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# The Anthropogenic Salt Cycle

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#### Abstract

Increasing salt production and use is shifting the natural balances of salt ions across Earth systems, causing interrelated effects across biophysical systems collectively known as freshwater salinization syndrome. In this Review, we conceptualize the natural salt cycle and synthesize increasing global trends of salt production and riverine salt concentrations and fluxes. The natural salt cycle is primarily driven by relatively slow geologic and hydrologic processes that bring different salts to the surface of the Earth. Anthropogenic activities have accelerated the processes, timescales and magnitudes of salt fluxes and altered their directionality, creating an anthropogenic salt cycle. Global salt production has increased rapidly over the past century for different salts, with approximately 300 Mt of NaCl produced per year. A salt budget for the USA suggests that salt fluxes in rivers can be within similar orders of magnitude as anthropogenic salt fluxes, and there can be substantial accumulation of salt in watersheds. Excess salt propagates along the anthropogenic salt cycle, causing freshwater salinization syndrome to extend beyond freshwater supplies and affect food and energy production, air quality, human health and infrastructure. There is a need to identify environmental limits and thresholds for salt ions and reduce salinization before planetary boundaries are exceeded, causing serious or irreversible damage across Earth systems.

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#### Introduction

Salt, which makes up the major ions dissolved in the ground and surface waters of the Earth, is essential for regulating a variety of biological, geological and chemical processes across natural and engineered systems. However, human activities have dramatically altered the amounts and compositions of different salt ions in terrestrial and aquatic environments. In the USA, well over 36 Mt of rock salt (NaCl) has been mined per year<sup>1</sup>. Approximately 20 Mt of NaCl is currently used for road salt in the USA<sup>2,3</sup> (Box 1). On a global scale, approximately 39.55 Mt of K+- rich potash is extracted annually for use as agricultural fertilizer<sup>4</sup>. At present rates of consumption without changes in lifestyles or management, an average newborn infant will require a supply of 13.6 t of salt throughout their lifetime<sup>5</sup>.

Starting in the twentieth century, an increase in anthropogenic salt inputs exceeding biological, geochemical and engineering demands has driven long-term increases in the pools, residence times and fluxes of different salt ions<sup>6,7</sup>. Salinization of water, soil and air has increased because of irrigation<sup>8</sup>, the application of road deicers and fertilizers, wastewater discharges, mining, resource extraction, human-accelerated weathering<sup>7</sup> and saltwater intrusion<sup>9</sup>. Salinization has the potential to extract and co-mobilize many other contaminants from soils and sediments, including nutrients, metals, radionuclides and organics<sup>10</sup>. These so-called chemical cocktails of dissolved salts and other contaminants can have shared sources, flowpaths or biogeochemical reaction pathways<sup>9–11</sup>, which are influenced by factors such as climate, geology, land use, human activities and time<sup>7</sup>. The suites of biological, physical and chemical impacts that commonly occur together are known as freshwater salinization syndrome (FSS)<sup>6–10,12</sup>.

Inland waters are a key conduit for salts, but the magnitude and extent of FSS are now expanding beyond only a freshwater issue. Dissolved salt ions and evaporites impact land, air and infrastructure through soil salinization, erosion rates, decreased crop growth; the mobilization of dust and air pollution<sup>7</sup>; impacting pathogen survival<sup>13</sup>; the corrosion and scaling of metal pipes; changing potability and taste of drinking water; and the mobilization of metals in soils and groundwater including radionuclides<sup>8</sup>. Across land, air and water, there are myriad human health impacts associated with inhaling, drinking and consuming excess salt and its chemical cocktails such as hypertensive disorders, cancers and respiratory diseases<sup>8</sup>. Therefore, there is a need to understand the sources, fate and transport of salt across land, air and water as a holistic biogeochemical cycle that accounts for natural and anthropogenic processes.

In this Review, we conceptualize the natural salt cycle and its major processes. We compare how the natural salt cycle has been disturbed by human activities and identify major sources, transport and transformations of salt ions along the anthropogenic salt cycle from watershed to global scales. We examine how salt concentrations and fluxes are exceeding biological demands (or limits within which ecological systems can handle salt ions) owing to increasing societal demands for salt; this occurrence is illustrated by a national salt budget for the USA and by examining global effects across the hydrosphere, pedosphere, lithosphere, biosphere and atmosphere. Finally, we discuss emerging research directions and impacts.

#### The Natural Salt Cycle

Salt cycle occurs naturally on land in the lithosphere, pedosphere and biosphere. However, the global salt cycle remains poorly conceptualized; instead, diverse disciplines (geology<sup>14,15</sup>, soil science, ecology, hydrology and oceanography) have considered how specific salt ions (mostly NaCl) impact particular components of the Earth such as its crust, soils, biota, freshwaters or oceans. For example, soil salinization is often considered independently of freshwater salinization, despite an obvious relationship between the two occurrences. Similarly, natural patterns of concentrations and fluxes of major ions in some of the major rivers of the world have been characterized<sup>16</sup>, but interactions and transport across soils, the atmosphere and deeper groundwater are not detailed. However, a baseline set of natural salt-related processes (referred to here as the natural salt cycle) must be established to understand anthropogenic disturbances (Fig. 1).

#### Salts on Earth

Salts are ionic compounds with cations and anions such that the product is neutral. Salts can be basic, acidic or neutral and each salt can have different solubility in water and subsequent ability to conduct electricity. The component ions can be inorganic (such as Cl–) or organic (such as CH3COO–), monoatomic (such as F–) or polyatomic (such as NO3–). Different salt ions have different affinities to form complexes with metals such as Cu and Pb, which have environmental and human health implications owing to toxicity. Abundant major salt ions such as Ca2+, Mg2+, K+, SO42– and dissolved inorganic carbon (DIC) are found in the ground and surface waters of the Earth. Here, the term salt is used to represent all of these major ions (concentrations typically >1 mg l–1 in Earth's water).

#### Geologic and terrestrial processes

Salts are released to the surface of Earth by the weathering of sedimentary rocks (including evaporites), igneous rocks and metamorphic rocks that are exposed at the surface of the Earth (Fig. 1). Evaporitescan be dissolved when exposed to water, including groundwater<sup>17–22</sup>, releasing salt ions. Additionally, some rock units include salt layers, which often uplift in unique ways to form diapirs or salt domes<sup>23</sup>. Salt diapirs are mobile masses that intrude into overlying harder rocks owing to the buoyancy of salt relative to other sediments and rocks.

Physical, chemical and biological weathering and erosion of rocks release salt ions to surface waters, soils and air. Salts in soils primarily come from the weathering of rocks and minerals, including carbonates and feldspars<sup>24,25</sup>. Plants<sup>26,27</sup>, fungi<sup>28</sup> and other organisms take up salt ions, depending on their nutrient needs and salt stress<sup>26–32</sup>. Evaporation and plant transpiration pull soil water in the rooting zone, and water and salt ions move toward the surface via capillary action. Salts in soils accumulate once the water at the surface is evaporated to the atmosphere<sup>24,25,33,34</sup>. In arid environments, this can lead to the formation of caliche or soil layers of calcium carbonates<sup>33,35</sup>. Salt ions also accumulate in biomass and certain elemental combinations accumulate in soils through biogeochemical transformations (such as chloride interactions with organic matter forming organochlorines in forest soils).

Salt ions are released from soils via ion exchange and dissolution. Once dissolved in soil water or groundwater, salt ions can be transported via diffusion, advection and dispersion and enriched during freeze-thaw cycles (including brine rejection, which can occur when salty water freezes and salt ions do not fit within the crystal structure of ice and are expelled into surrounding water). From the soil, salt ions can enter aquatic systems or the atmosphere, where they undergo additional transport and physicochemical processes.

Salt ions are eventually precipitated, buried and lithified as sedimentary rocks and/or percolated to deeper groundwater or ocean subduction zones, in which their elemental constituents can undergo igneous and metamorphic reactions to form minerals in rocks. Salts can also be reincorporated into the lithosphere through the formation of evaporites through desiccation.

#### Processes in water

The natural salt cycle of the Earth is intimately connected to sources, fluxes and storage of water. Natural weathering fluxes of salts from land to water depend on climate (temperature and precipitation), rock type or geology, location or flowpath, biologic factors and time<sup>7,36,37</sup>. The hydrological cycle is a major transporter of salt ions, and one of the largest salt fluxes is from rivers to oceans<sup>16,38</sup>, with additional transport in soil water and groundwater<sup>39,40</sup> (Table 1). For example, connate fluid, salty water that is trapped within rock during formation, can migrate and naturally lead to salinized groundwater<sup>41–45</sup>. There are also natural salt fluxes from marine environments to freshwaters, owing to tidal fluctuations, long-term climate change or marine transgression<sup>24,25,46</sup>, incidental flooding<sup>45</sup> and salt aerosol inputs through rain<sup>47</sup>.

In marine environments, salt cycling is driven by evaporation, biogeochemical transformations, ice formation (resulting in brines) and saltwater intrusion. Reverse weathering controls major ion ratios and alkalinity in seawater through the deposition of silicate minerals involving divalent cations, primarily forming clays<sup>48–50</sup>. Salt is precipitated from seawater, whereby carbonate deposits first at low salinities (most modern calcium carbonate is formed through biological processes<sup>51</sup>), then gypsum, and finally sodium chloride deposits last at high salinities<sup>23,52</sup>. In locations of high heat flow, such as rift valleys and subduction zones, the water in seawater is vaporized leaving salt ions, increasing brine salinity until salt precipitates<sup>45,53,54</sup>.

#### Processes in the atmosphere

Salt cycle also occurs naturally within the atmosphere of the Earth. Sea spray is also a primary source of aerosolized atmospheric salts<sup>55</sup> therefore, most deposition occurs over oceans or in coastal areas<sup>55</sup>. Dry deposition typically exceeds wet deposition, and fluxes depend on wind speed, size of source area, proximity to source area, residence time and concentration of salts in the atmosphere, and contributions of wet and dry deposition<sup>55–58</sup>. Natural salt in dust, including Na+ and Ca2+, comes from arid soils, dried lake beds, and salt flats<sup>33,59–61</sup>. Atmospheric deposition of salt ions can accelerate melting of snowpacks on nearby mountains, as is occurring in arid regions, which impacts regional

water supplies<sup>62</sup>. Dust deposition rates vary with climate conditions and can range from 0.004 to 0.34 g m-2 year-1 during glacial cycles, which is 2 to 20 times higher than during interglacial cycles<sup>57,63</sup>.

#### Disturbing the natural salt cycle

The natural salt cycle has been disturbed by the extraction of salt for anthropogenic uses, such as agriculture, road deicers, food preparation, industry, construction, and water and wastewater treatment. For example, salts are used heavily in the built environment — aggregate minerals in construction materials contain salt ions (CaSO42– and CaCO3). There are salt compounds and organic salts (including monosodium glutamate) in food additives, cosmetics, cleaning supplies, solvents and printing, which can contribute salt ions (including Na+, K+, Ca2+ and Mg2+) to the environment. There are also hydrated salts in household products (including bleaching powder (Ca(ClO)2), anti-caking agents (potassium ferrocyanide (K4[Fe(CN)6])(•xH2O)), reagents in manufacturing, borax (Na2[B4O5(OH)4] ·8H2O) and washing soda (Na2CO3•10H2O)), but the major ions of Na+, Ca2+, Mg2+, K+, Cl–, HCO3 – and SO42– are the primary focus here<sup>64,65</sup>.

Global production and distribution of NaCl and many other salt ions such as K+, SO42–, Ca2+, Mg2+ and carbonates are rapidly increasing. There has been a corresponding increase in calcium carbonate, gypsum and halite production on a global scale (Fig. 2a). In the USA, for instance, overall salt consumption is now greater than salt production, which demonstrates the importance and reliance on imports (Fig. 2b). Salt production in China overtook US production in 2015; the 2013–2017 average production (68.7 Gt) is 1.6 times greater than US production (42.3 Gt)66. Increasing salt use is a global issue and is probably because of various causes such as increased impervious surfaces (roadways and parking lots), increasing needs for road safety and traffic in colder climates, changes in human diet, agriculture and industry.

#### The anthropogenic salt cycle

Human disturbances of baseline processes drive an anthropogenic salt cycle (Fig. 1). Earth processes that influence the salt cycle naturally occur over long geologic periods (typically centuries to millions of years) but humans have dramatically accelerated this cycle by altering the forms, fluxes and fates of salts in the global cycle (Fig. 1). Land use change, agricultural practices, mining and urbanization have accelerated extraction, weathering and transport of salts from the lithosphere and pedosphere, diverting vast quantities of previously stored or geologically locked-up salts into the natural cycles of the Earth (Fig. 1). Furthermore, human-induced changes in the hydrological cycle and processes that increase salinization of freshwater are interacting to increase salt ion concentrations. As a result, there are rapidly increasing trends in salt ion concentrations in rivers over the past century<sup>8,10,67</sup>. In this section, salt cycling under current anthropogenic influences is described and conceptualized.

#### Natural waters

Human activities have altered salt fluxes in freshwater ecosystems. Based on historical data from world river inputs to oceans, global fluxes from the major rivers of the world in 1970 were estimated as 4.04 Ca2+, 1.01 Mg2+, 1.88 Na+, 0.60 K+, 2.0 Cl-, 2.87 SO42– and 16.54 HCO3–(in g m–2 year–1)<sup>16</sup> (Table 1). The estimated anthropogenic salt inputs to the ocean were 47 Ca2+, 10.5 Mg2+, 78 Na+, 5 K+, 93 Cl–, 124 SO42– and 100 HCO3– (in 106 t year–1)<sup>16</sup>. Over the past 50 years, all of these ions have increased in streams and rivers on continental and global scales<sup>8,10,67,68</sup> (Fig. 3), coinciding with a global increase in the consumption and production of salts<sup>10</sup>. For example, there are widespread increasing trends in electrical conductivity, chloride, sodium and other ions in freshwaters in North America<sup>9,69,70</sup>. There have been increasing trends in DIC concentrations and fluxes in some rivers<sup>68,71,72</sup>. DIC (primarily HCO3–) fluxes range from 55 × 109 to 2,450 × 109 mol year–1 in the 25 largest rivers in the world<sup>73</sup>, and there are perturbations of DIC transport from land to sea<sup>74</sup>. Rising salt concentrations suggest that the capacity of watersheds for diluting and attenuating salt ions to steady-state conditions, or salt assimilative capacity, can be exceeded<sup>6</sup> (Fig. 4).

Increasing salt trends in freshwaters are caused by diverse drivers such as human-accelerated weathering<sup>75</sup>, salt pollution from nonpoint and point sources, resource extraction<sup>76</sup> and groundwater pumping. Salt is concentrated in runoff enriched with road salts, agricultural fertilizers and lime, mine drainage, and ions from weathering of impervious surfaces. Salt ions in road runoff can be transported both rapidly to streams through conduits, subsurface piping and preferential flowpaths and more slowly through soil water and groundwater as salt plumes<sup>77,78</sup>. DIC can be increased from agricultural liming, urban impervious surfaces, changes in atmospheric deposition and human-accelerated weathering<sup>68,79,80</sup>. Salt is also added to groundwater as a result of pumping, injection of industrial and wastewater discharges, brines from fracking and gas and oil production<sup>81,82</sup>, accumulation of road salts and fertilizers in groundwater, and saltwater intrusion in coastal regions<sup>83</sup>.

Variations in salinization and in relationships between Na+ and Cl– reflect variations in the sources, transport and transformation of salt ions across climate, geology, human activities, flowpaths and time<sup>7</sup> (Fig. 4). For example, rivers draining semiarid and arid regions in South Africa can show enrichment in Na+ concentrations relative to Cl– concentrations whereas rivers draining humid regions can show enrichment in Cl– concentrations relative to Na+ owing to road salt and other pollution sources (Fig. 4). Retention of Na+ and Cl– in groundwater can occur because of incomplete flushing of soils in the vadose zone, uptake by microbes and plants and ion exchange of Na+ on soil exchange sites<sup>84,85</sup>. Overall, the attenuation effects of soil ion exchange, biological uptake and dilution effects of downstream water accumulation and/or precipitation events no longer drive salt concentrations below water quality limits.

The anthropogenic salt cycle can also be altered in marine waters through evaporative concentration from warming sea surface temperatures, sea level rise and saltwater intrusion. Saltwater intrusion impacts at least 501 cities worldwide, 108 of which support populations of more than 1 million<sup>86</sup>. In the continental USA alone, coastal aquifers supply potable

water to 95 million people, and many are considered vulnerable to seawater intrusion because they lie below sea level; 15% of West Coast, 22.6% of Gulf Coast and 34.7% of East Coast groundwater well elevations within 10 km of the coast are below sea level<sup>87</sup>. At present, landward hydraulic gradients are evident along at least 15% of the US coastline, suggesting that a large fraction of the coastal USA may be vulnerable to freshwater salinization<sup>87</sup>. Only 2–3% seawater is needed to make a coastal groundwater aquifer unsuitable for drinking<sup>87,88</sup>.

#### Engineered water systems

Although often overlooked, the engineered water cycle (including wastewaters) contributes to acceleration of the anthropogenic salt cycle. Salts originating from seawater and rocks are taken up by humans through consumption of food and beverages and released into domestic wastewater. An average person in the USA can consume 3,400-4,000 mg day-1 of Na+ and excrete 3,414-3,803 mg day-1 of Na+ (refs. 89,90). Wastewater loads from selected studies around the world show that households can discharge 90,629 mg of Na+ per person per week and 18,713 mg of K+ per person per week, and 38,768 mg of Cl- per week and 11,236 mg of Ca2+ per week can be released into wastewater from an Australian household<sup>91</sup> (Table 2).

Home products are a major contributor to salt loads, especially in effluent-dominated streams, which are a major and increasing fraction of the streams in the USA, for example. Chemical analysis of home products ranging from toothpaste to dishwashing soap suggests that salt ions such as Cl– are prevalent across products and are especially high in soaps, deodorants, in-cistern toilet fresheners and powdered laundry detergents (mean concentrations of 89, 67, 53 and 27 g kg–1 product, respectively12,64,65,91. Of these products, powdered detergents are expected to contribute most to overall salt mass loading, with estimates of Cl– loading ranging from 21 g person–1 week–1 to 86 g person–1 week–1 (ref. 64). Individual in-home products (and the wastewater streams they contribute to) contain unique chemical cocktails that are part of the anthropogenic salt cycle. For instance, although laundry and dishwasher powder have elevated concentrations of fluoride, chloride and, in some countries, phosphorus92 and low concentrations of cobalt, antimony and selenium, shower and bath products (such as shampoos and conditioners) are the opposite. Therefore, wastewater salt profiles, and their contribution to the anthropogenic salt cycle, are a signature of the products that societies use<sup>64</sup>.

Engineered systems also contribute to salinization through the addition of salts used in the treatment of drinking water and wastewater. As the population of the world continues to increase, water management, reuse and treatment will all increase. Adoption of treatment upgrades, such as reverse osmosis systems, to remove salts is one possible solution, but also raises concerns, including the energy and carbon footprint and the potential environmental impacts of brine disposal<sup>6,12</sup>. Salt ions are added at almost every step along engineered water systems, and thus, the ways water is used, reused and recycled for urban and agricultural uses can impact inland freshwater salinization.

#### Land

Although salts naturally occur in soils, approximately 1 billion ha of soils are impacted by anthropogenic salinization, primarily owing to irrigation, fertilization and liming<sup>93</sup> (Fig. 3a). Between 1970 and 1980, approximately 6.5% of the soils of the world capable of growing crops were either saline or sodic<sup>94</sup>, and over 50% of the croplands of the world could be impacted by salinization by the end of the twenty-first century<sup>95,96</sup>. These estimates have some inaccuracies — remote sensing-based techniques probably overestimate the spatial extent of soil salinization in Mexico and underestimate impacts in Australia<sup>93</sup> (Fig. 3a) — but the spatial extent of soil salinization at the global scale is unequivocally growing<sup>93,96</sup>.

The anthropogenic salt cycle on land is enhanced through increased evaporation rates<sup>97</sup>, vegetation disturbance, anthropogenic inputs of salt ions and human-accelerated weathering. Evaporation of saline lakes causes precipitation of salts such as mirabilite, halite, thenardite and gypsum<sup>98</sup>. Vegetation disturbance, deforestation and colonization by shallow-rooted plants can influence evaporation rates and concentrate salt ions in upper soil horizons through capillary action.

Anthropogenic inputs include potash, which is enriched in K+ and mined or concentrated through evaporation of lakes and subsurface brines<sup>99</sup>. In agricultural watersheds with rising chloride trends, chloride from potash fertilizers can be the dominant input (approximately 49 kg ha–1 year–1) and even exceed road salt sources (7.2–23.2 kg ha–1 year–1)100. Cation exchange dictates how salts are stored and transported in soils and migrate vertically in the soil profile owing to leaching and capillary action. There can be substantial accumulation of anthropogenic salts in soils owing to retention of Cl– because of incomplete water flushing, uptake by microbes and plants, chlorination of organic matter, and ion exchange of Na+ at soil exchange sites<sup>84,85</sup>.

The built environment impacts the salt cycle through weathering and the mining of materials for construction (including sand and gravel extracted from open pits and quarries; bedrock that is drilled, blasted, and mined to produce crushed stone; and limestone and dolomite produced from underground mines)<sup>101</sup>. A key example is the widespread extraction of salt ions to make concrete, dry wall and other construction materials. The rates of mineral weathering of this concrete and 'urban karst'<sup>75</sup> can be faster than those of other types of sedimentary rocks accelerating the release of salt ions. Eventually, chemical and physical weathering of rock aggregates in the built environment degrade surfaces and cause dissolution, honeycomb weathering, weathered rims and rinds, spalling and salt corrosions on stone<sup>102</sup> contributing to salt ion pollution to surface waters<sup>75</sup>.

#### Atmosphere

Human activities have altered sources and fluxes of salt in the atmosphere. In arid regions, the drying of the Aral Sea and other drying lake beds around the world (owing to diversions, irrigations, climate change and other factors) have increased mobilization of saline dust and particulate pollutants across vast regions<sup>103,104</sup>. Desertification and increased dry lake beds can have total suspended particulate concentrations of approximately 230–290  $\mu$ g m–3 of

air, with soluble salts comprising 3–20% of the total suspended particulates<sup>104</sup>. Drought and catchment scale disturbances resulting from human activities such as farming, water use and urban development alter atmospheric fluxes of salts<sup>105,106</sup>. For example, salts can be added to atmospheric deposition through dust from erosion of rangelands<sup>105</sup> and desertification in other locations worldwide<sup>103</sup>. In the Western USA, dust loads have increased by 500% compared with the late Holocene owing to livestock grazing<sup>105</sup>.

In colder regions, aerosolization of road salts (Na+ and Cl–) in PM 2.5 particles can increase during winter months<sup>107</sup>. Road salt can be an important ClNO2 source and influence atmospheric composition and air quality in the urban wintertime environment<sup>108</sup>. There have been long-term increasing trends in atmospheric deposition of Na+ and Cl– in some areas of the Northeastern USA probably owing to road salting<sup>8</sup>. Industrial activities, coal and biomass burning, traffic emissions, wear of asphalt, agricultural operations and wildfires can also increase mineral dust containing salt ions (for example, concentrations within urban dust are 2.4% Ca2+, 1.5% K+, 1.2% Na+ and 0.9% Mg2+)<sup>109</sup>.

#### **Cascading Impacts**

Salinization has cascading impacts, which have already been documented in groundwater systems<sup>110</sup> and ecological food webs<sup>111–113</sup>. These impacts arise from the mobilization of metals, nutrients and organic matter by salt ions, through cation exchange, chemical complexation, changes in pH, solubility and biogeochemical reactions<sup>9–11</sup>. As a result, metals and radionuclides in soils are mobilized and enter waterways, salt ions and metals from drying lake beds are lofted into the atmosphere, and drinking water pipes are corroded by salt ions.

Water cycling is also impacted by salinization, as it alters water density, freezing temperature and evaporation rates. For example, salt-rich dust from drying lakebeds has the potential to accelerate regional snow melt, which can be a problem for many communities on snowpack for water supply<sup>62</sup>, such as in the Western USA. Warming sea surface temperatures could increase evaporation and aerosolization of salts in droplets in air and cloud albedo<sup>114</sup>; aerosols scatter solar radiation and serve as seed particles for cloud drops, which could in turn affect precipitation. This section discusses cascading impacts across the Earth system.

#### Land and aquatic ecosystems

FSS refers to the suites of chemical, physical and biological impacts of salt ions on natural, built and social systems, but some ecosystem consequences are still complex to diagnose and connect comprehensively as interrelated symptoms of FSS. For example, salinization impacts on the physical and chemical properties of soils and waters lead to many overwhelming and coinciding changes in biological and human health impacts at the ecosystem scale. Atmospheric mobilization and deposition of salt ions can impact soil fertility, air quality, eutrophication and human health in distant locations<sup>115</sup>. Increasing soil salinization impacts aggregate structure, erosion, hydraulic conductivity and infiltration rates, contaminant mobilization in soils, and crop growth<sup>96,116</sup>. Salinization can also lead

to losses in sensitive species of animals and plants and decreases in soil biodiversity<sup>117</sup>. Overall, the impacts of FSS are broad and extensive, and some impacts are probably still yet to be discovered.

Similarly, salinization has led to reductions in survival, growth and reproduction among multiple freshwater organisms and over a wide range of salt concentrations and types<sup>118,119</sup>. As a few examples, increasing salt concentrations impacts organisms by changing osmotic pressures, desiccating plant roots, impacting development, survival, mortality and parasitism of amphibians and other organisms, and increasing the invasion of salt-tolerant organisms<sup>96,120</sup>. In freshwater ecosystems, increased salinization can also lead to declines in zooplankton, which leads to increased phytoplankton growth and potentially harmful algal blooms<sup>118</sup>. Impacts of algal blooms on water quality can be compounded by salinization impacts on density stratification and the vertical mixing of lakes<sup>3</sup>. Salinization can also enhance pathogen survival<sup>13</sup> and alter microbial communities in both terrestrial soils and aquatic sediments<sup>121</sup>. Overall, increased salinization in terrestrial and aquatic systems can cause ecological community shifts with wide-ranging ecosystem impacts.

#### Human health and infrastructure

There are cascading direct and indirect human health impacts associated with salinization<sup>8,122</sup>. There are direct impacts on respiratory illnesses, mosquito-borne diseases and mental health from salinization-related environmental degradation<sup>123</sup>. For example, sodium in drinking water impacts hypertension<sup>124</sup> and hypertensive disorders in mothers before birth<sup>125</sup>. Atmospheric deposition of urban dust enriched with salt ions can increase risk of pulmonary toxicity, cancers via inhalation, ingestion and dermal contact, and impacts on biodiversity<sup>115,126–128</sup>. Road salts can contribute to atmospheric deposition, but little is known about the health impacts of road salt aerosols and dust inhalation.

Increased salinization also has indirect effects on human health by mobilizing contaminants, metals and radionuclides in the environment, such as radium (Ra), radon (Rn), thorium (Th) and uranium  $(U)^{10,129,130}$ . For example, salinization increases water–rock interactions, which cause the release of trace elements, including radionuclides, into the aquifer<sup>131</sup>. The abundance of Na+ ions causes increased competitive adsorption of radium with other cations and forms soluble radium complexes with Cl– (refs. 130,132). Uranium is released by dissolution from minerals, desorption from mineral surfaces, formation of uranium complexes and precipitation<sup>129</sup>. In drinking water, excess radionuclides can cause a variety of health issues, including cancers and nephrotoxicity<sup>129</sup>.

Infrastructure and urban environments, like roads, bridges, stormwater management, vehicles, road safety and metal infrastructure, are also impacted by salinization<sup>13</sup>. For example, salts in pipes cause corrosion and metal leaching, impacting safe drinking water and human health<sup>122,129,133</sup>. Salinization also affects boiling points of water, energy production using steam and build-up of mineral scaling in piped infrastructure.

In the USA, for example, SO42– concentrations have decreased in freshwaters because of air pollution regulations and decreases in SO2 emissions from power plants<sup>134</sup>, whereas Cl–

concentrations have simultaneously increased in streams of this same region owing to lack of road salt regulations69. Increased Cl–:SO42– mass ratios increase corrosion potential and leaching of metals such as Pb and Cu from drinking water pipes<sup>133</sup>.

#### **Biogeochemical cycles**

The anthropogenic salt cycle has the potential to influence biogeochemical cascades. Salinization can influence episodic acidification<sup>67</sup> and long-term alkalinization of rivers<sup>68</sup>. For example, coupled relationships between DIC and Ca2+ and Mg2+ influence alkalinity and pH, microbial processes such as nitrification and denitrification<sup>135</sup>, desorption of P from sediments<sup>121</sup>, toxicity of metals and the ability of rivers to neutralize coastal ocean acidification<sup>68</sup>. Long-term increases in pH and alkalinity can further influence the solubility of certain fractions of organic matter and quality of dissolved organic carbon transported in rivers (which can then affect transport of metals bound to organic matter and other biogeochemical cycles)<sup>136</sup>. In addition, emissions of CO2 from streams and rivers are strongly influenced by pH, and river alkalinization from carbonate salts.

The magnitude and scope of FSS have expanded well beyond only a freshwater issue. Salinization impacts can extend along an increasingly saltier hydrologic cycle across air, land and water. Different salt ions are added along each step of the hydrologic cycle in response to compounded human impacts via polluted runoff, increased evaporation rates through impoundments, irrigation, and climate change, addition of salts during water and wastewater treatment and wastewater discharges, mobilization of saline dust from drying lakes, saltwater intrusion, and other processes. However, the anthropogenic salt cycle is also driven by human alteration of geological and atmospheric processes. Thus, the impacts of the anthropogenic salt cycle transcend past the apparent boundaries of freshwaters and extend into terrestrial and atmospheric systems. Ultimately, salt is an increasing agent of global change with cascading impacts across air, land and water similar to greenhouse gases and nutrients.

#### Summary and future perspectives

Humans have redistributed salts among the terrestrial and aquatic ecosystems of the Earth, disturbing the natural salt cycle by driving rising salt concentrations and fluxes in many pools across Earth's systems. The rates of change in these fluxes vary but are much more rapid than natural background rates and timescales (Table 2). There are consequences for natural and engineered environments, including FSS, soil salinization and infrastructure corrosion, which propagate impacts across the Earth system (Supplementary Table 1).

Many questions remain regarding constraining the global salt cycle and understanding its cascading impacts. Most fundamentally, the global extent of salinization of soils and freshwaters needs to be more accurately measured. There are important spatial gaps in our knowledge, particularly regarding the magnitude and scope of salinization of freshwaters in South America, Asia and Africa (Fig. 2). These gaps can be addressed through international collaborations with researchers, citizen science and standardized measurements and data sets. There is also a need to estimate historical baselines of freshwater salinity across

different geographic regions to understand disturbances to the natural salt cycle. This could be investigated through modelling or palaeoecological studies of diatoms or other salinity proxies.

Anthropogenic impacts on the salt cycle need to be better understood to manage impacts on the salt cycle. Quantifying and constraining salt budgets from watersheds to continental and global scales can help researchers and policymakers to better understand the storage and retention of salts in soils and groundwater. An understanding of social dimensions of salt use could also aid the management of salt budgets at local watershed scales. Beyond direct inputs of anthropogenic salts to the environment, there are also many anthropogenically enhanced processes contributing to salinization (Fig. 1). Land use change, hydrologic alterations and dredging of channels promoting saltwater intrusion, for example, impact salt cycling, but the mechanisms and magnitude of these impacts are poorly documented. Similarly, consequences probably arise from extreme events (droughts, saltwater intrusion events and road salting during blizzards) that cause rapid changes in salt ion concentrations, but have been poorly characterized. Targeted monitoring of extreme events using sensors and proxies such as electrical conductivity for salt ions and associated chemical cocktails could yield new insights and enable better environmental planning.

FSS could become an existential threat to our freshwater supplies and generate a freshwater crisis. The research community must address strategies for managing FSS, such as analysing the potential for releasing water from reservoirs and tributaries to dilute salinity or conservation, restoration and stormwater management strategies. Upstream approaches are also needed to prevent FSS, but options are currently fraught. Adoption of treatment upgrades, such as reverse osmosis systems, to remove salts increases the carbon footprint of water treatment and creates additional issues such as brine disposal and loss of water production capacity<sup>6,12</sup>. Reverse osmosis is also very expensive (both for installation and maintenance), and there are many regions of the world that cannot afford it. Salt ions are added at almost every step of engineered water management, and the ways water is reused and recycled for urban agricultural uses can further contribute to salinization of freshwater along the water cycle. Improving our understanding of the anthropogenic acceleration of the salt cycle is critical to generate measures to protect human health and the environment and to avoid shortfalls in high-demand salt resources.

A systems-level approach is now needed for studying, quantifying and constraining the anthropogenic salt cycle and can illuminate improved empirical and modelling studies and improve management approaches in the future. Human activities are increasing the production and consumption of multiple salt ions and accelerating the salt cycle. More work is necessary to identify when environmental limits are being exceeded for human health and ecosystem processes and services<sup>137</sup>. Given that water, land and air are now impacted by increasing salinization across Earth systems, there needs to be a planetary boundary for safe and sustainable salt use similar to other biogeochemical flows of nitrogen, phosphorus and carbon dioxide<sup>138</sup>. A new understanding of the anthropogenic salt cycle and constraining global, regional and watershed salt budgets are important for reducing salt consumption to decrease impacts on societies, ecosystems and infrastructure. More work is needed to understand how ecological communities change in response to salinization.

If environmental limits are exceeded, impacts along the anthropogenic salt cycle could cause harm to ecosystems, drinking water, human health, infrastructure and earth system processes, which can be difficult or nearly impossible to reverse over timescales relevant to human life spans.

#### **Supplementary Material**

Refer to Web version on PubMed Central for supplementary material.

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Box 1 The Shifting Balance of the U.S. Salt Budget



#### Figure 5 (Box 1).

A salt budget for the continental United States illustrating major natural and anthropogenic fluxes. The annual average NaCl production, consumption, imports, exports, and atmospheric deposition between 2013 and 2017, as well as riverine outputs of total dissolved solids (TDS) indicated by \* in the continental United States, reported in million metric tons (Mt)<sup>2</sup>. Data on production, consumption, imports, and exports were obtained from the United State Geological Survey (USGS) Annual Minerals Yearbook<sup>82</sup>. Atmospheric deposition values were interpolated and estimated from Kaushal et al. (2021) <sup>2,8</sup>.

Ice control + road stabilization: 21.5 Mt (43.19%)

Chemical: 18.7 Mt (37.48%)

Distributors

4.23 Mt (8.49%)

Food processing: 1.76 Mt (3.54%)

Atmospheric deposition: 2.29 Mt (NaCl)

-1.45 Cl -0.84 Na

U.S. production: 42.3 Mt (NaCl) Agricultural: 1.38 Mt (2.78%) General industrial:

0.87 Mt (1.74%)

Water treatment:

0.55 Mt (1.10%)

Other: 0.84 Mt (1.68%)

U.S. imports: 15.7 Mt (NaCl)

Riverine output: 272 Mt (TDS\*) -194 from geologic sources -37.8 from road deicers

-18.2 from pastures
-13.9 from urban areas
-7.90 from cultivated lands

-5.79 from Chile -4.75 from Canada

-1.80 from Mexico

In the continental USA, anthropogenic salt fluxes are within similar orders of magnitude as salt fluxes from atmospheric deposition and riverine transport<sup>2</sup> (see the figure), based on values reported in the US Geological Survey (USGS) annual Minerals Yearbook<sup>66</sup>. Atmospheric deposition values were interpolated and estimated from Anning and Flynn<sup>2</sup> and Kaushal et al.<sup>8</sup>.

Increased salt use in the USA impacts salt fluxes in streams and rivers. Approximately 271.9 Mt of total dissolved solids are transported to streams within the USA, with 71.4% originating from geologic sources, 13.9% originating from road deicers, 6.7% originating from pastures, 5.1% originating from urban areas and 2.9% originating from cultivated lands<sup>2</sup>. Road salt use increased rapidly after 1990 in the USA, as road deicers began replacing sand use, and road salt use has increased with increasing impervious surface cover over time<sup>182</sup>. Road salting is now a major use of salt in colder climates, accounting for approximately 44% of salt use in the USA between 2013 and 2017 (ref. 66).

Salt for chemical use increased rapidly until the 1970s in the USA, when there were plateaus and declines in manufacturing and certain industrial activities<sup>66</sup>. In addition, environmental regulations such as US Federal Clean Air, Clean Water, and Safe Drinking Water Acts were passed in the 1970s, leading to fundamental shifts in the uses and disposal of industrial chemicals across the USA<sup>183,184</sup>. Wastewater is also an important (and overlooked) source of salinization missing from previous estimates and budgets; chemicals used in wastewater and water treatment (for pH adjustment, chlorination, dechlorination and odour control) can account for almost 8% of the Na+ entering a drinking water reservoir whereas the contribution of wastewater effluent to total Na+ mass loading entering that same drinking water reservoir can be approximately 60–80% during dry weather<sup>12</sup>.

The relative importance of riverine salt fluxes and anthropogenic salt consumption in the USA is estimated and compared here. Assuming similar concentrations to global averages for unpolluted freshwaters<sup>185</sup>, NaCl is approximately 7.2% of the majority of total dissolved solids (6.71/93.71 mg l–1)185, suggesting a flux of 19.59 Mt (based on a total dissolved solid flux of 272 Mt). From 2013 to 2017, average US NaCl consumption and use (agriculture, industry and road salt, for example) is 49.8 Mt; therefore, riverine fluxes are equivalent to approximately 39% of NaCl consumption and use in the USA. While acknowledging uncertainties, these values conservatively suggest that NaCl transport in rivers can be within the same order of magnitude as anthropogenic salt use in the USA. As salts are stored in the subsurface (for example, as legacy pollution), riverine exports of non-geologic salts could be less than all inputs in some cases.

However, river outputs could be over or underestimated, and this example is missing inputs from natural geologic sources and internal sources from vertical groundwater transport. Irrigation is also not shown, which is a major driver of salinization in some river basins in the USA such as the Colorado River. Ultimately, quantifying the anthropogenic salt cycle in biogeochemical budgets is a difficult but urgent problem<sup>186</sup>.



#### THE NATURAL SALT CYCLE: SOURCES, TRANSPORT, AND STORAGE

#### THE ANTHROPOGENIC SALT CYCLE: SOURCES AND SALINIZATION PROCESSES



#### Figure 1.

a, The natural salt cycle is characterized by a balance in uplift of salts to the surface of the Earth and weathering and transport of salts to the oceans. Salinization is a natural process in many dryland environments (inset). b, The anthropogenic salt cycle is characterized by accelerated transport of salts to the surface of the Earth by mining and resource extraction, increased fluxes of salt to the atmosphere from saline dust, and increased soil salinity and evaporite formation owing to desiccation (inset). Anthropogenic sources of salts exceed natural sinks, with a wide variety of geological, chemical, biological, engineering and hydrological processes contributing to human alteration of the global salt cycle.

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a, Rising trends in global production of salts since 1929, as reported by the USGS<sup>66</sup>. b, Rising NaCl production (defined as the quantity of salt mined or manufactured that is available for sale) and consumption (defined as the quantity of salt sold or used, plus imports, minus exports) in the USA, with major uses of consumption shown<sup>66</sup>.



#### Figure 3.

Global heterogeneity in environmental salinity. a, Soils impacted by anthropogenic salinization (red-shaded areas) to at least some degree as of 2016 (ref. 93). b–e, Probability density plots showing the distributions of riverine (solid line) or groundwater (dashed line) freshwater conductivity values among countries or regions with 100 sites from an open-access global database of observations made over the past four decades178. Note that Eastern Australia includes Queensland, New South Wales, Victoria, Australian Capital Territory and Tasmania. Central Australia includes South Australia and Northern Territory. Electrical conductivity in water often serves as an easily measured proxy for total salinity in freshwater. Distributions reflect mean readings drawn from disparate river and stream or groundwater sampling locations from within each country or region. Coastal sites were omitted from distributions. Country or region site sample sizes are provided in the density plot legends.



#### Figure 4.

Sodium and chloride concentrations in rivers. a,b, Rising trends in concentrations of Na+ and Cl– salt ions in select world rivers. c, Deviations in Na+:Cl– molar ratios from 1:1 (refs. 179–181) (indicated by dashed line). Variations Na+:Cl– molar ratios reflect variations in the sources, transport and transformation of salt ions across climate, geology, human activities, flowpaths and time. Rivers draining semiarid and arid regions with more sodic soils can have higher Na+:Cl– molar ratios whereas rivers draining humid regions have lower relative Na+:Cl– molar ratios owing to road salt (and other pollution sources) and greater retention of Na+ on soil ion exchange sites compared with Cl–. Data from the Global Freshwater Quality Database GEMStat.

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groundwaters, where salt ions undergo ion exchange. Fluvial transport of salts to coastal margins can lead to sedimentation, biogenic precipitation of salts, Salt is brought to soil surfaces through evaporation, capillary action, and diffusion in dryland soils. Salts in dust, sea spray, and aerosols become airborne and transported through the atmosphere before deposition onto Earth's surface. Weathering increases the dissolution of rocks and increases the transport groundwaters can be exposed at Earth's surface by weathering. Salt layers formed or deposited deeper within the Earth are brought to the surface by salt Some examples of major processes and fluxes in Earth's natural salt cycle. Salinization can be a natural process, particularly in dryland environments. and reverse weathering at coastal margins. Evaporation of sea water can lead to the formation of evaporites. Salts, evaporite layers, and connate of salts in runoff. Salts can be taken up and stored in biological pools. However, a significant fraction of salt ions can infiltrate into soils and tectonics, uplift, exposure, and dessication. Natural saltwater intrusion can bring salts from marine environments back into freshwaters.

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Processes	Flux Directions	Descrintions of Fluxes	- Ryamulee of Rafee
Geological uplift	Lithosphere -> Lithosphere	Rock units including salt layers, which form diapirs, slowly rise to Earth's surface <sup>36-40</sup> .	Uplift velocity ranges from 0.1 to 10 mm year <sup>-1 38,40,158,159</sup> .
Chemical weathering	Lithosphere -> Hydrosphere	Evaporites dissolve from chemical weathering <sup>54–56,58,59</sup> . Base cations are liberated <i>via</i> weathering of silicate and carbonate rocks (bedrock) <sup>52</sup> .	Cation weathering (Na <sup>+</sup> , K <sup>+</sup> , Ca <sup>2+</sup> , Mg <sup>2+</sup> ) from silicates is 200 * $10^{6}$ tonnes yr <sup>-1</sup> globally. Total dissolved solids from evaporites is 290 * $10^{6}$ tonnes yr <sup>-1</sup> globally <sup>52</sup> .
	Lithosphere -> Pedosphere	Salts in mineral soils primarily come from the breakdown of rocks and minerals, including carbonates and feldspars <sup>41,42</sup> .	Soil Production Rate ranges between 0.1 and 594 mm kyr <sup>-1</sup> , with a mean of 67 mm kyr <sup>-1</sup> and median of 27 mm kyr <sup>-1 160</sup> .
	Pedosphere -> Hydrosphere	Weathering of soil minerals releases major ions.	Weathering rates can be 0.005 to 11 keq ha <sup><math>-1</math></sup> yr <sup><math>-145</math></sup> .
Atmospheric transport and deposition	Lithosphere and Pedosphere -> Atmosphere	Dust is mobilized from dry areas globally including arid soils, dried lake beds, and salt flats.	735 to 8186 Tg dust yr <sup>-1</sup> <sup>161</sup> . On an areal basis, global dust deposition ranges from -0 to >450 g m <sup>-2</sup> yr <sup>-1</sup> , but are typically <1 g m <sup>-2</sup> yr <sup>-1 33</sup> .
	Hydrosphere -> Atmosphere	Sea spray is the primary source of aerosolized atmospheric salts <sup>28</sup> .	Sea spray aerosol mass is about $3-70 * 10^{12}$ kg yr <sup>-1</sup> globally <sup>162</sup> . Global sea salt aerosol deposition is about $1-3 * 10^{16}$ g yr <sup>-1 31</sup> .
	Atmosphere -> Lithosphere, Pedosphere, Hydrosphere	Salts can be deposited through wet or dry deposition <sup><math>28-30</math></sup> .	In the U.S., atmospheric deposition ranges from: $0-5~kg~ha^{-1}$ of Na <sup>+</sup> , $0-3~kg~ha^{-1}$ of Ca <sup>2+</sup> , $0-1~kg~ha^{-1}$ of Cl <sup>+</sup> , $0-24~kg~ha^{-1}$ of SO <sub>4</sub> <sup>163</sup> .
	Atmosphere -> Cryosphere	Dust, including Na <sup>+</sup> and Ca <sup>2+</sup> , deposits on snow and ice. Snow and ice deposition can accelerate snowmelt <sup>35</sup> .	Less than 0.05 g m <sup>-2</sup> yr <sup>-1 32</sup> .
Biological uptake and release	Pedosphere, Lithosphere, Hydrosphere <-> Biosphere	Plants, fungi, and other biological organisms take up salt ions, depending on their nutrient needs and salt stress <sup>29,43–48</sup> . Salt ions can be released from organisms to soils and waters (such as through processes such as decomposition) <sup>29,164,165</sup> and soil organochlorine formation <sup>30,44,166</sup> .	Unknown globally, see examples at local and regional scales <sup>29,44,164,165,167,168</sup>
Evaporation and transpiration	Hydrosphere -> Lithosphere	Solar evaporation leads to increased salt concentrations in marine and non- marine waterways, from which minerals can precipitate and form evaporitic rocks.	Approximately mm to cm of evaporite thickness accumulate yr <sup>-1 59</sup> .

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	Hydrosphere -> Pedosphere	Evaporation and plant transpiration pull groundwater towards the surface <i>via</i> capillary action, accumulating salts in soils.	Unknown on global scale, see examples at local and regional scales <sup>169–</sup> 171
	Hydrosphere -> Hydrosphere	When evaporation exceeds precipitation, salinity increases in ocean water <sup><math>172,173</math></sup> and groundwater <sup>64</sup> .	Up to 0.3 psu month <sup>-1 172</sup> .
Mineral precipitation	Biosphere, Hydrosphere -> Lithosphere	Many salt ions can form mineral precipitates and we give calcite as an example. Most modern calcium carbonate is formed through biological processes <sup>70</sup> .	As an example of mineral precipitation, ~32 $\ast$ 10 <sup>12</sup> mol of CaCO <sub>3</sub> yr <sup>-1</sup> accumulates globally <sup>174</sup> .
Reverse weathering	Hydrosphere -> Lithosphere	Reverse weathering controls major ion ratios and alkalinity in sea water through the deposition of silica minerals involving divalent cations, primarily forming $clays^{71-73}$ .	The reverse weathering reaction coefficient varies between $4*10^{-6}$ to $10^{-10}$ µmol cm <sup>-3</sup> s <sup>-1175</sup> .
Hydrothermal circulation	Hydrosphere -> Hydrosphere, Lithosphere	In locations of high heat flow, such as rift valleys and subduction zones, the water in seawater is turned to vapor and expelled, increasing brine salinity until salt precipitates <sup>64,74,75</sup> . Processes occurring at hydrothermal vents can also be a sink for some elements, such as $Mg^{2+}$ , while other elements are enriched <sup>176,177</sup> .	The chemical fluxes of hydrothermal vent fluid of salt ions vary from a sink of 1.6 * $10^{12}$ mol Mg <sup>2+</sup> yr <sup>-1</sup> to a source of 2.3–6.9 * $10^{11}$ mol K <sup>+</sup> yr <sup>-1</sup> $177$ .
Brine rejection	Cryosphere -> Lithosphere	When sea ice forms, salt is rejected from the ice, increasing salt concentrations and forming brines in oceanic water <sup>178–183</sup> .	Salt flux from brine rejection has been observed at 6.4 * $10^{-7}$ to 1.5 * $10^{-6}~{\rm g~cm^{-2}~s^{-1182}}$ .
Saltwater intrusion	Hydrosphere -> Hydrosphere	Natural mechanisms leading to salt water intrusion include: tidal fluctuations, long term climate change/marine transgression, flooding, and salt aerosol inputs through rain <sup>41,42,64,67,68</sup> .	On a global scale, natural rates are variable depending on the process $^{67.68}$ .
Connate fluid mixing	Lithosphere -> Hydrosphere	Connate fluid is a very salty water that is trapped within rock during formation. However, eventually the fluid will migrate and can lead to salinized groundwater <sup>60–64</sup> .	Rates vary naturally based on uplift/erosion rates, rock permeability, glacial/interglacial cycles $^{60-63}$ .
Soil ion exchange and transport	Pedosphere -> Pedosphere	Cation exchange is a key process that dictates how salts are stored and transported in soils. Salt migration in soils typically follows a convective or diffusive pattern <sup>29,42,49,51,184</sup> .	Cation exchange capacities vary from <10 to 300 cmol charge $kg^{-1}$ , depending on the soil material <sup>185</sup> .
Hydrological transport	Hydrosphere -> Hydrosphere	The hydrological cycle is a key transporter of salt ions. The largest flux is from rivers to oceans $^{19,52}$ , although transport also happens within groundwater <sup>30</sup> .	Yields range from 0.60 to 4.04 g m <sup>-2</sup> basin area yr <sup>-1</sup> , depending on the $ion^{19}$ .

Earth's anthropogenic salt cycle. A growing research need is to identify and quantify perturbations to sources,	in the anthopogenic salt cycle.
l fluxes in Earth's anthropoge	ions within the anthopogenic
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Examples of Anthropogenic Processes	Examples of Flux Directions	Examples of Anthropogenic Fluxes	Examples of Rates and Magnitudes	Examples of Potential Implications
Uplift from mining	Lithosphere -> Pedosphere	Potash for agricultural production is mined or evaporated from salt lakes and subsurface brines <sup>101</sup> .	39.55 million metric tons extracted worldwide in 2014 <sup>4</sup> .	With waste to usable potash ratios upwards of 3:1, nearby freshwater rivers and streams are subject to high-salt runoff, which can alter river ecosystems <sup>4</sup> .
	Lithosphere -> Technosphere	Mining of salt rich materials for the built environment (such as limestone and dolomite) <sup>105</sup> .	22 billion tons extracted globally $yr^{-1.186}$ .	Noise, dust, pollution; decline in biodiversity; lower water table <sup>187</sup> ; depletion of resources <sup>186</sup> .
	Lithosphere -> Hydrosphere	Mountaintop mining releases salt ions in bedrock, including $Ca^{2+}$ , $Mg^{2+}$ , $K^+$ , $Na^+$ , $CI$ , $HCO_3^-$ , $SO_4^{2-110}$ .	Dissolved solids exported downstream from mined catchments at a rate of ~7600 kg ha <sup>-1</sup> yr <sup>-1</sup>	Predicted increases in coal mining over the next century. <sup>188</sup> may cause an increase in fluxes of ions exported from mines to downstream catchments.
Human-accelerated weathering	Technosphere -> Hydrosphere, pedosphere	Weathering of the built environment <sup>107</sup> causes dissolution, honeycomb weathering, weathered rims and rinds, spalling, and corrosion <sup>107</sup> .	0.0009 – 1.32 mm yr <sup>-1</sup> on average <sup>189</sup>	Increased major ions and pH levels impact aquatic life and infrastructure <sup>106</sup> .
Atmospheric transport and deposition	Technosphere -> Atmosphere, biosphere	Atmospheric deposition from industry, coal combustion, traffic emissions, wear of asphalt, agriculture, and wildfires <sup>98,190</sup> .	Base cation fractions of mineral dust mass are: Ca <sup>2+</sup> : 2.4%, K <sup>+</sup> : 1.5%, Na <sup>+</sup> : 1.2%, Mg <sup>2+</sup> : 0.9% <sup>98</sup> .	Urban dust can increase risk of pulmonary toxicity and cancers via inhalation, ingestion, and dermal contact; also correlated to declines in invertebrate populations
	Pedosphere, Lithosphere -> Atmosphere	Atmospheric dust transport from farming, grazing, water use, urban development <sup>24,25,93–</sup> 95,191,192,	Estimates range from small compared to natural sources <sup>24</sup> to 50 +/- 20% of atmospheric dust mass <sup>191</sup> .	Increased aeolian transport can increase rates of soil erosion, salt deposition, groundwater salinization, ocean and stream sedimentation, climate change, disease transmission, air pollution, and drinking water contamination <sup>95</sup> .
	Hydrosphere -> Atmosphere	Aerosolization of road salts, primarily $\mathrm{Na^+}$ and CI during winter months <sup>96</sup> .	During winter months, there is a four-to-five-fold increase in daily average concentration of CI aerosols <sup>193</sup> .	Increased Na $^+$ and Cl <sup>-</sup> concentrations occur during winter.
Wastewater and industrial discharges and water treatment	Technosphere -> Hydrosphere, Pedosphere	Salt ions, including $Ca^{2+}$ , $Mg^{2+}$ , $K^+$ , $Na^+$ , $CI^+$ , and $SO_4^{-2}$ in household products and cleaning supplies can be released into domestic wastewater and into streams and landfills.	Per household rates of release are: $Ca^{2+1}$ :11236 mg L <sup>-1</sup> ; C1: 38768 mg L <sup>-1</sup> ; K <sup>+</sup> : 18713.8 mg L <sup>-1</sup> ; Mg <sup>2+</sup> : 3528 mg L <sup>-1</sup> ; and Na <sup>+</sup> : 90629 mg L <sup>-1</sup> 118.	Adoption of treatment upgrades, such as reverse osmosis systems, to remove salts increases the carbon footprint of water treatment and creates additional issues with respect to brine disposal <sup>11</sup> .
	Hydrosphere -> Technosphere -> Biosphere	Salts are taken up by humans through consumption of food and beverages and released into domestic wastewater through excretion.	Average consumption of $Na^+$ day <sup>-1</sup> per person in the USA is 3400–4000 mg day <sup>-1</sup> ; Average excretion of $Na^+$ is 3414–3803 mg day <sup>-1</sup> 116,117.	High-salt diets increase risk of hypertension and high blood pressure <sup>194</sup> .

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Examples of Anthropogenic Processes	Examples of Flux Directions	Examples of Anthropogenic Fluxes	Examples of Rates and Magnitudes	Examples of Potential Implications
	Technosphere -> Hydrosphere	Salt ions from industrial plants are released from into water bodies.	Average Na <sup>+</sup> load discharged from a microfabrication facility is ~1000 kg day <sup>-111</sup> .	Stringent pre-treatment requirements for industrial effluent discharges should be developed and enforced <sup>11</sup> .
	Hydrosphere -> Hydrosphere, Biosphere	Na <sup>+</sup> and Cl <sup>-</sup> are added to water during conventional wastewater and drinking water treatment processes ( <i>e.g.</i> , softening, corrosion control, disinfection, coagulation).	Unknown globally, see examples at local and regional scales <sup>[11,195</sup> . In the US, an average of 1.04 Mt NaCl were used for water treatment and conditioning annually between 2013 and 2017 <sup>82</sup> .	Increased salt ions in receiving waters, which can harm aquatic life and impact mobilization of contaminants from sediments to streams and rivers.
	Technosphere -> Hydrosphere	There are excess loads of major ions transported in rivers from wastewater discharges.	$\begin{array}{l} \mbox{Per capita loads in urban sewage (kg cap^{-1} yr^{-1}): \\ 1.2-3.2 (Ca^{2+}), 0.65-1.5 (Mg^{2+}), 6.4-14 (Na^{+}), \\ 1-2 (K^{+}), 6.3-15 (Cl^{-}), 5.8-13.5 (SO_4^{2}), 14.5-24 \\ (HCO_3^{-})^{50}. \end{array}$	Increased major ions in rivers from urban sewage discharges could influence pH, alkalinity <sup>84</sup> , and microbial processes such as nitrification and denitrification, P desorption <sup>84</sup> , and also pathogen survival <sup>20</sup> .
Road salting	Pedosphere -> Lithosphere -> Hydrosphere	Road salts can be transported (a) rapidly to streams through conduits, sub-surface piping, and preferential flowpaths; (b) diffusely through aquifers <sup>111</sup> ; and (c) vertically as salt plumes through unsaturated zones and aquitards <sup>112</sup> .	Lateral salt transport in roadside soils at rate of $25-50 \text{ cm day}^{-1}$ , vertical salt transport at rate of $1.3-2.3 \text{ cm day}^{-1}$ 103. Cl <sup>-</sup> mass input to aquifer at average rate of 30 g m <sup>-2</sup> year <sup>-1</sup> 11 <sup>2</sup> .	Higher baseline concentrations of Cl <sup>-</sup> in streams, wells, and aquifers. Sodic conditions in roadside soils weaken soil structure, causing colloidal dispersion and accelerated adsorption- dispersion reactions <sup>104</sup> .
Agriculture	Hydrosphere -> Pedosphere, Hydrosphere	Irrigation causes evaporative concentration of salts in soil water, increasing salts in soils and groundwater <sup>64</sup> . Irrigation can raise the water table bringing saline groundwater towards the surface <sup>64,83</sup> .	Depending on the irrigation method, groundwater total dissolved solids can increase at rates ranging from $3-5$ to $15-17 \text{ mg L}^{-1} \text{ yr}^{-1196}$ .	Increased soil salinity can lead to crop yield reductions between 10–25% in salt-sensitive species <sup>83</sup> .
	Technosphere -> Pedosphere, Hydrosphere	Fertilizer application, including potash, increases salts (such as $K^+$ , $Cl^+$ , and $SO_4^{-2}$ ) in soils and salt export to waterways <sup>102,197,198</sup> .	Globally ~32.2 Tg K <sup>+</sup> yr <sup>-1</sup> and ~70 Tg S yr <sup>-1</sup> are applied through fertilizer (Schlesinger 2021; Hinckley et al., 2022). Watershed Cl <sup>-</sup> inputs from potash are around 49 kg ha <sup>-1</sup> yr <sup>1102</sup> .	Increased S and salinity change pH, acid-base status, alkalinity, and buffering capacity, and can lead to methyl mercury production <sup>102</sup> .
	Technosphere -> Pedosphere, Hydrosphere	Aglime application adds HCO <sub>3</sub> , $Ca^{2+}$ , and $Mg^{2+}$ to soils, part of which is transported to waterways <sup>87,199</sup> .	In the USA in 2018, 86 thousand metric tons of lime was sold or used as fertilizer or aglime <sup>82</sup> .	Aglime application increases pH, emits CO <sub>2</sub> , and influences downstream carbonate precipitation <sup>87,199</sup> .
Anthropogenic riverine transport to oceans	Pedosphere -> Lithosphere -> Hydrosphere	Increased concentrations of major ions in streams and rivers have been documented over the past 50 years on continental and global scales <sup>8,16,108,109</sup> .	In 1970, estimated anthropogenic salt inputs to the ocean were (in $10^{6}$ t yr <sup>-1</sup> ): 47 Ca <sup>2+</sup> , 10.5 Mg <sup>2+</sup> , 78 Na <sup>+</sup> , 5 K <sup>+</sup> , 93 Cl <sup>-</sup> , 124 SO <sub>4</sub> <sup>2-</sup> , and 100 (HCO <sub>3</sub> <sup>-</sup> ) <sup>19</sup> .	DIC, $Ca^{2+}$ , and $Mg^{2+}$ influence alkalinity and pH, $CO_2$ emissions, desorption of P from sediments, toxicity of metals, and the ability of rivers to neutralize coastal ocean acidification <sup>108</sup> .

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