b}$$

From these synthetic time series, we constructed daily time series for the percentage contribution of the Occoquan River watershed ('OccRiv'), Bull Run watershed ('BullRun'), and UOSA reclaimed water ('UOSA') to the total sodium mass entering the reservoir from the Occoquan River and Bull Run (which, as noted earlier, contributes 95% of freshwater flow into the reservoir):

$$\%\mathrm{Load}_{\mathrm{OccRiv}} = 100\frac{\langle L_{\mathrm{ST10}}\rangle}{\langle L_{\mathrm{ST10}}\rangle + \langle L_{\mathrm{ST45}}\rangle} \tag{2a}$$

$$\%\mathrm{Load}_{\mathrm{BullRun}} = 100\frac{\langle L_{\mathrm{BR}}\rangle}{\langle L_{\mathrm{ST10}}\rangle + \langle L_{\mathrm{ST45}}\rangle} \tag{2b}$$

$$\%\mathrm{Load}_{\mathrm{UOSA}} = 100\frac{\langle L_{\mathrm{UOSA}}\rangle}{\langle L_{\mathrm{ST10}}\rangle + \langle L_{\mathrm{ST45}}\rangle} \tag{2c}$$

Construction of bivariate distributions and conditional probabilities. Equations (2a)–(2c) provide daily predictions for the relative contribution of each source to sodium mass discharged to the reservoir from the Occoquan River and Bull Run. How are these predictions modulated by local weather conditions? To answer this question, we adopted the cumulative daily discharge of water flowing into the reservoir from the Occoquan River and Bull Run as a proxy of local weather conditions: $\langle Q_{\mathrm{Total}}\rangle = \langle Q_{\mathrm{ST10}}\rangle + \langle Q_{\mathrm{ST45}}\rangle$. Marginal probability distributions of percentage sodium mass load from Equations (2a)–(2c) ($\%\mathrm{Load}_{\mathrm{OccRiv}}$, $\%\mathrm{Load}_{\mathrm{BullRun}}$, $\%\mathrm{Load}_{\mathrm{UOSA}}$) and log-transformed values of cumulative stream flow from the Occoquan River and Bull Run ($\ln\langle Q_{\mathrm{Total}}\rangle$) were then joined by a copula to yield three bivariate cumulative distribution functions of the form $F_{LQ}(l,q) = C[F_l(l),F_Q(q)]$, where L and Q are random variables for the percentage sodium mass load from a particular source and cumulative discharge from the Occoquan River and Bull Run, respectively, l and q are specific values of these random variables, and C is the cumulative distribution function form of the copula function[71]. The copula was selected on the basis of BIC ranking of the Plackett and Archimedean copula families optimized to our daily time series of percentage mass load (from Equations (2a)–(2c)) and measured daily cumulative discharge from the Occoquan River and Bull Run using the MATLAB software package MvCAT[72]. The probability density function (PDF) of percentage sodium mass load from each of the three sources conditioned on a specific cumulative discharge was then calculated as follows:[71]

$$f_{L|Q}\,(l|q) = c\,[F_L\,(l)\,,F_Q\,(q)]\,f_L(l) \tag{3}$$

Here, the function c is the PDF form of the copula function and $f_L(l)$ is the PDF form of the marginal distribution for the percentage of sodium mass load to the Occoquan Reservoir from a particular source. We focused on three conditioning events corresponding to low (10th percentile), medium (50th percentile) and high (90th percentile) cumulative discharge ($\langle Q_{\mathrm{Total}}\rangle = 2.55\,\mathrm{m^3\,s^{-1}}$, $6.91\,\mathrm{m^3\,s^{-1}}$ and $31.0\,\mathrm{m^3\,s^{-1}}$, respectively). These three conditioning events represent dry, average and wet weather conditions, respectively.

Stationarity. The time-series data used for the copula analysis and to generate the MLR models were tested for stationarity (tseries package in R Statistical Software, R Core Team[73]) using the Augmented Dickey-Fuller test, the Phillips-Perron test and the Kwiatkowski-Phillips-Schmidt-Shin test (Supplementary Table 5 and Supplementary Note 1). These test statistics indicate that measured sodium concentration and all independent variables included in our analysis are stationary over the period for which the MLR and copula analyses were conducted (2010–2018).

Reporting Summary. Further information on research design is available in the Nature Research Reporting Summary linked to this article.

Data availability

All data used in this study are publicly available (https://doi.org/10.4211/hs.61a19724394643fca62a4fb3ce881efe).

Received: 16 August 2020; Accepted: 17 March 2021;
Published online: 19 April 2021

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Acknowledgements

Funding was provided by a US National Science Foundation Growing Convergence Research (GCR) Program award to S.B.G., M.A.R., S.K., P.V., M.E., T.A.B. and T.S. (NSF award #2021015) and by a Metropolitan Washington Council of Governments award to S.B.G. and S.K. (contract #21-001). The authors thank the Upper Occoquan Service Authority—in particular, B. Angelotti and M. Noble-Blair—for providing critical data and guidance, H. Zhang at The Water Research Foundation for a national perspective on freshwater salinization, D. Sedlak for valuable comments on the manuscript and participants in the Occoquan Watershed Monitoring Lab's Freshwater Salinization Workshop held on 14 January 2020.

Author contributions

S.V.B. and S.B.G. conceived and drafted the article. E.A.P., M.A.R., A.N.G., P.V., A.M.-M., M.E., G.P., N.S. and S.C. contributed text and analysis. All authors contributed edits.

Competing interests

The authors declare no competing interests.

Additional information

Supplementary information The online version contains supplementary material available at https://doi.org/10.1038/s41893-021-00713-7.

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Peer review information *Nature Sustainability* thanks the anonymous reviewers for their contribution to the peer review of this work.

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Last updated by author(s): Mar 11, 2021

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Our web collection on statistics for biologists contains articles on many of the points above.

Software and code

Policy information about availability of computer code

Data collection	No software code was used for data collection.
Data analysis	The following softwares and software packages were used for data analysis: R software (RStudio version 1.2.5001), USGS-R/loadflex (version 1.1.11), MvCAT (version 2.02), Mathematica (version 12.0.0.0).

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Field-specific reporting

Ecological, evolutionary & environmental sciences study design

All studies must disclose on these points even when the disclosure is negative.

Study description	In this study we quantify the contribution of three salinity sources (one wastewater reclamation facility and two rapidly urbanizing watersheds) to rising concentration of sodium in a regionally important drinking water reservoir using historical data of sodium concentration, specific conductance and other environmental variables like discharge, rainfall, snowfall, temperature.
Research sample	This study involves the use of existing datasets for (a) sodium concentration, specific conductance, discharge and watershed rainfall obtained from the Occoquan Watershed Monitoring Laboratory, Manassas VA; (b) snow depth and minimum air temperature obtained from National Oceanic and Atmospheric Administration.
Sampling strategy	The sample size for each parameter varied by location depending on the extent of historical data available at that location. The final analysis included data from 2010-2018, spanning multiple years and all seasons.
Data collection	Used existing historical datasets, collected by different entities.
Timing and spatial scale	The timescales for sodium concentration and specific conductance measurements varied by site location. At ST45, sodium concentration data is available from 12/04/06 to 08/20/18, and specific conductance data from 11/05/06 to 12/31/18. At ST10, sodium concentration data is available from 01/05/06 to 11/28/18. At UOSA, sodium concentration data is available from 07/13/10 to 06/26/18, and specific conductance data from 06/10/10 to 06/30/18.
Data exclusions	No data were excluded for the MLR analysis. Bivariate distribution analysis involved data only for the time period when data from all three site locations overlapped.
Reproducibility	N/A
Randomization	N/A. Not relevant to this study as no clinical research or randomized control trials involved.
Blinding	N/A. Not relevant to this study as no clinical research or randomized control trials involved.

Did the study involve field work? ☐ Yes ☒ No

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Methods

n/a	Involved in the study
☒	☐ ChIP-seq
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☒	☐ MRI-based neuroimaging