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Resetting of soil compositions by irrigation in urban watersheds: Evidence from Sr isotope variations in Austin, TX

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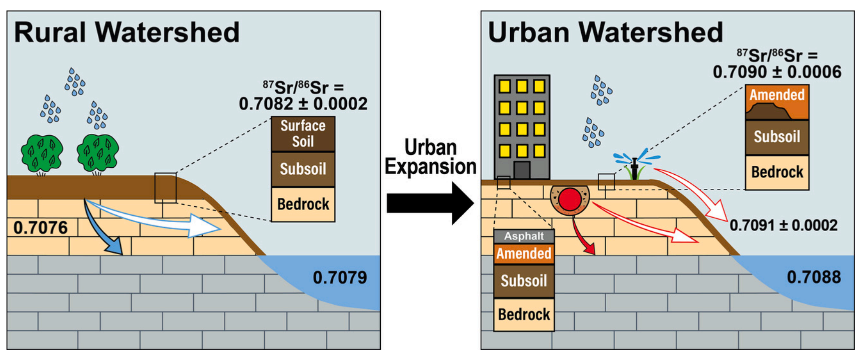
HIGHLIGHTS

- Soil Sr isotope ratios ($^{87}Sr/^{86}Sr$) are used to assess the impacts of urbanization.
- Elevated urban streamwater $^{87}Sr/^{86}Sr$ values are not from interaction with soils.
- Soil $^{87}Sr/^{86}Sr$ values are reset by irrigation in urbanized watersheds.
- Resetting of soil $^{87}Sr/^{86}Sr$ values is spatially and chemically extensive.

GRAPHICAL ABSTRACT



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ABSTRACT

Human activities in urban areas disturb the natural landscape upon which they develop, disrupting pedogenic processes and ultimately limiting the ecosystem services urban soils provide. To better understand the impacts on and resiliency of soils in response to urban development, it is essential to understand the processes by which and degree to which soil physical and chemical properties are altered in urban systems. Here, we apply the source-tracing capabilities of Sr isotopes ($^{87}Sr/^{86}Sr$) to understand the impacts of urban processes on the composition of soils in eight watersheds in Austin, Texas. We evaluate natural and anthropogenic Sr sources in watersheds spanning a wide range of urbanization, comparing soils by variations in their natural (including mineralogy, thickness, soil type, and watershed) and anthropogenic (including irrigation, soil amendments, and fertilization) characteristics. A strong positive correlation between soil thickness and $^{87}Sr/^{86}Sr$ is observed among unirrigated soils ($R^2 = 0.83$). In contrast, this relationship is not observed among irrigated soils ($R^2 = 0.004$). 95 % of 42 irrigated soil samples have $^{87}Sr/^{86}Sr$ values approaching or within the range for municipal supply water. These results indicate soil interaction with municipal water through irrigation and/or water infrastructure leakage. Soils irrigated with municipal water have elevated $^{87}Sr/^{86}Sr$ values relative to unirrigated soils in seven of eight watersheds. We propose that original soil $^{87}Sr/^{86}Sr$ values are partially to completely reset by irrigation with municipal water via ion exchange processes. Our results demonstrate that in urban systems, Sr isotopes can serve as an environmental tracer to assess the overprint of urbanization on natural soil characteristics. In the Austin

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region, resetting of natural soil compositions via urban development is extensive, and the continued expansion of urban areas and irrigation systems may affect the ability of soils to retain nutrients, filter contaminants, and provide other ecosystem services that support environmental resilience.

1. Introduction

Urbanization has long been a major agent of land use and land cover change, driven by the increasing proportion of the global population residing in urban areas. Soils are susceptible to urbanization processes via impacts on soil quality, structure, and composition (Mironova et al., 2022; Marcotullio et al., 2008; Pickett and Cadenasso, 2009). Urbanization is projected to accelerate during the 21st century (Chen et al., 2022), which will lead to the disturbance of natural soils via land clearing, shifts in plant/soil resource availability, and alteration of physicochemical conditions—resulting in urban ecosystems that are distinct from their nonurban counterparts (Pavao-Zuckerman, 2008; Kaye et al., 2006).

Physical urbanization processes include topsoil removal, grading, and establishment of amendments (e.g., externally-derived soil, fertilizer, mulch, lime) to address the effects of interrupting soil development (Chen et al., 2014; Alaoui et al., 2011; Jim, 1998; Effland and Pouyat, 1997; Pavao-Zuckerman and Pouyat, 2017). Chemical urbanization impacts are less visible and occur through changes in land management and hydrology. For example, soil compaction and impervious cover can reduce infiltration rates, increasing surface water runoff and mobilization of accumulated pollutants such as anthropogenic heavy metals and volatile organic compounds (Jia et al., 2018; Pouyat et al., 2008). In addition, urbanization is associated with high concentrations of exchangeable soil cations and increased pH (Huang et al., 2015; Lovett et al., 2000), although this influence is not consistent across urban centers (Takahashi et al., 2015).

The irrigation of urban landscapes can cause vertical changes in soil physicochemical properties (Pouyat et al., 2020; Çadraku, 2021; Steele and Aitkenhead-Peterson, 2012). In the U.S., a cultural predilection for lush, manicured green spaces contributes to the rapid expansion of irrigation in urbanizing areas (Milesi et al., 2009). However, there is little published research on the chemical impacts of municipal water irrigation on soils in urban settings, despite their relevance for understanding and monitoring fragile urban ecosystems (Filho et al., 2020). Such information may better inform resilient land management and urban expansion practices. For this reason, we aim to understand the chemical impacts of urban expansion practices, focusing on urban irrigation, by evaluating the evolution of soils that have experienced a range of extents of urbanization.

Previous studies have used anthropogenic particulates, heavy metals, trace elements, and polycyclic aromatic hydrocarbons (PAHs) to trace urbanization impacts and assess soil alteration (Cannon and Horton, 2009; Argyraki and Kelepertzis, 2014; Peng et al., 2013). While elevated concentrations of these contaminants can primarily be attributed to anthropogenic activities, there are several limitations in tracing their source(s) (Sun et al., 2018). For example, evaporation, biological fractionation, dilution, and mineral precipitation may affect the concentrations of these species (Christian et al., 2011). In contrast, naturally occurring isotopes of heavy elements (e.g., Sr, Nd, Pb) are not significantly fractionated by these processes (Åberg, 1995) and have been used to identify and distinguish sources of pollution in urban settings (Delile et al., 2017; Sun et al., 2018)—making them useful for tracing urbanization impacts on soils (Li et al., 2010). Sr isotopes ($^{87}Sr/^{86}Sr$), in particular, have been used as diagnostic tracers of specific environmental reservoirs, both natural and anthropogenic (Capo et al., 1998; Garcia et al., 2021; Zieliński et al., 2016).

The Austin, TX region is well-suited for evaluating the impact of urbanization on soils by using the tracing capabilities of Sr isotopes. It is one of the fastest-growing areas in the U.S., with a population increase of 560 % between 1970 and 2019 and a projected increase of 200 % between 2019 and 2050 to 4.5 million residents (Robinson, 2021). Additionally, Austin-area Sr reservoirs such as bedrock, soils, urban materials, municipal water, stream water, and groundwater can have characteristic $^{87}Sr/^{86}Sr$ values and ranges (Christian et al., 2011). Christian et al. (2011) and Beal et al. (2020) found a strong correlation between urbanization extent and stream water $^{87}Sr/^{86}Sr$ values in the Austin area, showing that streams in more urbanized watersheds have elevated $^{87}Sr/^{86}Sr$ values compared to those in rural watersheds. Elevated stream water $^{87}Sr/^{86}Sr$ values are hypothesized to be primarily controlled by inputs from municipal water with high $^{87}Sr/^{86}Sr$ values via irrigation runoff and/or water infrastructure failure. However, the isotopic influence of urban soils on stream water $^{87}Sr/^{86}Sr$ values is not fully understood, and uncertainty exists about the relationship between soil $^{87}Sr/^{86}Sr$ values and the extent of irrigation in urban settings. This is due, in part, to a poor understanding of the natural and anthropogenic controls on urban soil $^{87}Sr/^{86}Sr$ variability.

The main aims of this study are to 1) investigate the role of soils in urban stream water geochemical evolution in eight Austin-area watersheds (Fig. 1a) and 2) evaluate the impacts of urban development on soils by using Sr isotopes as tracers and analyzing natural and anthropogenic parameters, including soil series, thickness, mineralogy, watershed characteristics, and irrigation status. By evaluating Sr isotope variability in irrigated and unirrigated soils, and comparing regional soils from natural settings, we identify the extent to which urbanization processes impact soil compositions. The results allow us to infer that soil irrigation with municipal water strongly controls soil $^{87}Sr/^{86}Sr$ values. Understanding and characterizing urban soil alteration, chemical or otherwise, in relation to urban expansion is the first step for making informed decisions about land use and the provision of ecosystem services (Kumar and Hundal, 2016). As such, our results are relevant for sustainable urban development. The following two sections give an overview of the study area and methodology. Sections 4 and 5 present and discuss the natural and anthropogenic parameters that may be responsible for soil $^{87}Sr/^{86}Sr$ variability.

2. Study area

Eight watersheds in the Austin, Texas metropolitan area were investigated: Barton, Bull, Onion, Shoal, Slaughter, Waller, West Bull, and Williamson Creeks (Fig. 1). These watersheds are characterized by ephemeral and perennially flowing streams. The bedrock the Austin-area watersheds is comprised of 95 % Lower Cretaceous marine limestone and dolostone and 5 % mudstone and shale (Fig. 1c). Soil thicknesses in the area range from approximately 0.15 m in the uplands (Cooke et al., 2007) to up to 0.25 to several meters on flood plains. The dominant soil series in the watersheds of interest include Brackett, Tarrant, Volente, and Urban Land (Fig. 1d). Hereafter, all soils that are not classified as Urban Land will be described as Austin-natural soils, including Brackett, Tarrant, Volente, Austin Clay, Comfort, Houston Black, Krum Clay, Oakalla, Speck, and Lewisville. Soil series are detailed in Werchan et al. (1974) and summarized here.

Brackett soils are shallow, well-drained Inceptisols. They are clayey, loamy soils with clasts of chalk or limestone, with low available water capacity and moderately low permeability. Tarrant soils are shallow, stony, clayey Mollisols that are well-drained, moderately permeable, and have low water capacity. Volente soils are silty clays developed in slope alluvium and are deep, well-drained Mollisols. They have a high available water capacity and are moderately permeable. Urban Land soils are amended and reclaimed via urban development. Texture, composition, permeability, and water-holding capacity are highly variable among Urban Land soils. They are most prevalent in the densely-urbanized watersheds (i.e., Waller and Shoal Creek).

This study defines the degree of urbanization within a watershed as the percentage of impervious cover in 2019 (*City of Austin, Texas, Watershed Protection Department,* 2019). The City of Austin Watershed Protection Department's methodology for calculating impervious cover per watershed is given in Table 1. The eight watersheds sampled for this study are characterized by a wide range of urbanization (Fig. 2b; Table 1) and also exhibit a range in the average age and timing of urbanization onset—beginning by 1880 for Waller and Shoal Creeks, 1950 for Williamson Creek, and within the last few decades for Onion, Barton, Bull, and West Bull Creeks (Christian et al., 2011; Garcia-Fresca, 2004). The average building ages within Shoal, Waller, and Williamson Creeks range from 1964 to 1985, while Slaughter, Bull, West Bull, Barton, and Onion Creek range between 1988 and 1991 (Christian et al., 2011).

Additionally, municipal water infrastructure in Austin has expanded in recent decades to meet increasing water demand from a rapidly growing population. Austin had 132 km of municipal water mains and 121 km of sewer mains citywide in 1917. These infrastructure networks have grown to >6100 km of municipal water mains and >4650 km of sewer mains.

Table 1
Physical characteristics of Austin-area watersheds.

Watershed	Area[a] (km^2)	2003 impervious cover[b] (%)	2019 impervious cover[c] (%)
Barton	281	7.6	8
Bull	64	22	21.6
West Bull	18	9.8	10.6
Onion	547	6.2	6.8
Shoal	34	44	55.4
Slaughter	79	15	22
Waller	15	43	60.7
Williamson	80	29	37

[a] Watershed area calculations from City of Austin, Texas, Watershed Protection Department (2015).

[b] 2003 impervious cover calculations from Christian et al. (2011). Percent impervious cover determined by R. Botto using locally derived relationship between land use and impervious cover, and the City of Austin's 2003 parcel-based land use layer and impervious cover polygon layer.

[c] 2019 impervious cover calculations from *City of Austin, Texas, Watershed Protection Department* (2019). The City of Austin Watershed Protection Department classifies surfaces that cause water to runoff into drainage systems as impervious cover. We slightly modify this definition to not include above-ground and in-ground pools, compacted soils, courtyards, golf courses, gravel/sandpits, open space, quarries, unpaved athletic fields, and paved ditches.

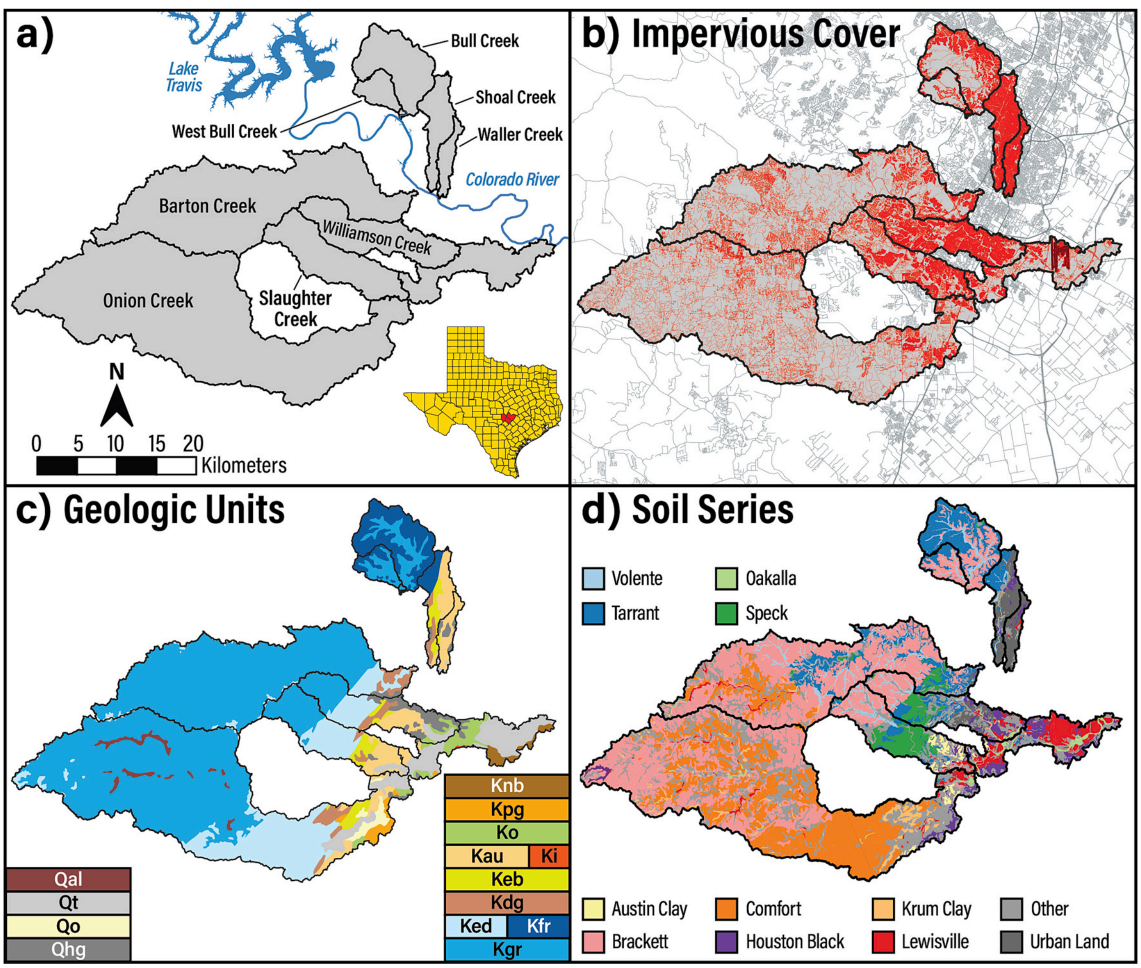

Fig. 1. (a) Location and boundaries of the eight watersheds of study in the Austin, Texas metropolitan area. (b) Impervious cover (red) (*City of Austin, Texas, Watershed Protection Department,* 2019) and main road densities outside of the eight watersheds of study (grey lines) (Texas Department of Transportation, 2023). (c) Geologic units (Stoeser et al., 2005). Quaternary units include Alluvium (Qal), Terrace deposits (Qt), Onion Creek Marl (Qo), and High gravel deposits (Qhg). Cretaceous units include the Navarro Group and Marlbrook Marl (undivided; Knb), Pecan Gap Chalk (Kpg), Ozan Formation (Ko), Austin Chalk (Kau), Igneous rocks of Austin age (Ki), Eagle Ford Formation and Buda Limestone (undivided; Keb), Del Rio Clay and Georgetown Limestone (undivided; Kdg), Edwards Limestone (Ked), Fredericksburg Group (Kfr), and Glen Rose Limestone (Kgr). (d) Soil series (SSURGO, 2019).

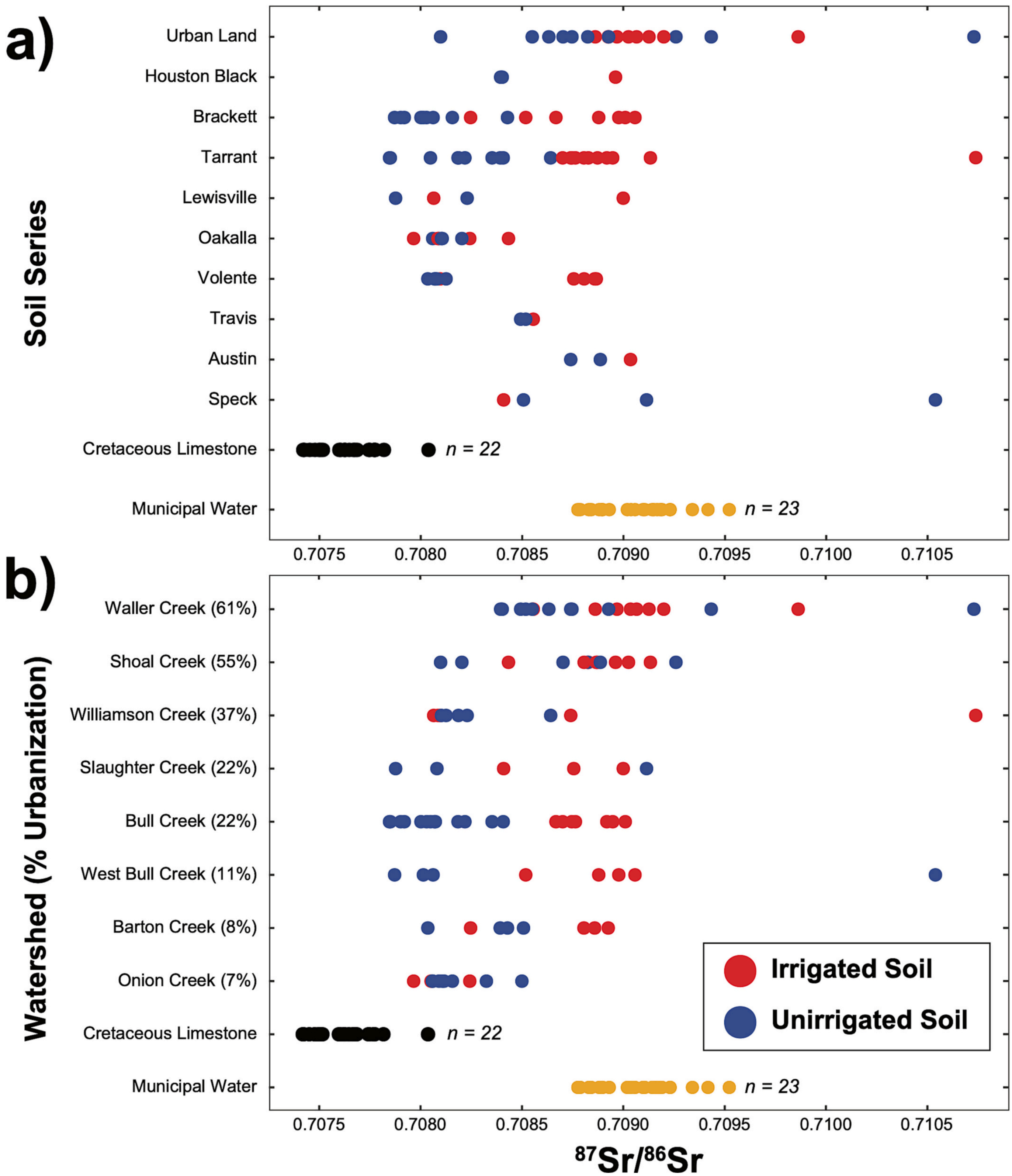

Fig. 2. Distribution of $^{87}Sr/^{86}Sr$ values for Austin-area (a) soil series, and (b) watersheds. $^{87}Sr/^{86}Sr$ values for municipal supply water (yellow circles; Beal et al., 2020; Christian et al., 2011) and Cretaceous limestone (black circles; Beal et al., 2020; Christian et al., 2011) are shown. Blue (unirrigated) and red (irrigated) circles denote soil irrigation status. Analytical uncertainty for $^{87}Sr/^{86}Sr$ analyses is smaller than the size of an individual data point.

3. Methods

3.1. Soil sampling methods

This study uses new and existing soil samples to assess differences in soil chemical and physical characteristics as a function of watershed urbanization. Soil samples ($n = 30$) were collected during the summer of 2019 from public parks, private lands, nature reserves, and residential areas within the eight Austin-area watersheds. Soil sample sites were selected based on soil series (Austin-natural soils vs. Urban Land), irrigation status (irrigated vs. unirrigated), and proximity to infrastructure (at least ~5 m from permanent buildings/structures). Site irrigation status was determined using the following criteria: 1) observed presence of irrigation systems in the field (e.g., sprinkler heads, drip irrigation valves); 2) verification of irrigation status by land managers for private lands, City of Austin Parks and Recreation personnel for public parks, and business owners for businesses and other establishments; and 3) samples were not collected nearby or downslope of residential areas where irrigation runoff could influence soil $^{87}Sr/^{86}Sr$ values.

Soil samples were collected from approximately 8–15 cm wide and 10–40 cm deep pits, dug with a stainless-steel trowel. Approximately 7–25 g of soil were collected at each site and refrigerated immediately after collection. At sites that lacked clear horizonation, a single sample was collected. One sample was collected from each horizon at sites with multiple soil horizons. Soil thickness (i.e., depth to bedrock) was measured at each site using a hammer and rebar. We compiled additional $^{87}Sr/^{86}Sr$ data from previous studies ($n = 90$; Beal et al., 2020; Christian et al., 2011; Cooke et al., 2003; Musgrove and Banner, 2004; Appendix A). We used the Soil Survey Geographic Database (SSURGO) to assign soil thicknesses and series to some samples from previous studies (SSURGO, 2019).

3.2. Silicate mineral extraction and analysis

Soil samples were analyzed to determine each sample's relative percentage of silicate, carbonate, and magnetic iron oxide minerals. Soil organic matter was removed via loss on ignition (LOI; Davies, 1974). Soil carbonate mineral content was determined following a procedure detailed in Shang and Zelazny (2008). Magnetic iron oxides were extracted from the samples using a neodymium magnet. The remaining material in each sample was assumed to be predominantly composed of silicate minerals. This assumption was verified by selecting one representative soil sample from each watershed for scanning electron microscope (SEM) analysis. Silicate mineral extraction and SEM point counts (100 points per sample) were conducted in the Keck Laboratory at Macalester College (Saint Paul, Minnesota), Department of Geology. Additional details can be found in the Supplementary Methods.

3.3. $^{87}Sr/^{86}Sr$ analysis

Organic matter and lithic fragments larger than 2 mm were removed from all samples using forceps rinsed with isopropanol. Each soil sample was homogenized using a quartering method to ensure that subsamples were representative of the bulk sample (Schumacher et al., 1990).

Soil leachate extractions followed Montañez et al. (1996). Approximately 1.5 g of each soil sample was leached with 6 mL of 0.2 M ammonium acetate (NH_4OAc; buffered to pH ~8.2) and reacted for 1 h, shaken periodically to ensure that all sample components were reacting. Samples were then centrifuged, and the supernatant (leachate) was collected. This process was repeated twice with 3 mL 0.2 M NH_4OAc. Soil leachates were acidified with 3 mL of 7 N reagent-grade nitric acid (HNO_3), dried overnight, and rehydrated with 700 μL of 3 N HNO_3 for ion exchange chemistry.

Soil leachate Sr was isolated and $^{87}Sr/^{86}Sr$ values measured following the methods of Banner and Kaufman (1994) and Musgrove and Banner (2004). $^{87}Sr/^{86}Sr$ values were measured using a Thermo Scientific Triton thermal ionization mass spectrometer (TIMS) in the Department of Geological Sciences at the University of Texas at Austin. All standard and sample analyses were corrected for instrumental fractionation and ^{87}Rb. All reported $^{87}Sr/^{86}Sr$ data have been adjusted for our measured values of the NBS-987 standard as follows. Three rounds of measurements were conducted using the TIMS. The $^{87}Sr/^{86}Sr$ value measured for the NBS-987 standard was 0.710251 for the first round, 0.710252 for the second, and 0.710254 for the third. Analytical uncertainty, based on the 2-sigma external reproducibility of the NBS-987 standard, is ±0.000008 ($n = 9$) for the first round, ±0.000011 ($n = 9$) for the second, and ±0.000013 ($n = 9$) for the third. The analytical uncertainty is ±0.000011 for reported sample $^{87}Sr/^{86}Sr$ values, based on 2-sigma for standard NBS-987 measurements for all three rounds (0.710252) and a sample replicate (difference of 0.000016; LPS5; Appendix A). All data reported here have been adjusted to an interlaboratory value of 0.710248 for the NBS-987 standard. Three procedural blanks were measured by isotope dilution to assess potential contamination from sample handling, NH_4OAc leaching, ion exchange chemistry, and filament loading procedures. The procedural blanks are <75 pg of Sr, which is insignificant relative to the estimated amount of Sr in the samples (3 μg; Christian et al., 2011). The statistical analyses and graphical visualizations were done using the open-source statistical computing software R.

4. Results

Several factors were evaluated when analyzing variations in soil $^{87}Sr/^{86}Sr$ values, including soil series, mineralogy, and thickness. $^{87}Sr/^{86}Sr$ values of all soil samples ranged between 0.7078 and 0.7107 (Table S1). The main soil series in this study, from most prevalent to least, are Urban Land, Brackett, Tarrant, Volente, Oakalla, Lewisville, Speck, Austin, Travis, and Houston Black (Fig. 1d). The non-carbonate mineralogy of eight representative samples was analyzed via SEM (Appendix A). Phyllosilicate minerals identified include montmorillonite, kaolinite, vermiculite, biotite, and illite. Non-clay silicate minerals include quartz, albite, orthoclase, anorthoclase, anorthite, and zircon. Non-silicate minerals include ilmenite, cassiterite, monazite, and xenotime. Soil thickness ranged from 15 to 74 cm (mean$n = 31$ cm) in the Austin area. Our results are organized by first introducing the natural parameters and then the anthropogenic impacts that may be responsible for variations in soil $^{87}Sr/^{86}Sr$ values.

4.1. Natural variations in Soil $^{87}Sr/^{86}Sr$

Here, we consider variations in soil leachate $^{87}Sr/^{86}Sr$ values that may be a function of several natural parameters: soil series, mineralogy, and thickness.

4.1.1. Soil series

The Austin-area soil series are categories of uniform, nationwide soil classifications based on major horizons, thickness, arrangement, and other features. We do not distinguish between the various phases within a soil series that differ in texture of the surface layer, slope, stoniness, and other characteristics that affect soil management (Werchan et al., 1974). $^{87}Sr/^{86}Sr$ values from Austin-natural soils show variable distributions across watersheds, ranging from 0.7078 to 0.7107 (Fig. 2a; Table 2). There is no clear relationship between soil series and $^{87}Sr/^{86}Sr$ value. However, the Urban Land $^{87}Sr/^{86}Sr$ distribution is elevated relative to Austin-natural soils (Fig. 2a).

4.1.2. Mineralogy

Soil silicate mineral content ranges between 26.1 wt% (Fig. 3; $^{87}Sr/^{86}Sr = 0.7082$) and 96.3 wt% (Fig. 3; $^{87}Sr/^{86}Sr = 0.7105$), with a total $^{87}Sr/^{86}Sr$ range of 0.7079–0.7107. There is a weak trend among all soils of increasing $^{87}Sr/^{86}Sr$ value with increasing silicate mineral content (Fig. 3a; $n = 31$; $R^2 = 0.32$). This trend is expected, as most silicate

Table 2
Soil $^{87}Sr/^{86}Sr$ data categorized by watershed, irrigation status, and soil series[a].

Watershed	Number of samples		Mean $^{87}Sr/^{86}Sr$ (1σ)		$^{87}Sr/^{86}Sr$ range	
	Irrigated	Unirrigated	Irrigated	Unirrigated	Irrigated	Unirrigated
Barton	4	5	0.7087 (0.0003)	0.7088 (0.0010)	0.7082–0.7089	0.7080–0.7105
Bull	8	13	0.7091 (0.0007)	0.7081 (0.0002)	0.7087–0.7107	0.7078–0.7084
West Bull	4	4	0.7089 (0.0002)	0.7080 (0.0001)	0.7085–0.7091	0.7079–0.7081
Onion	3	8	0.7081 (0.0001)	0.7082 (0.0002)	0.7080–0.7082	0.7081–0.7085
Shoal	8	6	0.7089 (0.0002)	0.7087 (0.0004)	0.7084–0.7091	0.7081–0.7093
Slaughter	3	3	0.7087 (0.0003)	0.7084 (0.0007)	0.7084–0.7090	0.7079–0.7091
Waller	8	11	0.7091 (0.0004)	0.7089 (0.0007)	0.7086–0.7099	0.7084–0.7107
Williamson	4	5	0.7082 (0.0003)	0.7083 (0.0002)	0.7081–0.7087	0.7081–0.7086
Soil Series						
Austin	1	2	0.7090	0.7088 (0.0001)	0.7090	0.7087–0.7089
Brackett	7	9	0.7088 (0.0003)	0.7080 (0.0002)	0.7082–0.7091	0.7079–0.7084
Houston Black	1	2	0.7090	0.7084 (0.000005)	0.7090	0.7084
Lewisville	2	2	0.7085 (0.0007)	0.7081 (0.0002)	0.7081–0.7090	0.7079–0.7082
Oakalla	4	5	0.7082 (0.0002)	0.7081 (0.00005)	0.7080–0.7084	0.7081–0.7082
Speck	1	3	0.7084	0.7094 (0.0010)	0.7084	0.7085–0.7105
Tarrant	11	10	0.7090 (0.0006)	0.7082 (0.0003)	0.7087–0.7107	0.7078–0.7087
Travis	1	2	0.7086	0.7085 (0.00002)	0.7086	0.7085
Urban Land	8	10	0.7091 (0.0003)	0.7090 (0.0007)	0.7089–0.7099	0.7081–0.7107
Volente	5	5	0.7087 (0.0003)	0.7081 (0.00003)	0.7081–0.7089	0.7080–0.7081

[a] Data sources include this study, Christian et al. (2011), and Beal et al. (2020).

minerals will contribute Sr with high $^{87}Sr/^{86}Sr$ values. Silicate and carbonate minerals constitute the majority of each sample. The concentration of magnetic iron oxide minerals is insignificant, with a maximum of 3.05 wt% (Fig. 3b; Table S2).

SEM point counts (100 points) on eight representative soil samples revealed distinct silicate and non-silicate mineral phases (Table 3). Phyllosilicate counts ranged from 36 to 88 points. Non-clay silicates ranged from 11 to 62 points. Counts of other mineral phases (e.g., sulfides, phosphates, etc.) ranged from 0 to 3 points. Of the samples analyzed via SEM, the two thickest samples (74 and 46 cm) had similar mineralogies to thick, relict soils within the same region (Cooke et al., 2007).

4.1.3. *Thickness*

Soil thickness varies within the Austin area, ranging between 15 and 74 cm. Among soils not extensively influenced by urban land management practices, there is a strong correlation between $^{87}Sr/^{86}Sr$ value and thickness (Fig. 4; $R^2 = 0.83$; slope $= 149.7\ \frac{meters}{^{87}Sr/^{86}Sr}$), with thicker soils having higher $^{87}Sr/^{86}Sr$ values. Soils not extensively influenced by urban land management practices exhibit the same general trend as rural, undisturbed soils from central Texas (Fig. 4; $R^2 = 0.89$; slope $= 170\ \frac{meters}{^{87}Sr/^{86}Sr}$; Cooke et al., 2003; Musgrove and Banner, 2004). Soil thickness is the only natural parameter that strongly correlates with $^{87}Sr/^{86}Sr$ value.

We consider that soils in urban areas may not preserve their original thickness due to various anthropogenic activities. To account for this complication, we evaluate soil $^{87}Sr/^{86}Sr$ values as a function of sampling distance from the base of the soil profile (Fig. S2). A weak correlation was observed between soil $^{87}Sr/^{86}Sr$ value and sampling distance from the base of the soil profile ($R^2 = 0.33$; slope $= 0.0038\ \frac{meters}{^{87}Sr/^{86}Sr}$). This weak correlation is consistent with a similarly weak correlation observed among rural, undisturbed soils from central Texas ($R^2 = 0.15$; slope $= 0.0023\ \frac{meters}{^{87}Sr/^{86}Sr}$; Appendix A; Cooke et al., 2003).

4.2. *Anthropogenic impacts on soil $^{87}Sr/^{86}Sr$*

Here, we consider variations in soil $^{87}Sr/^{86}Sr$ values that may be a function of human activity via urbanization processes.

4.2.1. *Irrigation*

Austin municipal supply water has a high and narrow distribution of $^{87}Sr/^{86}Sr$ values (0.7088–0.7095, mean = 0.7091) relative to rural stream and spring water (0.7078–0.7081; Beal et al., 2020; Christian et al., 2011). Weathering of soils with naturally high $^{87}Sr/^{86}Sr$ values have been previously hypothesized to be a potential source of Sr with high $^{87}Sr/^{86}Sr$ values (e.g., $^{87}Sr/^{86}Sr > 0.7085$) to urban stream waters in the Austin area (Christian et al., 2011). We find that $^{87}Sr/^{86}Sr$ values of soils irrigated with municipal supply water are elevated on average relative to unirrigated soils in seven of eight watersheds (Fig. 2b). Regardless of soil series, mineralogy, and thickness, most soil $^{87}Sr/^{86}Sr$ values are within a $^{87}Sr/^{86}Sr$ range bounded by limestone bedrock and municipal water (Figs. 2–4). However, irrigated soil $^{87}Sr/^{86}Sr$ values resemble $^{87}Sr/^{86}Sr$ values for Austin municipal supply water. They are generally higher than unirrigated soil $^{87}Sr/^{86}Sr$ values, which resemble $^{87}Sr/^{86}Sr$ values of the local Cretaceous limestone upon which they developed (0.7074–0.7080, mean = 0.7076; Beal et al., 2020; Christian et al., 2011). However, some unirrigated soils from densely urbanized watersheds (e.g., Waller and Shoal Creek) exhibit elevated $^{87}Sr/^{86}Sr$ values greater than Austin municipal water and irrigated soil $^{87}Sr/^{86}Sr$ values.

When only unirrigated soils are considered, the relationship between $^{87}Sr/^{86}Sr$ value and concentration of silicate minerals have a moderately strong correlation ($R^2 = 0.57$; Fig. 3a), but when only irrigated soils are considered, the correlation is much weaker ($R^2 = 0.14$; Fig. 3a). The relationship between $^{87}Sr/^{86}Sr$ value and thickness is strong among unirrigated soils ($R^2 = 0.83$; Fig. 4). However, the relationship between $^{87}Sr/^{86}Sr$ value and soil thickness is substantially weaker among irrigated soils ($R^2 = 0.004$; slope $= 21.1\ \frac{meters}{^{87}Sr/^{86}Sr}$; Fig. 4). No natural parameter in this study significantly correlates with irrigated soil $^{87}Sr/^{86}Sr$ values. Similarly, the relationship between $^{87}Sr/^{86}Sr$ value and sampling distance from the base of the soil profile is strong among unirrigated soils ($R^2 = 0.80$; Fig. S2) but substantially weaker among irrigated soils ($R^2 = 0.003$; Fig. S2).

4.2.2. *Watershed urbanization and Urban Land soils*

Soils from the densely-urbanized watersheds (≥55 % impervious cover) have elevated $^{87}Sr/^{86}Sr$ values (mean = 0.7089) relative to soils from the other watersheds (mean = 0.7084), regardless of irrigation status (Fig. 2b; Table S1). Onion Creek is the most rural watershed (7 % impervious cover) and has the lowest range of soil $^{87}Sr/^{86}Sr$ values, from

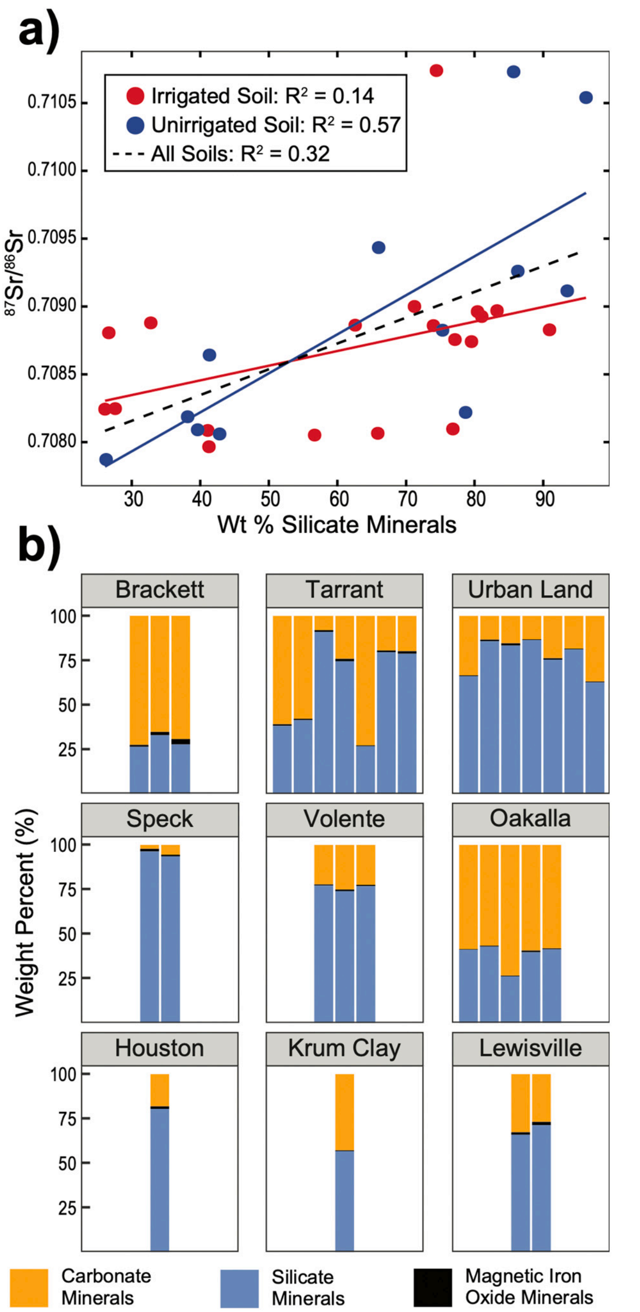

Fig. 3. (a) Co-variation of mean soil $^{87}Sr/^{86}Sr$ values and weight percentage of silicate minerals for irrigated (red) and unirrigated (blue) soils. The equations for the regression lines are $y = 0.00001x + 0.70802$ and $y = 0.00003x + 0.70707$ for irrigated and unirrigated soils, respectively. The equation for the regression line for all samples (black dashed line) is $y = 0.00002x + 0.70759$. (b) Weight percentages of the carbonate (orange), silicate (blue), and magnetic iron oxide (black) mineral phases, organized by soil series.

0.7080 to 0.7085 (Table S1). Additionally, all soil samples classified as Urban Land were collected from the densely urbanized watersheds, except for one sample from the Barton Creek watershed.

Austin-area Urban Land soils do not resemble the other Austin soil series. The $^{87}Sr/^{86}Sr$ values for irrigated and unirrigated Urban Land soils ($n = 26$) range between 0.7081 and 0.7107 (Table 2). This range is elevated and larger relative to the other Austin-area soil series (Fig. 2a). When only considering irrigated Urban Land soils, $^{87}Sr/^{86}Sr$ values cluster between 0.7089 and 0.7094, with one exception (0.7099; Fig. 2a).

5. Discussion

Our findings suggest that the origins of soil water—namely, the extent of irrigation—control much of the variability in $^{87}Sr/^{86}Sr$ values measured for Austin-area soils. However, natural factors, such as soil series, thickness, and mineralogy, may also exert some control on soil $^{87}Sr/^{86}Sr$ values in urban settings.

5.1. Mineralogic and geologic controls on soil $^{87}Sr/^{86}Sr$

One possible control on soil $^{87}Sr/^{86}Sr$ is mineralogy, as the $^{87}Sr/^{86}Sr$ of Sr released by mineral weathering will reflect the weighted average of the weathered minerals (Åberg, 1995). To determine the degree to which soil mineralogy controls soil $^{87}Sr/^{86}Sr$, we measured the carbonate and silicate mineral concentrations of all samples and identified concentrations of specific silicate mineral phases via SEM point counts. We focus on silicate minerals because they are the largest group of primary and secondary soil minerals (Karathanasis, 2006). Additionally, most soils in the region developed on lower Cretaceous marine carbonate bedrock (Fig. 1c), resulting in high carbonate mineral concentrations (Fig. 3b and Fig. S1; Table S2). The low $^{87}Sr/^{86}Sr$ values of marine carbonate rocks reflect the $^{87}Sr/^{86}Sr$ of the seawater from which they were deposited (and of subsequent diagenetic fluids) due to low concentrations of parent ^{87}Rb in most carbonates and negligible isotopic fractionation during mineral precipitation (Faure, 1991). In the Austin area, carbonate bedrock samples have $^{87}Sr/^{86}Sr$ values similar to or slightly higher than values for lower Cretaceous seawater (mean = 0.7076; Christian et al., 2011; Musgrove and Banner, 2004). On the other hand, silicate mineral $^{87}Sr/^{86}Sr$ values are generally higher than carbonate values due to high Rb/Sr ratios and time-dependent radioactive decay of ^{87}Rb to ^{87}Sr (Harrington and Herczeg, 2003).

During pedogenesis, bedrock minerals are differentially weathered and release Sr with the same $^{87}Sr/^{86}Sr$ value as the bulk mineral (Aguzzoni et al., 2019; Bullen and Kendall, 1998). Therefore, soils that develop on carbonate bedrock will likely have high concentrations of carbonate minerals and relatively low $^{87}Sr/^{86}Sr$ values. In contrast, soils with high concentrations of silicate minerals exhibit higher $^{87}Sr/^{86}Sr$ values. The soil series with the highest concentration of silicate minerals are Urban Land and Speck Soils (Figs. 1d and 3b). Speck soils have the highest concentration of silicate minerals among all samples despite forming on carbonate bedrock, with unirrigated Speck samples exhibiting higher $^{87}Sr/^{86}Sr$ values (0.7091 and 0.7105) relative to all unirrigated Austin-natural soils. The high silicate concentrations in these samples are likely due, in part, to the collection depth. They were collected from the Bt horizon, which is typically noncalcareous and clay-rich (Werchan et al., 1974). SEM analyses indicate that the silicate mineralogy of these samples is largely homogenous, with eight major unique silicate phases (Appendix A). Given that most silicate minerals, regardless of phase, will contribute Sr with high $^{87}Sr/^{86}Sr$ values (Cooke et al., 2003), and unirrigated soil $^{87}Sr/^{86}Sr$ values correlate with silicate mineral concentration ($R^2 = 0.57$; Fig. 3a), silicate mineral concentrations may partially control Austin-area soil $^{87}Sr/^{86}Sr$ values. However, irrigated soil samples exhibit a much weaker correlation ($R^2 = 0.14$; Fig. 3a), suggesting that irrigation has a stronger control than mineralogy on soil $^{87}Sr/^{86}Sr$ values—potentially overprinting original soil $^{87}Sr/^{86}Sr$ values.

5.2. Alteration of soil mineralogy via urban development

While irrigation may overprint the relationship between soil mineralogy and $^{87}Sr/^{86}Sr$, other urban land management practices can directly affect urban soils' mineralogical and chemical composition, causing variable and often negative impacts on the surrounding environment. Typically, soils overlying limestone will have elevated concentrations of calcite and expandable phyllosilicate minerals (Norra et al., 2006). In the Austin area, elevated concentrations of silicate

Table 3
SEM point count results.

Sample ID	Watershed	Soil series	$^{87}Sr/^{86}Sr$	Point counts		
				Phyllosilicates	Non-clays	Other
2S02B-080619	Barton	Volente	0.7089	36	62	2
1S34WB-070319	West Bull	Brackett	0.7079	88	11	1
2S07WL-080619	Williamson	Volente	0.7081	46	54	0
2S05O-080619	Onion	Oakalla	0.7080	72	26	2
2S04SL-080619	Slaughter	Lewisville	0.7090	57	40	3
1S40S-070319	Shoal	Urban Land	0.7088	77	22	1
1S10W-061719	Waller	Urban Land	0.7107	48	52	0
LPS5	Bull	Tarrant/Speck	0.7082	44	55	1

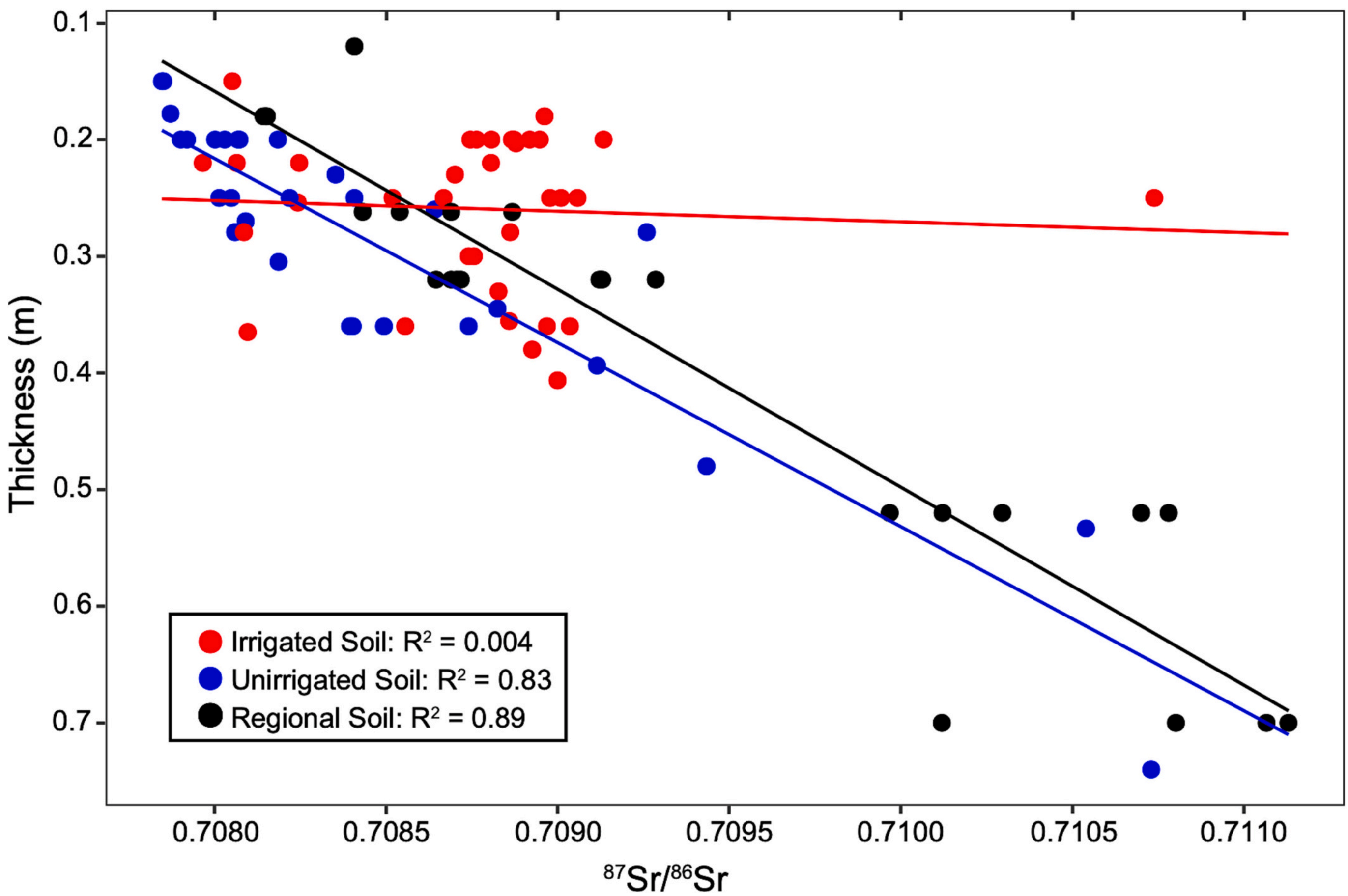

Fig. 4. Co-variation of mean $^{87}Sr/^{86}Sr$ values and soil thickness for Austin-area irrigated (red) and unirrigated (blue) soils. Note that the y-axis is inverted. The equations for the regression lines are $y = 21.1x - 14.7$ and $y = 149.7x - 105.8$ for irrigated and unirrigated soils, respectively. The equation for the regression line for the regionals soils is $y = 170.0x - 120.2$. Regional soils (black) are a compilation of data from Cooke et al. (2003) and Musgrove and Banner (2004).

minerals (including phyllosilicates; Fig. 3b; Table S2) may be due to either 1) the in situ weathering of clay-rich strata and loss of carbonate strata or 2) soil amendments introduced from sources outside the watersheds of interest. Rural soils from central Texas also have high concentrations of silicate minerals despite overlying the silicate-poor Edwards Limestone (Cooke et al., 2007). Cooke et al. (2007) attribute this discrepancy to the in situ weathering of the Del Rio Clay—a Cretaceous clay-rich marly geologic formation (Cooke et al., 2007; Fig. 1c). Provided that the Del Rio Clay and other clay-rich units comprise part of the stratigraphic section in the Austin area (Rabenhorst and Wilding, 1986), high silicate concentrations measured in some soils may be sourced from the Del Rio Clay. However, because we deliberately sampled soils that did not directly overly the Del Rio Clay or other clay/silicate-rich strata, elevated concentrations of silicate minerals found in some soils are likely due to the sourcing of soil amendments not formed on carbonate bedrock.

Urban Land soils have a higher mean concentration of silicate minerals (79 wt%) than the other Austin-area soil series (57 wt%). Since Urban Land soils are characterized as extensively altered and obscured by construction and other urban expansion efforts, the elevated concentrations of silicate minerals are most likely a result of urbanization processes. Although it is difficult to determine the exact origin of urban soils without detailed knowledge of a site's construction history, Urban Land soils may primarily be amended soils that did not originate on the same carbonate parent material but were sourced for construction purposes and/or other urban management activities (e.g., mixing, topsoil removal, filling, pulverization; Craul, 1992).

5.3. Soil thickness and $^{87}Sr/^{86}Sr$

Soil thickness is the only natural parameter strongly correlated with $^{87}Sr/^{86}Sr$ values in this study. The positive correlation was especially strong among Austin-area unirrigated soils (R^2 = 0.83; Fig. 4) and among modern and relict soils from the Edwards Plateau (R^2 = 0.89; Fig. 4; Cooke et al., 2003; Musgrove and Banner, 2004). This is likely due to the low $^{87}Sr/^{86}Sr$ value of the region's limestone bedrock (mean = 0.7076), where thin soils (~20 cm) will have a more bedrock-derived Sr (with low $^{87}Sr/^{86}Sr$ values) than thick soils (~50 cm). Thick soils will not incorporate as much bedrock-derived Sr and exhibit high $^{87}Sr/^{86}Sr$ values reflecting that of silicate minerals (Cooke et al., 2003, 2007). Plants rooted in thin soils are more likely to penetrate the limestone with their roots to access water that inherited low $^{87}Sr/^{86}Sr$ values from the bedrock. The plant-derived water becomes incorporated into the soil—effectively resetting the soil's original $^{87}Sr/^{86}Sr$ value via ion exchange processes. Additionally, bioturbation via plant roots will incorporate more carbonate minerals from the underlying bedrock into the soil (Gabet et al., 2003). However, because soils in urban areas may not preserve their original thickness due to various anthropogenic activities, we assessed soil $^{87}Sr/^{86}Sr$ variability by sampling distance from the base of the soil profile (Fig. S2). This relationship was strong among unirrigated soils (R^2 = 0.80; Fig. S2), where higher $^{87}Sr/^{86}Sr$ values were observed among soils sampled further away from the base relative to samples near the base—opposite to values observed among thick relict soils from the greater Edwards Plateau (Cooke et al., 2003). This is likely because soils sampled closer to the base have higher concentrations of bedrock-derived carbonate minerals (Appendix A).

Ultimately, both thickness and sampling distance above base strongly correlate with $^{87}Sr/^{86}Sr$ value among unirrigated soils, but almost no correlation exists when evaluating the same relationships among irrigated soils (Fig. 4 and Fig. S2). Despite the large thickness range of the irrigated soils (~15–41 cm), $^{87}Sr/^{86}Sr$ values did not increase with depth. Likewise, soil $^{87}Sr/^{86}Sr$ values did not increase further away from the base of the soil profile. Instead, most irrigated soils exhibited $^{87}Sr/^{86}Sr$ values between 0.7085 and 0.7091, the upper end of which is the same as mean Austin municipal supply water (Fig. 2). Collectively, these results indicate that irrigation with municipal supply water strongly controls soil $^{87}Sr/^{86}Sr$ values. The following sections consider the chemical processes that may drive irrigation controls.

5.4. Tracing abilities of $^{87}Sr/^{86}Sr$ and Austin-area reservoirs

The tracer utility of Sr isotopes lies in the foundation that $^{87}Sr/^{86}Sr$ values of surface water, groundwater, and natural soils are determined primarily by the isotopic characteristics of a watershed's lithology. However, mixing between reservoirs can occur via water-rock-soil interactions, and mixing Sr from two or more reservoirs with distinct $^{87}Sr/^{86}Sr$ values results in mixtures with diagnostic values between endmember reservoirs (Beal et al., 2020; Vitòria et al., 2004). In the Austin area, natural groundwater and stream water $^{87}Sr/^{86}Sr$ values (mean = 0.7079; Christian et al., 2011) approach values for Cretaceous seawater (0.7072–0.7080; NBS-987 standard = 0.71014; Koepnick et al., 1985), reflecting interactions with the region's bedrock. In contrast, Austin municipal supply water is withdrawn from the Colorado River, with distinctly higher $^{87}Sr/^{86}Sr$ values (mean = 0.7091; Christian et al., 2011) relative to Austin-area stream water. Elevated $^{87}Sr/^{86}Sr$ values in the Colorado River come from dissolved Sr from silicate-rich lithologic units upstream of Austin (Christian et al., 2011). For example, the Llano River, a tributary of the Colorado River, drains the Llano Uplift and contributes dissolved Sr with high $^{87}Sr/^{86}Sr$ values (mean $^{87}Sr/^{86}Sr$ = 0.7102; n = 2; Christian et al., 2011). The isotopic differences between the Austin-area Sr reservoirs allow for the evaluation of irrigation impacts on natural soil physiochemical characteristics as a function of urbanization.

5.5. Influence of municipal irrigation on soil

In agricultural settings, soil chemical properties evolve to resemble the properties of the water with which they are irrigated (Haliru and Japheth, 2019; Mostafazadeh-Fard et al., 2007), and our results suggest the same for soils in urban settings. Cation exchange between soil water and minerals is the dominant process controlling Sr partitioning at the soil pH (~8.7; Christian et al., 2011) observed in this study (Serne and LeGore, 1996). Exchangeable Sr inputs from precipitation may also alter soil $^{87}Sr/^{86}Sr$ values (Green et al., 2004). However, rainwater is a negligible Sr source in the Austin area, with a concentration of 6 ppb (Christian et al., 2011). Much higher concentrations of Sr are observed in Austin-area groundwaters (1.1 ppm), non-urbanized stream waters (0.28 ppm), highly urbanized stream waters (0.43 ppm), and municipal wastewaters (0.14 ppm; Christian et al., 2011; Beal et al., 2020). Municipal supply water Sr concentrations are within the same order of magnitude as the anthropogenic and natural reservoirs (0.14 ppm; Christian et al., 2011), with a substantially higher mean $^{87}Sr/^{86}Sr$ value. As such, we expect that prolonged irrigation with municipal supply water alters natural soil $^{87}Sr/^{86}Sr$ values.

Our findings, and those from Beal et al. (2020) in the Bull Creek watershed, suggest that cation exchange with municipal water controls the $^{87}Sr/^{86}Sr$ values in irrigated soils across soil series and watersheds (Fig. 2). In addition to ion exchange processes, municipal water can evaporate and deposit soluble salts in soils (containing Sr with municipal water $^{87}Sr/^{86}Sr$ values), which can be incorporated in soil leachates during sample treatment (Robinson et al., 2017). However, the high solubility of salts and the mobility of Sr in soil water will likely result in the minimal incorporation of salt-derived Sr into the soil leachates (Borg and Banner, 1996). In watersheds exhibiting ≤37 % urbanization, unirrigated soil $^{87}Sr/^{86}Sr$ values are generally low relative to irrigated soils (Fig. 2b), indicating that irrigation resets soil $^{87}Sr/^{86}Sr$ values toward values for municipal water with time. Irrigated soils mostly fall within the municipal water $^{87}Sr/^{86}Sr$ range in the densely-urbanized watersheds, but there is greater $^{87}Sr/^{86}Sr$ variability among unirrigated soils. Additionally, some unirrigated soils have unusually high $^{87}Sr/^{86}Sr$ values, even among irrigated soils of the same soil series and watershed (Fig. 2b; Table 2). Ultimately, while extensive irrigation with municipal water primarily controls soil $^{87}Sr/^{86}Sr$ values, additional factors appear to influence soil $^{87}Sr/^{86}Sr$ values in densely-urbanized watersheds.

In the Austin area, bedrock-derived Ca-rich minerals are likely preferentially weathered, rather than the less-abundant K- and Rb-rich minerals, which contribute more radiogenic Sr with higher $^{87}Sr/^{86}Sr$ values. Thus, high soil Ca and Sr concentrations that are typical in the Austin area (Christian et al., 2011; Werchan et al., 1974) likely increase the ability of Ca (and Sr) to compete against Rb and K for exchange sites, resulting in lower $^{87}Sr/^{86}Sr$ values similar to those of the carbonate bedrock (Oi et al., 1992; Werchan et al., 1974). However, it is important to note that unirrigated soil $^{87}Sr/^{86}Sr$ values are not identical to bedrock $^{87}Sr/^{86}Sr$ values, as Sr with elevated $^{87}Sr/^{86}Sr$ values may be contributed by differentially weathered Ca-poor minerals and external Sr sources (e.g., dust; Borg and Banner, 1996).

The natural variability observed among soil $^{87}Sr/^{86}Sr$ values (Fig. 2b; Table S1) was previously hypothesized to account for elevated $^{87}Sr/^{86}Sr$ values of Austin-area urban streams (0.7077–0.7095; Beal et al., 2020). However, the elevated $^{87}Sr/^{86}Sr$ values of irrigated soils relative to unirrigated soils, and the similarly high $^{87}Sr/^{86}Sr$ range of irrigated soils and municipal water, support our inference that soils evolve toward municipal water $^{87}Sr/^{86}Sr$ values via ion exchange processes as a consequence of extensive irrigation. As such, the hypothesis for the origin of the high $^{87}Sr/^{86}Sr$ values of Austin-area streams—that the natural variability of soils may elevate urban stream $^{87}Sr/^{86}Sr$ values—is not supported. Irrigated soils most likely acquire elevated $^{87}Sr/^{86}Sr$ values via irrigation with municipal water and thus would not naturally contribute the requisite values for explaining elevated stream water $^{87}Sr/^{86}Sr$ values. Instead, our results support the hypothesis that municipal supply water can alter urban stream water $^{87}Sr/^{86}Sr$ values, as proposed by Christian et al. (2011) and Beal et al. (2020). Therefore, high urban stream water $^{87}Sr/^{86}Sr$ values (e.g., Waller Creek: 0.7082–0.7092) relative to rural stream water (e.g., Onion Creek: 0.7079–0.7081) indicate that leakage from failing water infrastructure and/or irrigation runoff is likely an important contribution to urban stream waters.

5.6. Temporal constraints on the geochemical evolution of soils via irrigation

Time is a crucial factor when evaluating irrigated soil $^{87}Sr/^{86}Sr$ values. Irrigated soil $^{87}Sr/^{86}Sr$ values in densely-urbanized watersheds are within the range of municipal supply water but are more variable in less urbanized watersheds (Fig. 2b). This is likely because watersheds exhibiting higher degrees of urbanization are more likely to have older and more extensive irrigation systems in their neighborhoods, business fronts, parks, and green spaces (Gooch, 2009). Given a longer history of irrigation (and thus a larger volume of municipal water) within a soil, labile Sr will be more extensively exchanged, and the $^{87}Sr/^{86}Sr$ value of the soil leachate will reflect that of the water (Åberg, 1995; Capo et al., 1998).

Urban development has only recently begun over the last several decades in Onion Creek watershed relative to the more urbanized watersheds. Therefore, irrigation in Onion Creek is not as extensive or long-term, resulting in low $^{87}Sr/^{86}Sr$ values among irrigated soils from Onion Creek, even when compared to unirrigated soils (Fig. 2b). This is likely because the overall duration of irrigation is short, so there has not been sufficient time for ion exchange processes to reset soil $^{87}Sr/^{86}Sr$ values. Ultimately, soils from densely-urbanized watersheds are more likely to experience longer-term irrigation compared to soils from more rural watersheds, leading to higher $^{87}Sr/^{86}Sr$ values observed among soils from Waller and Shoal Creek watersheds compared to soils from Onion and Barton Creek watersheds (Fig. 2b).

5.7. Additional sources of Sr to soils in urban landscapes

Here, we consider several other processes and Sr sources that can impact soil $^{87}Sr/^{86}Sr$ values in urban areas. Specifically, we consider the $^{87}Sr/^{86}Sr$ values of Urban Land soils, amendments, fertilizers, and other building/infrastructure materials.

5.7.1. Urban Land soils

In densely-urbanized watersheds, the most common soil series is Urban Land soil (Fig. 1d)—defined as altered and/or obscured by the construction and installment of buildings, roads, and other infrastructure (Werchan et al., 1974). Considering that the range of Urban Land $^{87}Sr/^{86}Sr$ values is distinctly higher relative to the distribution of Austin-natural soils (Fig. 2a), elevated $^{87}Sr/^{86}Sr$ values of unirrigated soils from densely-urbanized watersheds (Fig. 2b) are due to the prevalence of Urban Land soils. In addition, Urban Land soils have a higher mean $^{87}Sr/^{86}Sr$ value (0.7091), likely because most Urban Land samples were amended (amendments identified in the field through spatial relationships with surrounding soil) and, in many cases, subsequently irrigated. Ultimately, unirrigated Urban Land soils have distinctly higher $^{87}Sr/^{86}Sr$ values and higher concentrations of silicate minerals than unirrigated Austin-natural soils, indicating that they did not develop (or extensively develop) on the same parent material and/or are extensively altered beyond isotopic recognition by urban development.

5.7.2. Soil amendments

Most soils in the Austin area are relatively thin (~15–20 cm) and do not exhibit pronounced horizonation between soil layers. However, some locations exhibited abrupt changes in urban soil profiles, which indicate soil amendments (Craul, 1985). It is common to supplement urban soils with various amendments, including imported soil, fertilizer, compost, mulch, and lime (Pavao-Zuckerman and Pouyat, 2017). However, anthropogenic soil materials are also commonly added during urban development, including construction materials, waste products, earthy materials from industrial activities, building rubble, and human artifacts (Effland and Pouyat, 1997). Additionally, significant urban expansion efforts in Austin include new home construction, where construction companies for new housing developments implement topsoil amendments (Werchan et al., 1974).

Three soil samples from two sites had distinct upper and lower soil layers (Appendix A). Two were classified as Tarrant soils and one as Volente soil, with irrigation systems at both sites. The upper layers were organic-rich compared to the lower layers and had a noticeable layer from the application of sod. The lower layers were much lighter in color, less organic-rich, sandier, and rockier. These changes between upper and lower soil layers within ~15 cm of the surface were only observed in residential/commercial areas. The sharp transitions and layer truncation suggest an anthropogenic origin for the upper layer. The largest difference in $^{87}Sr/^{86}Sr$ between upper (0.708807) and lower (0.708867) soil layers occurred in the Volente soil with a difference of 0.000060. The upper and lower layers in the two Tarrant soils had essentially negligible differences of 0.000029 and 0.000019 relative to the analytical uncertainty (Appendix A). The differences between the upper and lower layers at the sample sites are small and support our conclusion that differences in soil types/origins between layers do not substantially affect $^{87}Sr/^{86}Sr$ values. Rather, irrigation with municipal water at these sites primarily controls soil $^{87}Sr/^{86}Sr$ values.

5.7.3. Soil fertilization

The application of fertilizers can impact soil $^{87}Sr/^{86}Sr$ values. Some soil samples were obtained from areas that may use fertilizer, including public parks, private lands, residential areas, and golf courses. Sr in fertilizers can be associated with sulfate, phosphate, carbonate, or potassium-bearing minerals used during manufacturing. The University of Texas (UT) at Austin uses Dr. Grow™ fertilizer, procured from Justin Seed Company, with an NPK rating (percentages of nitrogen, phosphorus, and potassium) of 23–3-5 and $^{87}Sr/^{86}Sr$ value of 0.70839 (Christian et al., 2011). Several soil samples were collected on UT Austin property (Appendix A), and fertilizer application may have influenced their $^{87}Sr/^{86}Sr$ values. Outside of UT Austin, various sources of Sr in commercial fertilizers lead to a large range of $^{87}Sr/^{86}Sr$ values (0.70335–0.71522; Vitòria et al., 2004). Fertilizer use in the site where sample 1S42BL was collected may explain the unusually high $^{87}Sr/^{86}Sr$ value of soil from this location (0.7107; Appendix A). 1S42BL is from the Bull Creek watershed and is an irrigated Tarrant soil, so an $^{87}Sr/^{86}Sr$ value within the municipal supply water range was expected. However, this sample was collected from Great Hills Country Club, where the soils are extensively fertilized with an assortment of fertilizers with different NPK values (personal communication, Great Hills Country Club Golf Course Superintendent).

5.7.4. Additional urban Sr sources

Several additional anthropogenic sources of Sr may cause soil $^{87}Sr/^{86}Sr$ values to fall within the municipal supply water $^{87}Sr/^{86}Sr$ range, including dust, concrete, asphalt, and road aggregate. These materials may be incorporated into urban soils via weathering, erosion, and/or atmospheric deposition. However, based on local isotopic constraints, these sources do not appear to be a major component of (or substantially alter) Austin-area urban soils for the following reasons: 1) Weathered concrete and asphalt have $^{87}Sr/^{86}Sr$ values that are similar to those of the local Cretaceous limestone bedrock (0.7079 and 0.7075; $n = 2$; Christian et al., 2011); 2) Aggregate commonly used in repaving roadways in Austin is quarried from volcanic/intrusive rock near Uvalde, Texas and has a $^{87}Sr/^{86}Sr$ value that is substantially lower than that of the local Cretaceous limestone bedrock (0.7035; Christian et al., 2011); 3) Atmospheric dust deposited in the Austin area contains a component of marine limestone, with values between 0.7079 and

0.7085 (Christian et al., 2011). The silicate fraction of dust contains Sr with elevated $^{87}Sr/^{86}Sr$ values (0.7123; Christian et al., 2011) but is less mobile due to lower solubilities of most silicate minerals relative to calcite (Siever, 1962); and 4) SEM point counts of representative samples from each watershed did not yield any evidence of anthropogenic materials (Table 3).

6. Implications for sustainable urban development

Our results provide a framework for understanding how the geochemical compositions of soils in the Austin metropolitan area are impacted by municipal water as a function of urbanization. Mean $^{87}Sr/^{86}Sr$ values of irrigated soils are elevated relative to unirrigated soils for seven of the eight watersheds studied (Fig. 2b). In addition, the relationship between soil thickness and $^{87}Sr/^{86}Sr$ values among unirrigated soils is consistent with that for natural soils in the region, but this relationship does not hold for irrigated soils (Fig. 4). These relationships can be accounted for by the alteration/resetting of natural soil compositions via irrigation with municipal water, which is spatially and chemically extensive in the Austin area. Lawns in arid areas, such as central Texas, require more irrigation than those in temperate regions (Pouyat et al., 2010). With projected temperature increases in the Southern Great Plains region, including Texas, of 2.4–4.7 °C by the late-21st century (USGCRP, 2018) and an anticipated 30–60 additional days per year above 37.8 °C compared to current conditions (USGCRP, 2017), the demand for irrigation is expected to rise in the coming decades.

Expected future irrigation demands, coupled with rapid urban expansion and development in central Texas, may have a detrimental effect on urban soils and their capacity to provide the necessary ecosystem services to maintain environmental health and resilience. Extensive, long-term irrigation has been shown to leach large concentrations of cations (K^+ and Na^+) and reduce soil cation exchange capacity, which can have a detrimental impact on soil fertility, nutrient retention, and capacity to protect groundwater from cation contamination (Rahman et al., 2016; Adrover et al., 2012; Mulidzi et al., 2019). Extensive irrigation can also cause bulk soil electrical conductivity to increase, hindering plant nutrient uptake and limiting plant development via nutrient leaching (Guillen-Cruz et al., 2021; Ding et al., 2018; Fernández Cirelli et al., 2009). Ultimately, extensive long-term irrigation may impact soil and vegetation health in the Austin area.

The novel isotopic approach of assessing soil alteration in response to municipal irrigation presented in this study can be applied in areas (with varying degrees of urban development) where the $^{87}Sr/^{86}Sr$ values of municipal water are distinct from other local Sr reservoirs. For example, the municipal water supply for St. Louis, MO (0.7090 ± 0.0001; Hasenmueller and Banner, 2020) is isotopically distinct from the Paleozoic marine carbonate rocks underlying the region ($^{87}Sr/^{86}Sr$ = 0.7085; Beal et al., 2020). Many urban centers are expected to exhibit isotopic distinctions between watershed geology and municipal water, as observed across the U.S. (Chesson et al., 2012). Our approach cannot be applied in regions where Sr isotopic distinctions do not exist. For example, the $^{87}Sr/^{86}Sr$ signature of municipal water was similar to the signature of the bedrock across multiple provinces in South Korea (Shin et al., 2022). In areas where the isotopic distinction between various Sr reservoirs is small or non-diagnostic, one can employ a multi-proxy approach with other tracers of urbanization, including heavy metals, PAHs, and microplastics.

7. Conclusions

This study examines the role of soils in urban stream water geochemical evolution, assesses the chemical influence of municipal water, and evaluates the impacts of urbanization on soils by using the tracing capabilities of Sr isotopes in eight Austin, TX watersheds spanning a wide range of urbanization (7–61 % impervious cover). We compare soils by their natural (e.g., mineralogy, thickness, soil type, watershed) and anthropogenic (e.g., extent of irrigation, soil amendments, fertilization) parameters. Among the natural parameters examined, a strong positive correlation between soil thickness and $^{87}Sr/^{86}Sr$ value was observed in the Austin area and other regional studies in central Texas and is attributed to the influence of the region's Cretaceous marine limestone bedrock ($^{87}Sr/^{86}Sr$ = 0.7078), where thin soils (~20 cm) likely inherit more bedrock-derived Sr (with low $^{87}Sr/^{86}Sr$) compared to thick soils (~50 cm). However, the strong relationship between soil thickness and $^{87}Sr/^{86}Sr$ value is only observed among unirrigated soils, while mean $^{87}Sr/^{86}Sr$ values of irrigated soils are systematically shifted toward and within the $^{87}Sr/^{86}Sr$ range of municipal water for seven of the eight watersheds (Fig. 2b). These results contribute to our understanding of the chemical influence of municipal water on soils, indicating that soils evolve toward municipal water $^{87}Sr/^{86}Sr$ values via ion exchange processes as a consequence of extensive irrigation in urban settings. These results further suggest that measured increases in urban stream water $^{87}Sr/^{86}Sr$ values relative to rural stream water can be explained by municipal water inputs from irrigation runoff and/or failing water infrastructure rather than from stream water interactions with soils with high $^{87}Sr/^{86}Sr$ values.

Our results demonstrate that in urban systems, Sr isotopes can serve as an environmental tracer to assess the overprint of urbanization on soil physiochemical characteristics. The resetting of natural soil compositions in central Texas via urbanization is both spatially and chemically extensive, and the continued expansion of urbanization may limit the ability of soils to provide the necessary ecosystem services to maintain environmental health and resilience. The findings of this study suggest that urban communities should examine and better maintain their water infrastructure to minimize leakage and water loss via irrigation.

CRediT authorship contribution statement

Alessandro A. Mauceri: Conceptualization, Formal analysis, Data curation, Visualization, Writing – original draft, Writing – review & editing. **Jay L. Banner:** Funding acquisition, Project administration, Supervision, Conceptualization, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The data used for the research described in the article is in Appendix A.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2023.166928.

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