Developing the scientific framework for urban geochemistry


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Abstract

Urban geochemistry is a unique discipline that is distinguished from general geochemistry by the complex infrastructure and intense human activities associated with concentrated population centers. As stated by Thornton (1991) “This subject is concerned with the complex interactions and relationships between chemical elements and their compounds in the urban environment, the influence of past and present human and industrial activities on these, and the impacts or effects of geochemical parameters in urban areas on plant, animal and human health.” Urban areas present special challenges to geochemists attempting to understand geochemical states and fluxes. On the 5–6 of August, 2014, the first meeting of the reorganized Urban Geochemistry Working Group of the International Association of GeoChemistry (IAGC) was held in Columbus, Ohio, United States. Two goals of the meeting were to develop the overall scope, and a general definition of urban geochemistry. Five grand themes were developed: 1) recognizing the urban geochemical signature; 2) recognizing the legacy of altered hydrologic and geochemical cycles in urban environments; 3) measuring the urban geochemical signature; 4) understanding the urban influence on geochemical cycles from the continuous development and erosion of physical infrastructure and episodic perturbations; and 5) relating urban geochemistry to human and environmental health and policy. After synthesizing the discussion of these themes we offer the following perspective on the science of urban geochemistry building on the work of Thornton (1991): Urban geochemistry as a scientific discipline provides valuable information on the chemical composition of environments that support large populations and are critical to human health and well-being. Research into urban geochemistry seeks to 1) elucidate and quantify the sources, transport, transformations, and fate of chemicals in the urban environment, 2) recognize the spatial and temporal (including legacies) variability in these processes, and 3) integrate urban studies into global perspectives on climate change, biogeochemical cycles, and human and ecosystem health. We hope that this discussion will encourage...
other geochemists to engage in challenges unique to urban systems, as well as provide a framework for the future of urban geochemistry research.
anthropogenic sources that alter the composition of air, soil, and water within the urban system.

Much of the work in urban geochemistry has focused on nutrients that effect urban ecosystems such as C, N, or P (Bettez and Groffman, 2012; Duan et al., 2012; Divers et al., 2013; Janke et al., 2014; Kaushal et al., 2014; Newcomer Johnson et al., 2014; Potter et al., 2014), on compounds that affect human health through acute or chronic exposure such as trace metals (Pouyat et al., 2008; Ajmone-Marsan and Biasioli, 2010; Gallagher et al., 2011; Bain et al., 2012), and organic solvents and compounds (Kannan et al., 2005; Augusto et al., 2011; Tabor et al., 2014; Witter et al., 2014).

It has long been known that the land uses associated with urban growth may leave long-lived contamination that is only revealed at a later date (Colten, 1990, 1994). This legacy contamination needs to be documented in order to know how to neutralize or eliminate human and ecosystem exposures to deleterious substances (see Johnson et al. (2011) for a series of excellent case studies on the geochemistry of urban systems). Thus, in addition to anticipating future geochemical impacts, recognizing both the past and present human geochemical influence on the environment is an important aspect of urban geochemical studies.

Urban geochemistry is not the purview of geochemists alone, but rather, it is a truly interdisciplinary and cross-disciplinary subject. In addition, by measuring and understanding the sources, transport/transformations, and fate of chemicals, urban geochemistry is an important component of the concept of urban metabolism. Urban metabolism attempts to model the flows of mass and energy through urban settings at multiple scales and has a rich history of research (e.g., Wolman, 1965; Kaye et al., 2006; Chen, 2015).

Understanding and solving the present and future biogeochemical problems facing urbanized areas requires collaborations among geochemists, ecologists, engineers, health scientists, atmospheric scientists, city planners, microbiologists, hydrologists, soil scientists, urban gardeners, geographers, and political and social scientists. On the 5–6 of August, 2014, the first meeting of the reorganized Urban Geochemistry Working Group of the International Association of GeoChemistry (IAGC) was held in Columbus, Ohio, United States (Gardner et al., 2014). The goal of the meeting was to define key themes to be advanced in the future of urban geochemical research. From these themes, we attempted to develop a general definition of urban geochemistry, set objectives for the Working Group, and produce a white paper summarizing these results. This manuscript represents one of the products of that meeting.

This paper is intended to serve as a collective opinion piece on the status of urban geochemistry as a research priority. As such, this work builds upon, but does not review in detail, all of the past significant accomplishments by numerous international researchers (e.g., Bowen, 1966; Settle and Patterson, 1980; Thornton, 1990, 1992; Mielke, 1994; Chaney and Ryan, 1994; Mielke, 1999). These early studies were the building blocks upon which future work was based, and were very important in informing policy in the 1980s and beyond. Here, we review some of the major issues in urban geochemistry and propose five themes to help shape the future urban geochemical investigations: 1) recognizing the urban geochemical signatures and dynamics; 2) recognizing the legacy of altered hydrologic and geochemical cycles in urban environments; 3) measuring the urban geochemical signature; 4) understanding the urban influence on geochemical cycles from the continuous development and erosion of physical infrastructure and episodic perturbations; and 5) relating urban geochemistry to human and environmental health and policy. While these are certainly not the only important biogeochemical issues facing urban centers in the future, the working group clearly agreed that these are important themes. We hope that this discussion will aid in encouraging other geochemists to become engaged in urban problems, as well as help move the agenda of urban geochemistry forward.

2. Recognizing the urban geochemical signatures and dynamics

Urban areas likely possess geochemical signatures that are distinct from not only the natural environment, but also from other urban areas due to unique development histories. The term geochemical signatures is used here in a broad sense, which includes those sourced synthetically (e.g., PCBs), geogenically (e.g., Pb), and biogenically (e.g., C). We contend that there are commonalities amongst all cities that form an urban baseline and can guide the understanding of urban areas. Research should be geared towards establishing both the unifying properties of urban geochemistry and the unique aspects that define a given city or city district. Below, we outline major components of the built environment and human activities that contribute to the urban geochemical signature.

The geochemical signature of an urban area is not only influenced by the disturbance of the landscape, but also by the many non-local materials introduced during the development and erosion of urban infrastructure.

Geologic materials such as limestone, gyspum, rock salt, and gravel to boulder-size pieces of various rock types are used in the construction of roads, buildings, retaining walls, and other structures. This infrastructure is largely composed of reworked earth materials and is dominated by silicates, carbonates, and ore minerals (Gopi, 2010; Cevik, 2011). Concrete, one of the most ubiquitous building materials on earth, is composed of Si-rich crushed rock and Ca-rich cement binder mixed with various additives, including fly ash and natural impurities such as uranium. Aging infrastructure may be a major source of dissolved weathering products to urban streams (Kaushal et al., 2014). For example, dissolution of cement and leaching from lime may increase concentrations of calcium and bicarbonate in urban streams and sediments (Barnes and Raymond, 2009; Bain et al., 2012). Infrastructure can also include silicate-based materials such as brick, stone, and roofing tiles, and other calcium-rich carbonates such as limestone and marble. Pure metals or mixtures of metals such as Cu, Pb, and galvanized steel (Zn) are used in pipes, wiring, and roofing material. Aluminum and steel (an iron-carbon alloy) are common metals used in construction and are primary components of vehicles and many plumbing systems. Numerous other metals are used for specific functions (e.g., Cu wiring, pipes, and protective coatings) or are trace constituents of building materials (Davis et al., 2001). In addition to inorganic compounds, urban areas contain abundant organic polymers in the form of timber and petrochemical products (e.g., asphalt and plastics). Finally, the introduction of nano-materials of inorganic and organic composition in a myriad of industrial and commercial applications results in a potential concentration of these materials in the air, soil, and water of densely populated areas. Describing geochemical sources and processes in this context often requires a multi element approach, including isotopic analysis to identify and distinguish urban geochemical signatures from local geological contributions (e.g., Conner et al., 2014; Vystavna et al., 2012; Jiang, 2012; Chetelat et al., 2009; Bottrell et al., 2008). Quantification of the transport and transformations of chemicals through mass balance or source apportionment modeling, for example, is also required (Kaye et al., 2006; Vystavna et al., 2013; Teixeira et al., 2015). Here we discuss some of the processes and activities that may produce an urban geochemical signature: corrosion of infrastructure, fossil fuel combustion in a concentrated area, stormwater runoff, industrial waste streams, and treated sewage outputs.
The release of compounds from urban infrastructure alters the chemistry of the surrounding environment and affects the quality of air, soil, and water. Corrosion of metallic objects and leaching from building materials enrich urban environments in metals that may pose a risk to human health. For example, Zn and Pb are readily leached from materials present in building exteriors (e.g., brick, lead carbonate paint, concrete, galvanized metal) (Davis et al., 2001). Metal contamination is especially pronounced near roadways which receive contaminants from vehicular exhaust (Pb, Mn), brake emissions (Cu), tire wear (Zn, Cd), corrosion of welded metal plating (V, Ce, Ni, Cr) and possible combustion of lubrication oils (Cd, Cu, V, Zn, Mo) (Ward, 1990; Lytle et al., 1995; Davis et al., 2001; Ressler et al., 2000; Yesilonis et al., 2008; Rauch and Pacyna, 2009).

The input to the environment of platinum group elements (e.g., Pt, Pd, Rh) associated with catalytic converters is becoming of increasing interest (Cinti et al., 2002; Dubiella-Jackowska and Namešník, 2009; Neira et al., 2015). Some common elemental and organic compounds emanating from urban areas and their potential sources are summarized in Table 1. Although this list is thorough, it does not include all possible sources of urban-derived elements and compounds.

An increasing number of studies have found that alkalinity, and often Ca concentrations are elevated in urban streams, including locations with little to no carbonate bedrock present (Prowse, 1987; Lewis et al., 2007; Rose, 2007; Barnes and Raymond, 2009; Peters, 2009; Kaushal et al., 2013, 2014; Connor et al., 2014; Halstead et al., 2014; Stets et al., 2014). In the absence of carbonate bedrock, concrete may be a major contributor of Ca and alkalinity to urban streams (Wright et al., 2011; Tippler et al., 2014; Kaushal et al., 2014). High Ca concentrations found in riparian sediments, likely due to included concrete, represent a pool for future Ca and alkalinity contributions to urban streams (Bain et al., 2012). Elevated Ca concentrations and alkalinity might also result from enhanced chemical weathering of urban soils (Prowse, 1987), perhaps due to elevated inputs of fossil fuel-produced sulfuric and nitric acids, higher CO2 concentrations in urban areas, or the oxidation of NH4 fertilizers on urban lawns. The accelerated weathering of infrastructure has helped lead to “urban karst” development (Kaushal et al., 2014). Concentrations of Si are also relatively high in some urban watersheds (Peters, 2009; Carey and Fulweiler, 2012), though fewer Si data exist. Effects of these major ions and Si concentrations on stream ecosystems are unclear though likely to be deleterious in many cases.

The dominant source affecting the geochemistry of urban air is fossil fuel combustion. Both stationary point sources (e.g., industrial smoke stacks) and mobile non-point sources (e.g., vehicular

### Table 1

<table>
<thead>
<tr>
<th>Constituent</th>
<th>Sources</th>
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<tr>
<td><strong>Major and minor elements</strong></td>
<td></td>
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<tr>
<td>Na</td>
<td>Road salt; wastewater treatment plants. Kaushal et al. (2005). Steele and Artkenhead-Peterson (2011); Moore et al. (2013)</td>
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<tr>
<td>Ca</td>
<td>Building materials, e.g. concrete, cement, drywall, carbonate stone. Cevik et al. (2011); Wright et al. (2011); Tippler et al. (2014)</td>
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<tr>
<td>Al</td>
<td>Building siding; automobiles; coagulant for treating drinking water. Bray (2015); Pikaar et al. (2014)</td>
</tr>
<tr>
<td>Si</td>
<td>Silicate building materials, e.g. clay, sandstone. Cevik et al. (2011)</td>
</tr>
<tr>
<td>Fe</td>
<td>Steel used in building construction; automobile parts.</td>
</tr>
<tr>
<td>N</td>
<td>Fossil fuel combustion; sewage; wastewater; fertilizer. Kaye et al. (2006); Divers et al. (2013); Heatwole and McCray (2007)</td>
</tr>
<tr>
<td>P</td>
<td>Fertilizer; laundry detergent; sewage; food waste. Filippelli (2008); Kalmykova et al. (2012)</td>
</tr>
<tr>
<td>S</td>
<td>Fossil fuel electrical plants; drywall; coagulant for treating drinking water. Cevik et al. (2011); Pikaar et al. (2014)</td>
</tr>
<tr>
<td>Cl</td>
<td>Road salt; wastewater treatment plants. Kaushal et al. (2005); Steele and Artkenhead-Peterson (2011); Moore et al. (2013); Corsi et al. (2015)</td>
</tr>
<tr>
<td>HCO3</td>
<td>Concrete/cement, carbonate-bearing building materials. Wright et al. (2011); Tippler et al. (2014)</td>
</tr>
<tr>
<td>As</td>
<td>Pressure-treated wood; pesticides. ATSIR (2007); Loebenstein (1994)</td>
</tr>
<tr>
<td>B</td>
<td>Fiberglass; cleaning products. Butterwick et al. (1989)</td>
</tr>
<tr>
<td>Br</td>
<td>Fire retardants; pesticides; gasoline additive. Flury and Papritz (1993)</td>
</tr>
<tr>
<td>Cd</td>
<td>Rechargeable batteries; pigments and coatings; siding; photovoltaics. Davis et al. (2001); Tolcin (2013)</td>
</tr>
<tr>
<td>Cr</td>
<td>Welded metal plating; yellow road paint (as lead chromate); wood preservative; pesticides. Kessley (2010)</td>
</tr>
<tr>
<td>Cu</td>
<td>Brake pads; building siding or roofing; electrical wiring; Cured in Place Pipe (PVC); pressure-treated wood; pesticides. Loebenstein (1994); Davis et al. (2001); Edelstein (2013); Wicke et al. (2014)</td>
</tr>
<tr>
<td>F</td>
<td>Drinking water additive</td>
</tr>
<tr>
<td>Gd</td>
<td>Magnetic Resonance Imaging (MRI) contrast agents. Telgmann et al. (2013)</td>
</tr>
<tr>
<td>Hg</td>
<td>Coal-fired power plants; refuse incinerators; compact fluorescent light bulbs; button batteries. Nriagu and Pacyna (1988); Cheng and Hu (2012)</td>
</tr>
<tr>
<td>Li</td>
<td>Pharmaceuticals (e.g., for bipolar disorder). Barber et al. (2006)</td>
</tr>
<tr>
<td>Mn</td>
<td>Vehicular exhaust; releases from power plants and steel production; refuse incineration. Nriagu and Pacyna (1988); Lytle et al. (1995)</td>
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<tr>
<td>Pb</td>
<td>Paint; leaded gasoline; pipes and soldering; automotive batteries; urban waste incineration; cement production. Davis et al. (2001); Guberian (2013); Cloquet et al. (2006); Wang et al. (2000)</td>
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<tr>
<td>Zn</td>
<td>Galvanized steel (e.g., pipes/silverts, roofing); brick; tires; Cured in Place Pipe (PVC); waste incineration; automobile exhaust. Davis et al. (2001); Wicke et al. (2014); Cloquet et al. (2006); Goldberg (2006); Tolcin (2014)</td>
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<tr>
<td>Ce, Ni, V</td>
<td>Welded metal plating. Ward (1990)</td>
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<tr>
<td>Pt group elements</td>
<td>Catalytic converters. De Vos et al. (2002)</td>
</tr>
<tr>
<td><strong>Rare Earth Elements</strong></td>
<td>Diverse technological applications, including catalysts, metal alloys, batteries, lighting, pharmaceuticals and other medical sources. Gambogi (2014); Moller et al. (2002)</td>
</tr>
<tr>
<td><strong>Organic compounds</strong></td>
<td>Industrial and domestic solvents (e.g., degreasers, dry-cleaning); pesticides (e.g. DDT); PVC for pipes, roofing, refrigerators. Amster and Ross (2001); Pankow and Cherry (1996); Tabor et al. (2014)</td>
</tr>
<tr>
<td>PCBs</td>
<td>Polychlorinated Biphenyls: dielectric fluid in transformers, hydraulic fluid, adhesives, die-casting, concrete expansion joints, and paint. Voogt and Brinkman (1989); Diamond et al. (2010)</td>
</tr>
<tr>
<td>Dioxins/Furans</td>
<td>Byproduct of industrial chemistry and combustion; Detergents. Green et al. (2004); Rapp et al. (1990)</td>
</tr>
<tr>
<td>PBDE</td>
<td>Polychlorinated diphenyl ether; flame retardants. Stapleton et al. (2008)</td>
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<tr>
<td>Benzene</td>
<td>Gasoline stations and other petroleum-related facilities; Cured in Place Pipe (PVC). Moran et al. (2005); Pankow et al. (1997)</td>
</tr>
<tr>
<td>MTBE</td>
<td>Methyl Tert-Butyl Ether: gasoline additive. Moran et al. (2005); Pankow et al. (1997); Chusala et al. (2007)</td>
</tr>
<tr>
<td>PAHs</td>
<td>Polycyclic Aromatic Hydrocarbons: fossil fuel and biomass combustion; petroleum-derived products such as creosote and asphalt; waste from manufactured gas plants. Jensen et al. (2011); Van Metre et al. (2006); Emsbo-Mattingly et al. (2006); Stout et al. (2004)</td>
</tr>
<tr>
<td>Antibiotics</td>
<td>Pharmaceuticals and personal care products; treated wastewater and sludge. Kolpin et al. (2002)</td>
</tr>
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</table>
exhaust) enrich urban air in greenhouse gases, organic particulates, and heavy metals relative to non-urban systems (Bing-Quan et al., 2001; Wong et al., 2003; Rauch and Pacyna, 2009; Herndon et al., 2011; Moore and Jacobson, 2015). Recent work has also shown the imprint of combustion on the isotopic composition of urban water vapor (Gorski et al., 2015). With the exception of Pb, stationary sources dominate the release of metals during fuel combustion (Rauch and Pacyna, 2009). Compounds that are released to the atmosphere are subsequently deposited to surface soils and retained for indefinite periods of time. Although air contaminants have decreased in parts of the world, mainly in the United States and Europe due to environmental regulation and changing industry (e.g., Nriagu, 1990), surface soils record a legacy of past deposition of contaminants (e.g., Laidlaw and Filippelli, 2008; Herndon et al., 2011; Morrison et al., 2014).

Much of the early work in urban geochemical studies began with the investigation of contaminated urban soils (e.g., Patterson, 1965; Chaney and Ryan, 1994; Thornton, 1996), which continues to be of high interest as evidence by the tens of thousands of results in literature queries. Sources of contaminants that include heavy metals (e.g., Pb, Cd, As), persistent organic pollutants (e.g., PCBs, polycyclic aromatic hydrocarbons [PAHs]), and fine particulates (e.g., PM 2.5) are similar to those listed above for air and water, and include parent material, automobile traffic, mining activities, dust, brownfields, decaying infrastructure (e.g., roads and buildings), and industrial activities (e.g., fossil fuel burning, steel/manufacturing, industrial zinc, metal plating) (Manta et al., 2002; Cinti et al., 2002; Buzcu-Guven et al., 2007; Meuser, 2010; Jensen et al., 2011; Gualdiardi et al., 2012; Almeida et al., 2015).

Lead has been particularly well studied. For example, the early work of Patterson and his colleagues clearly documented the role of leaded gasoline consumption as a significant environmental/urban contaminant (Settle and Patterson, 1980). Lead quantities in urban areas in the United States are positively correlated with city population, with soil Pb concentrations increasing towards city centers (Mielke et al., 2010). Soil Pb content in the urban environment is 2–4-fold higher than background levels (e.g., Cicchella et al., 2008; Locutura and Bellan, 2011; Ander et al., 2013) and varies with land use (Lark and Scheib, 2013). The study of Pb in soils has also demonstrated the influence of mining activities. It has been shown that this influence is not only related to the close proximity of the urban area to active such as smelting (Meuser, 2010), but also, as in the case of St. Louis and Detroit, the “legacy” of previous smelters that have contributed to high levels of Pb in the soils and lake sediment cores (Brugam et al., 2003; Vermillion et al., 2005; MDEQ, 2015).

Urban streams receive dissolved and particulate chemical loadings from runoff, sewer connections, direct discharge from other waterways, and interactions with groundwater. Urban runoff chemistry tends to be dominated by material associated with or accumulated on impervious surfaces, such as heavy metals and de-icing salt from roadways (Turer et al., 2001; Kaushal et al., 2005). Elevated Cl concentrations are found consistently in urban areas across a range of geological and climate settings (Prowse, 1987; Williams et al., 2005; Lewis et al., 2007; Rose, 2007; Barnes and Raymond, 2009; Casey et al., 2013; Dailey et al., 2014; Long et al., 2015) and can sometimes reach toxic levels (Gallagher et al., 2011; Van Metre et al., 2011; Kaushal et al., 2005). Since the most common deicing agent is halite, Na concentrations also tend to be elevated, which can negatively impact environmental health by replacing nutrient cations (e.g. Ca, Mg, K) on exchange sites in soils (Howard and Maier, 2007; Green et al., 2008).

Many metals, especially Zn, assumed to be from tire wear, have been found in urban roadway stormwater runoff at concentrations orders of magnitude higher than receiving stream waters (Gardner and Carey, 2004), sometimes exceeding permit limits (Fernandez, 2012). Because of the changes in the hydrology of the urban watersheds, a storm event’s “first flush” can profoundly change the biogeochemistry of streams with negative influences on human and ecosystem health (Gobel et al., 2007; Daley et al., 2009; Long et al., 2015). Weathered petroleum and pyrogenic waste products from industrial, domestic, and distributed infrastructure sources are ubiquitous in urban streams, primarily adsorbed to suspended particulates (Delzer et al., 1996; Foster et al., 2000). As these particulates settle out, they add to the accumulations of urban stream sediments, which reflect the history of certain recalcitrant chemical loadings over time. The relative concentrations of specific chemicals (e.g., trichloroethylene, polychlorinated biphenyl congeners) or chemical groups (e.g., PAH homologs) in sediments can be informative as to their sources and environmental fate (Morrison, 2000; Priemer and Diamond, 2002; Diamond et al., 2010; Saber et al., 2006; Cincinelli et al., 2012).

Although it is recognized that urbanization impacts stream hydrology and biogeochemistry (e.g., Wayland et al., 2003; Walsh et al., 2005; Fitzpatrick et al., 2007; McElmurry et al., 2014), the full diversity and magnitude of urban geochemical inputs to streams remain to be discovered. For example, human activities can affect water quality indirectly by reducing the ecosystem services of urban-impacted streams. Increased nutrient concentrations in urban streams are not only due to local inputs but also a result of decreased uptake by aquatic organisms. Meyer et al. (2005) found that the uptake rates of ammonium and phosphate were negatively correlated with the degree of urbanization of streams. Hope et al. (2014) identified similar findings and further suggested that the degree of piping may play a substantial role in determining the biogeochemical properties of urban streams.

Studies of the dispersion and transport of sediment contaminated with urban byproducts have shown substantial variability in trace metals due to reach-scale variation related to hydrologic and geomorphological conditions (Rhodes and Cahill, 1999). These physical differences within urban streams lead to sediment sorting and preferential contaminant accumulation that vary not just in space, but in time. Variations in stream flows and decreases in natural sediment yields due to increased impervious surface area and decreases in recharge exacerbate sediment dynamics within urban streams (Wolman, 1967).

Sewage treatment plants are typically designed to remove some, but not all human-produced compounds and suspended material from water. Examples of compounds not removed during treatment include hospital waste such as iodinated x-ray contrast media and Gd-chelates for Magnetic Resonance Imaging (MRI) (Kormos et al., 2011; Kulaksiz and Bau, 2011), pharmaceuticals such as ibuprofen and Li through the use of lithium carbonate for bipolar disorder, carbamazepine used to treat seizures (Barber et al., 2006; Veach and Bernot, 2011), estrogens (Martinovic-Weigelt et al., 2013), personal care products such as antimicrobial agents (e.g., triclosan) (Anger et al., 2013), crotamiton for itchy skin (Kuroda et al., 2012), as well as illicit drugs such as cocaine and methamphetamine (Thomas et al., 2012; Bijlsma et al., 2014). The combination of such a diverse assemblage of chemicals in urban waters presents a unique problem that challenges both traditional treatment practices as well as the application of toxicity assessment models. For example, the complex mixture of chemicals found in treated wastewater (e.g., detergent metabolites such as 4-nonylphenol) have been found to enhance the toxicity of diazinon, an insecticide (Zein et al., 2015). These compounds can also be used as markers of modern activities. For example, the Gd-chelates used as MRI contrast agents since 1988 in the United States, cause Gd anomalies in the rare earth element signatures of urban sediments relative to analogous
background lithologies (Telgmann et al., 2013; Soderberg and Hennett, 2014). Many of these compounds can be found in sewage effluent, while others may be removed more effectively by the wastewater treatment process (Bijlsma et al., 2014).

In many urban areas, large rainfall events result in direct raw sewage release to waterways from combined sewage overflow systems, dramatically increasing biological oxygen demand (BOD) in the water column and sediments and also posing immediate human health threats (Gasperi et al., 2008; Lau et al., 2002). Runoff and leaking sewer systems can also contaminate urban groundwater with various chemicals that are released to soils, streams, and sediments due to transport in the dissolved phase (Feenstra, 1997; Pankow et al., 1997; Trauth and Xanthopoulos, 1997). Leaking and aging sewer pipes may contaminate underlying aquifers (Kuroda et al., 2012; Nakada et al., 2008). For example, Nakada et al. (2008) found carbamazepine and crotamiton in an aquifer below Tokyo as well as downstream in rivers and estuaries, which also demonstrates the resistance of some pharmaceuticals and personal care products (PPCP) to degradation. The authors also concluded that PPCP concentrations in water were directly related to population size.

The determination that concentrations of trace metals, toxic organic compounds, and solvents are elevated in urban areas compared to surrounding lands (from Patterson [1965] through Fitzpatrick et al., 2007; Pouyat et al., 2008; Peters, 2009; Ajmone-Marsan and Biasiol, 2010; Bain et al., 2012) has led to changes in policy. Work on trace metals and organic compounds continues to focus on the relevance to human health (Wcisto et al., 2002; Filippelli et al., 2012), problems stemming from the legacy of industrial and other urban activities (Albanese and Cicchella, 2012), analyzing trends through time (Kannan et al., 2005; Parsons et al., 2014), establishing emerging issues around infrastructure practices change (Gallagher et al., 2011; Tabor et al., 2014), fate and transport (Davis and Birch, 2010), and tracking sources of potentially harmful compounds (Augusto et al., 2011; Witter et al., 2014).

### 3. Recognizing the legacy of altered hydrologic and geochemical cycles in urban environments

#### 3.1. Water budgets

The water budgets and hydrologic flow paths in urban systems have a large impact on the geochemical reactions and fluxes in and out of the urban landscape. The legacy effects that urban areas have on the hydrologic cycle are well known, at least in a qualitative sense, and include reduced infiltration and increased runoff (Weiske et al., 2007). However, few studies have attempted to calculate a full water budget for urban areas that includes the complexity of water inputs and outputs from engineered systems, including leaks from water supply pipes, stormwater infiltration into sewage pipes, and the addition or removal of groundwater to/from sewage pipes (e.g., Bhaskar and Welty, 2012; Birkle et al., 2013; Marsan and Biasioli, 2010; Bain et al., 2012).

**Fig. 1.** Annual hydrologic cycles for (A) Baltimore, Maryland, USA, (B) Luleå, Sweden, and (C) San Luis Potosi, Mexico. (A) Baltimore is located in the mid-Atlantic of the United States, has a metropolitan population of ~2.5 million people, has a humid, subtropical climate (Bhaskar and Welty, 2012), Bhaskar and Welty (2012) independently estimated inputs and outputs to watersheds with the result that their models do not balance exactly but also do not include errors that propagate through the models because some parameters are determined by subtraction. Their study is one of only a few that quantitatively and directly compare hydrologic cycle models for rural and urban watersheds. (B) Luleå, Sweden lies 100 km south of the Arctic Circle, has 71,000 inhabitants, and is covered by snow for 5—6 months per year with frozen precipitation representing 39% of the annual total (Semadeni-Davies and Bengtsson, 1999). Leakage from water supply pipes was not calculated for Luleå due to the lack of data. (C) San Luis Potosi, Mexico is located ~400 km northwest of Mexico City in northern Mexico in an interior basin with no drainage to the ocean, has a metropolitan population of ~1.1 million, has a semiarid climate (Martínez et al., 2011). Despite being situated in a semiarid climate, years of recharge from leaking water supply pipes has resulted in a rising water table in a shallow groundwater aquifer that occasionally floods basements and other subsurface structures. These models make clear that urbanization alters local hydrologic cycles, but that the specifics of alteration depend on the climate, geology, and infrastructure, including method of water supply. The Baltimore model contains data for individual watersheds and focuses on a somewhat different scale than the Luleå and San Luis Potosi models, which are for entire urban areas. For all three urban areas, piped water fluxes leaving urban areas nearly equal or exceed surface water fluxes. The wet climates and associated high water tables in Baltimore and Luleå result in groundwater infiltration into sewage pipes, but in semiarid San Luis Potosi, water and waste effluent from sewage pipes into the vadose zone and then downward to groundwater.
water imported from outside the watershed boundaries dominated input (86%) to urban watersheds. In urban watersheds, precipitation represents the sole input and output was estimated independently so the overall sparse data for leaking pipes and groundwater in and evapotranspiration (26%). The city of Baltimore had somewhat constituted the largest recharge source to groundwater in some surface catchment-scale water budgets found that precipitation except in the driest of years (Mitchell et al., 2003). Bengtsson, 1999) or that precipitation was the dominant source leaching from sewage pipes into the vadose zone and then down to groundwater. Even in the semiarid climate in San Luis Potosi, the leaching wastewater has resulted in a rising water table in a shallow aquifer that occasionally floods basements and other subsurface structures.

The effect of water supply and sewage infrastructure on water budgets of urban areas can vary greatly. Investigations focused on groundwater (62%) and have been done (31%) that rely on groundwater for the public supply (Birkle et al., 1998; Kim et al., 2001; Martinez et al., 2011; Vizintin et al., 2009), and have shown that leaking water supply pipes can contribute greatly to groundwater recharge. Leakage from water supply pipes and other water imported from outside the urban watershed can represent 15% to more than 50% of inputs to catchments (Lerner, 1990; Bhaskar and Welty, 2012; Grimmond and Oke, 1986; Mitchell et al., 2003). This is an expensive loop in the urban hydrological system, as natural waters are effectively recharged with costly finished water. Water supply pipes that maintain high pressures for distribution to users can leak at rates as low as 4% in newer infrastructure (Mitchell et al., 2003), but more commonly at rates of 10–40% in older infrastructure (Bhaskar and Welty, 2012; Birkle et al., 1998; Martinez et al., 2011). It has been shown that groundwater recharge sometimes does not decrease as much as expected due to increased impervious surfaces in urban areas because recharge from leaking water supply pipes can constitute 50–90% of groundwater recharge in some areas (Kim et al., 2001; Vizintin et al., 2009). In Mexico City, where groundwater has been extremely overdrawn, leaking water supply pipes somewhat alleviate the lowered water table by artificially recharging groundwater (Birkle et al., 1998). In contrast, studies focused on surface catchment-scale water budgets found that precipitation constituted the largest recharge source to groundwater in some urban areas (Bhaskar and Welty, 2012; Semădeni-Davies and Bengtsson, 1999) or that precipitation was the dominant source except in the driest of years (Mitchell et al., 2003).

Bhaskar and Welty (2012) present an excellent case study in their comparison of urban and rural water budgets at the watershed scale for Baltimore, Maryland region (Fig. 1). Each hydrologic input and output was estimated independently so the overall budget for inputs may not precisely match the total outputs. Precipitation represents the sole input to rural watersheds and the dominant input (86%) to urban watersheds. In urban watersheds, water imported from outside the watershed boundaries — leaking pipes (12%) and lawn irrigation (2%) — represented the remainder of the inputs. The only outputs from rural watersheds are evapotranspiration (67%) and stream runoff (33%); urban watershed outputs are much different, with groundwater infiltration to sewer pipes as the largest output (41%) followed by stream runoff (33%) and evapotranspiration (26%). The city of Baltimore had somewhat sparse data for leaking pipes and groundwater infiltration into sewer pipes, but most municipalities have either little data or inaccessible information, adding to the challenge of constructing urban water budgets.

### 3.2. Geochemical cycles

Legacy issues in urban areas are wide-ranging in source loadings (e.g., past industrial releases, underground storage tanks, decaying infrastructure, past use of fertilizers and pesticides), in character (e.g., organic, inorganic, radiogenic) and in time-scale (i.e., decades to millennia) (Boult et al., 2001; Yohn, 2004; Gallagher et al., 2008; Meuser, 2010; Goody et al., 2014). Legacies affect all segments of the urban environment. Sources of information for quantifying legacy issues include historical data (e.g., sources, releases, chemical compositions) from the published and gray literature, as well as studies of environmental records such as lake sediments and tree rings. This information helps to identify geochemical hot spots and can predict future impacts on the urban environment and its effluent.

To assess whether geochemical changes induced by urbanization are potentially detrimental to human and ecosystem health, the pre-settlement conditions and attributes must be established. The geology of an area—the underlying bedrock—changes little over the relatively short occupation times of humans. Thus, geologic characteristics of urban regions can often be deduced from present day geologic mapping. Given the shorter residence time, soil geochemistry is subject to change, therefore early soil maps are particularly valuable. However, scientific soil mapping in the United States was not authorized until the late 19th century (Soil Survey Division Staff, 1993), which was well after the mid-18th to early 19th century Industrial Revolution occurred and before any environmental regulations were in effect. Although early cities occupied smaller spatial areas than cities today, they were well established before comprehensive scientific soil characterization, making other types of records of geochemical history crucial to understanding trends and processes. As cities expand over time, human habitation often grows in regions where industrial and other waste products were previously placed (Colten, 1990). The urban soil geochemical mapping exercise completed by Jensen et al. (2011) stands as a very forward-looking example of what needs to be done in other urban centers. In this work, surface soils (0–2 cm) were collected in high density Oslo, Bergen, and Trondheim, Norway and analyzed for PAHs, producing detailed maps to which different emission sources could be linked.

Legacy information can also be gained from environmental records such as tree rings and sediment accumulations. Annual growth rings on trees can record the geochemical conditions in the soil pore water at the time of water uptake by roots, and are also reflective of atmospheric deposition (Ragsdale and Berish, 1988; Wallner, 1998). Phreatophytic trees can also provide information on the geochemistry of shallow groundwater (Vroblysek and Yanosky, 1990). Laser ablation ICP-MS technology has simplified this type of dendrochemistry analysis, and is particularly useful for the analysis of trace metals (Outridge et al., 1995) including certain isotopic compositions (Novak et al., 2010). Epiphytic lichens have also been useful as natural recorders of atmospheric deposition (Getty et al., 1999; Spiro et al., 2004). Sediment accumulations in urban streams, urban estuaries, lakes/ponds and even sewer systems can provide robust information on the timing of releases of both inorganic and organic chemicals, and are long-term markers of human influence on the environment (e.g., Yohn et al., 2004; James, 2013).

Historical information about the use of chemicals in urban areas provides clues about potential latent releases associated with urban infrastructure. For example, underground storage tanks for...
petroleum products (e.g., domestic heating oil, diesel), sometimes with documented leaks, are ubiquitous and can leave a residue of non-aqueous phase liquid (NAPL) that migrates to the water table and acts as a long-term source of groundwater contamination with variable degradation rates (Mayer and Hassanizadeh, 2005; USEPA, 2012). Chlorinated solvents such as tetrachloroethylene (“PERC”), commonly used in dry-cleaning and industrial applications, have a density greater than water, allowing high chlorinated solvent concentrations to sink below the water table. These solvents are a common long-term geochemical problem in areas that hosted industrial activity during the 20th century (Feenstra, 1997). Systems within sewer systems represent an important long-term source of heavy metals and POPs when resuspended and transported during high flows (Gasperi et al., 2008).

There is also a radiological legacy in urban areas from the use of radionuclides in hospitals, building products with naturally high radioactivity, and the accumulation of atmospheric deposition through the urban network of impervious surfaces. These additional radiological sources are superimposed on the underlying geology, which is the primary determinant of how much naturally occurring radioactivity will be encountered at a given location (e.g., Rn exhalation from U-rich bedrock). Urban infrastructure also plays an important role in Rn exposure by providing enclosed spaces for Rn gas to accumulate. Naturally occurring radioactive materials (NORMs) can also be elevated in certain building materials such as gypsum and red brick (Somlai et al., 2008). Transportation via aerosols will bring contamination indoors where it will be deposited on walls, furniture and skin (Tschiirsch et al., 1995). The pumping of groundwater rich in Ra-226 has significantly increased the levels of this radionuclide in surficial sediments of lakes around Tampa, FL since the 1960s (Brenner et al., 2006). Clearly, the mineralogy and lithology of the underlying geologic material also contributes to the radiochemistry of urban areas. In order to understand and differentiate between the human contributions of radioactivity to urban areas, a detailed understanding of the sub-surface geology is important.

4. Measuring the urban geochemical signature

Accurate measurements of biogeochemically relevant compounds coupled with the myriad of synthetic substances (both organic and inorganic) are needed to assess the degree to which human activities have altered “natural” processes within urban watersheds. The development of Atomic Adsorption Spectroscopy (AAS) in the 1970s through the advent of extremely sensitive mass spectroscopy and optical methods today has added to our ability to make precise and accurate measurements of various milieu in urban settings. With the advent of bench scale high precision analytical instrumentation, researchers been able to detect more types of chemical species than ever before at exceedingly low levels (Kolpin et al., 2002). Powerful mass spectrometers coupled to innovative sample interfaces (e.g., electrospray ionization, matrix assisted laser desorption) have enabled researchers to detect compounds in all types of environmental matrices. Cavity ring down spectroscopy instruments have made the analysis of common C, H, O isotopes routine, inexpensive, and fast, enabling high throughput of samples relative to more traditional isotope ratio mass spectroscopy (Gorski et al., 2015; Moore and Jacobson, 2015). The development of ultra-performance liquid chromatography (UPLC) has allowed for the identification of separate complex mixtures in a fraction of the time it took traditional liquid chromatography, and at higher resolution. While these are just a few examples, the tools are currently available to measure a myriad of substances that are of concern to ecosystems and human health from discretely collected samples. Chemical measurements in urban settings face special challenges not encountered in less disturbed environments. Alterations to the landscape by impervious surfaces, sewer lines, population density, and human structures require us to rethink when, how, and where samples are collected. For example, these changes have rerouted the water cycle by limiting precipitation infiltration and altering overland flow (e.g., “first flush” mentioned above). As a consequence, siting sampling locations and determining the number of geochemical measurements becomes challenging. The presence of impervious surfaces can also prevent access to certain locations (e.g., covered urban streams, contaminated soils from leaking storage facilities).

The above issues reveal our need to better monitor and/or take multiple samples that capture the transient nature of biogeochemical, hydrological, and environmental processes that occur in urban settings (e.g., Long et al., 2015). The urban LTER sites in the United States have done an excellent job in this regard (e.g. Collins et al., 2010; Bain et al., 2012). In addition to the direct impact that urban activities have on the urban areas themselves, they exert impacts, such as geochemical fluxes, on areas “downstream.” This so-called “urban halo” effect discussed by Diamond and Hodge (2007) has great importance in regional, hemispheric, and even global contexts. Material can be transported by both water and air from urban locales. Sediment and ice core records clearly reveal long-traveled export of fossil-fuel burning products such as Pb, Hg, NO3 and SO4 (e.g., Mayewski et al., 1990). The dispersion of these urban-produced by-products of human activities will depend on their physiochemical form and their volatility and solubility (Lyons and Harmon, 2012).

Temporal resolution at the fine scale, at the diurnal scale, and at the annual scale, are important, but depending on the problem to be solved, spatial scale measurements are equally as important. Given that urban growth occurs over years and decades, long-term data collection is also necessary. While discrete samples provide snapshots into urban processes, urban systems are highly dynamic and require an even greater level of temporal resolution. With this aim, autonomous sensors are being developed to monitor water (e.g., Yang et al., 2013) and air (e.g. Piedrahita et al., 2014) in-situ in near real-time. With increasing availability and affordability of autonomous sensors, both spatial and temporal characteristics unique to urban landscapes will become better defined. To date, many watersheds are equipped with cost-effective and robust Sondes that measure basic water quality parameters (e.g., temperature, pH, conductivity, dissolved oxygen) at programmed intervals. However, Sondes capable of measuring other non-traditional water quality parameters (e.g., nitrate), are comparatively costly and can be less reliable. Many are based on detecting changes in the optical nature of the sample matrix or use electrochemical approaches such as selective-ion electrodes. Procuring, operating, and maintaining enough Sondes to instrument an urban watershed is likely prohibitively expensive relative to their net benefit to the science. Further, the technology needed to reliably measure other relevant geochemical parameters either does not exist or is of dubious value (e.g., Sondes that use optical measurements to extrapolate other important measurements such as dissolved organic carbon or BOD). Automated water samplers (e.g., Teledyne ISCO samplers) are capable of discretely capturing samples for transient water quality parameters not easily measured by sensors. They are ideal for collecting samples containing analytes that are stable over the collection periods and are non-reactive toward the sampling materials. These samplers, however, are relatively costly and ineffective for reactive analytes. Additionally, portable air quality monitors to determine particulate matter concentration and air geochemistry are available but are expensive and can be difficult to maintain.
The U.S. Environmental Protection Agency (EPA) is making a concerted effort to expand the use and usefulness of lower-priced sensors that can be applied at the community level—indeed, there may be an opportunity to utilize crowd-sourced data from mobile technologies to greatly increase data coverage in urban areas as it relates to human health, for example, but the development and implementation of these networks is still some years away. Thus, until advances in sensor technology occur, researchers will be unable to capture many of the transient environmentally and geochemically relevant processes in urban environments.

The other issues related to high frequency data collected by Sondes, water collected by automated samplers, or data obtained by remote sensing (e.g. from satellites) includes the transmission and archiving of large sets. For Sondes that take measurements over high temporal resolution, even annual data sets will be extremely large and require dedicated servers where the raw data are organized into easily accessible metadata. Large complicated datasets may even require additional personnel to maintain, organize, and manage. Finally, issues exist with respect to deployment and location. For example, can the monitoring stations be properly ruggized to withstand storms and secured properly to prevent vandalism, a significant issue in most urban settings? The built environment poses numerous challenges to making accurate, precise, and meaningful measurements. Significant advances have been made in bench scale analytical instrumentation, but more technologal advances need to be made in Sonde/sensor development coupled with enhanced computing ability to handle the very large amount of data acquired by these technologies.

The wide accessibility of Geographic Information Systems (GIS) has simplified spatially explicit data dissemination and visual representation, and is a critical tool for analyzing land use history in urban areas (Worboys and Duckham, 2004; Plumlee et al., 2012). GIS tools continually evolve, for example a recent development is the use of web-based GIS to build thematic maps that can easily be made publicly available on the Internet (Kerski, 2013, 2014), and can help facilitate citizen science programs. Base map information, such as topography and geology, is available in the USA, for example, through the U.S. Geological Survey, including historic digitized topographic data as old as 1884 (USGS, 2014). Geospatial datasets such as groundwater pollution potential maps are available through GeoCommunity (2014), as well as through various state (USA) agency web sites in some, but not all states (Carleton College, 2014). Site-specific geochemical data, such as those collected for Environmental Impact Statements (EISs) required by the US EPA are currently being placed in a publicly accessible database (USEPA, 2014a), and historic EISs are available in many libraries through a private vendor (Proquest, 2014). Geochemistry data from US EPA-designated Superfund Sites are being placed in the Superfund Enterprise Management System (USEPA, 2014b). Both the USGS and US EPA maintain active databases for groundwater and surface water chemistry data from sites across the U.S. with data going back to 1900 (USGS, 2014b; USEPA, 2014c). However, a universal compilation of city-level data does not exist currently, and we see this as a future need.

5. Understanding the urban influence on geochemical cycles from the continuous development and erosion of physical infrastructure and episodic perturbations

5.1. Continuous development and erosion of the physical infrastructure

As the duration and intensity of development increases in an urban area, it is likely that its geochemical signature will more closely resemble that of other urban areas while becoming less like the pre-urban geochemical signature, similar to how ecosystem properties of urban areas may converge and become relatively homogeneous compared to the pre-urban environment (Griﬃn et al., 2014). Many cities have developed over time and their environmental signature reﬂects changes in technology and economy, environmental laws, and population. The development and erosion of the urban infrastructure through time has been recently described by Kaushal et al., 2014. The environmental problems in any one city are related to where the city is in its particular stage of development in these issues (Lyons and Harmon, 2012). Still, the underlying geology will always play some role in differentiating urban centers, such as where urban soils are enriched in Cr or Ni from underlying mafic or ultramafic rocks (Pouyat et al., 2008) or As because of shale components (Smith et al., 1998).

It is very likely that the geochemical ﬂuxes from portions of urban infrastructure will vary through time. For example, variation in roadway materials over time, from brick, to concrete, to asphalt has led to a change in solutes derived from these surfaces (e.g., Mahler et al., 2005). Additionally, roofing materials associated with older infrastructure (i.e., galvanized steel or copper roofs) may produce higher ﬂuxes of certain chemicals/compounds compared to their non-metal, modern counterparts (i.e., asphalt shingles, roofing tar, etc.) (Wickel et al., 2014). Decaying infrastructure along with population increase in urban regions can lead to greatly enhanced nutrient ﬂuxes (Potter et al., 2014). The long-term maintenance of infrastructure dealing with waste plays a signiﬁcant role in the urban geochemistry of many elements and compounds. Finally, the quantity and quality of fossil fuel combustion, including changes in patterns and amount of vehicular trafﬁc, has had major in-situ and also far-reaching impacts on the geochemical output from urban areas.

Estuarine and lake sediments have proven a particularly useful tool in evaluating the impact of this changing urban infrastructure over time. For example, US Geological Survey scientists have measured both hydrophobic organic compounds and metals in these milieu to determine urban/suburban ﬂuxes through time (e.g. Van Metre and Mahler, 2004, 2014). Neumann et al. (2005) used a combination of historic reservoir sediment records and river water concentrations from the Chattahoochee River in metro Atlanta, Georgia, USA collected in the 1990s to “back cast” trace metal concentrations from the 1980s into the 1920s. Modeling of the sediment record suggested that the per capita input of dissolved metals decreased in recent decades, but anthropogenic metal input to the river was still a water quality issue. A series of chemical chronologies based on sediments from inland lakes across the state of Michigan, USA revealed 1) the success of environmental regulations in reducing metal contamination (e.g., Pb), 2) the impact of chemical loadings to the environment from urban areas (e.g., PAHs), and 3) that problems remain (e.g., Hg) (Yohn et al., 2004; Kannan et al., 2005; Parsons et al., 2007, 2014). Similarly, Church et al. (2006) describe periods of large increases in industrial metals post-1950, coupled with evidence for increased eutrophication and runoff to the Delaware Estuary — a heavily urbanized coastal estuary.

Although this type of work provides important insights into the elemental cycling within urban systems over time, it only yields the total surface ﬂux from the larger watershed containing the urban area. Thus, details of the speciﬁc sources of the compounds/elements are usually not well constrained. Although urbanization leads to increased environmental patchiness and hydrological connectivity (Grimm et al., 2008), the importance of these factors in the transport of chemicals in urban aquatic systems is not well quantiﬁed, especially from an urban development standpoint. Nonetheless, the use of traditional geochemical analyses (e.g., chronology of sediment cores) provides valuable information that
can assess urban ecosystem function and directs scientists to more specific process alterations or problems. For example, the loading of hydrophobic organic contaminants observed by Van Metre and Mahler (2004) in sediment cores led to the discovery of significant PAH loadings from coal-tar sealants commonly used in urban environment (Van Metre and Mahler, 2010). These findings have led to regulatory action in some areas which have in turn resulted in improved environmental health (Van Metre and Mahler, 2014).

5.2. Episodic perturbations

Episodic perturbations affecting the urban environment can result from a wide range of natural disasters, including hurricanes and large storms, droughts, floods, tornadoes, tsunamis, mass wasting, earthquakes, volcanic eruptions and wild fires, as well as human-induced disasters such as chemical spills, arson or industrial fires, and terrorist acts. These events can have significant impacts on the distribution, state, reactivity, and potential human exposure to pre-existing contaminants in the urban environment “baseline” (Plumlee et al., 2013). In addition, disasters have the potential to cause infrastructure failure, introduce new contaminants, and even flush-out, or dilute, pre-existing contaminants.

Understanding and documenting the existing urban conditions (i.e., characterization of the geochemical environment including associated infrastructure and geochemical fluxes) is a crucial first step to identifying the effects of urban disasters and perturbations on geochemical distribution and physiochemical state. For example, several studies have documented high concentrations of metals, volatile organic compounds, and other contaminants in the floodwater and sediments that were deposited throughout New Orleans following hurricane Katrina (Presley et al., 2006; Mielke et al., 2006; Adams et al., 2007). However, the observed concentrations for many of these contaminants were in agreement with those measured in the city’s storm-water runoff and soils prior to Katrina, and were not necessarily a result of the disturbance (Pardue et al., 2005; Abel et al., 2007; Zahran et al., 2010). Some pollutants were actually found to decrease following flooding resulting in improvements to public health (Zahran et al., 2010). Clearly, a key aspect of preparedness for future episodic perturbations includes thorough documentation of the baseline conditions regarding the distribution, state and concentration level of potential urban contaminants.

Although the impacts of any given disturbance or disaster will be highly event-specific, we summarize below select, recent events that demonstrate the diversity of potential impacts on urban geochemistry that could result from future episodic perturbations, both natural and anthropogenic.

5.2.1. Tsunamis and tropical cyclones

The impacts of tsunamis and tropical cyclones on urban centers can range from the redistribution of sediments, to the loss of hundreds of thousands of lives (e.g., Sumatra tsunami of 2004), to far-reaching radiation releases (e.g., Japan tsunami of 2011). Damage to coastal infrastructure by high winds can release or mobilize building materials and hence potential contaminants from these materials in the urban environment similar to tornadoes, while flooding can bring people in contact with water-borne pathogens and increase the abundance of molds and aerosolized fungal spores (Maegle et al., 2005; Solomon et al., 2006; Rao et al., 2007; Adhikari et al., 2009). Additionally, saltwater can also be corrosive to building materials and concrete, and cause the contamination of drinking water wells (Nair et al., 2013). Salt water intrusion and contamination of municipal drinking water will be especially problematic for coastal areas given future projections of sea-level rise and increased frequency of storm surge flooding associated with tropical cyclones (Gornitz et al., 2001, 1997; Ericson et al., 2006; Woodruff et al., 2013; IPCC, 2013).

5.2.2. Flooding

River flooding can mobilize metals and other contaminants 1) within an urban area (Hanjiang River, China, Guo et al., 2014; Perm, Russia, Vodyanitskii and Savichev, 2014; West Yorkshire, UK, Old et al., 2004), 2) into an urban area from upstream (Montousse River, France, Roussiez et al., 2013), especially when mining districts are upstream (e.g., Pb in Wales, Foulds et al., 2014; Hg in California Central Valley, USA, Singer et al., 2013), and 3) out of an urban area (Gironde Estuary, France, Schafer et al., 2009). Flooding can cause oil spills and other chemical releases during electrical power outages, and these releases can occur during remediation processes (Coffeeville, Kansas, USA; Santella and Steinberg, 2011). Flooding leaves behind wet sediment, potentially exposing occupants to airborne particulates, fungi and bacteria if cleanup is not rapid (Brisbane, Australia, He et al., 2014).

In coastal cities, sea-level rise in the second half of the 21st century and beyond is expected to create a much higher recurrence of episodic flooding due to both natural tidal cycles and in association with major storm events (Gornitz et al., 1997, 2001; Woodruff et al., 2013; Soleciki et al., 2014). This is turn will have major implications for infrastructure that will affect urban biogeochemical cycles, such as wastewater, stormwater, and drinking water systems. In addition, the potential for legacy contaminants to invade coastal and estuarine systems becomes higher over the next several centuries as sea-level continues to rise and the most vulnerable urban areas (e.g. New Orleans, Miami, Bangladesh) are faced with abandonment of coastal infrastructure.

5.2.3. Drought

Drought brings other potential issues such as increases in evaporation and dust generation. Evaporation concentrates solutes, and may render the water toxic to plants (Kabul, Afghanistan; Houben et al., 2009). Rainwater-collection systems can collect contaminant-rich dry deposition from natural and anthropogenic sources; Pb is of particular concern (Brisbane, Australia; Huston et al., 2012). In addition, metal buildup in arid urban areas may become a source of contamination to nearby water-supply lakes through dust transport (Las Vegas, Nevada, USA and Lake Mead, Rosen and Van Metre, 2010). Drought increases an area’s susceptibility to dust production and fire (Finch, 2012). A 120% increase in PM2.5 aerosols from the 2011 drought in the southern USA was primarily attributed to increases in organic carbon produced from forest fires (Wang et al., 2015). Population migration into urban areas because of drought is common, and can further concentrate contamination as well as stress water supplies (Finch, 2012).

5.2.4. Tornadoes

Highly focused disturbances such as tornadoes can completely destroy an urban environment, exposing, pulverizing and disseminating building materials (e.g. asbestos) and building contents (e.g. household hazardous waste), roads and sidewalks, electrical systems including PCB-containing transformers, plumbing, liquid storage tanks, rail cars, etc. (Greensburg, Kansas, Santella and Steinberg, 2011). Intentional releases of chemicals, such as insecticides used to reduce vector-born disease, may also follow a tornado (Young et al., 2004). Although the areal footprint of a tornado is small and thus the likelihood that it will hit an urban area small, the total destruction that occurs practically guarantees release of many kinds of toxic materials.

5.2.5. Volcanic eruptions

Two of the most likely adverse effects of volcanic eruptions on
urban environments include dispersal of volcanic ash and release into the atmosphere of acidic gases including SO$_2$, HCl, HF and CO$_2$. The fine particulate size of volcanic ash can result in wide dispersal and highly respirable/inhalable particulates that can lead to lung disease (e.g. Baxter, 2000). Leaching of toxic metals and gases (e.g. fluorine) by rainwater or resulting from direct deposition or transport of ash into water bodies can also contaminate drinking water supplies for animals and humans. Volcanic gases emitted to the atmosphere can be irritants and in high enough concentrations, can be toxic, and they form acidic solutions that can leach and transport a range of soluble toxic metals (e.g. Pb, As, Cr$^{6+}$, Cu, Zn) from the built environment that can contaminate food sources and water supplies (e.g. Plumlee et al., 2013). Because many major urban centers around the world are located in close proximity to active volcanoes, these issues are of global significance.

5.2.6. Fires

Wildfires or anthropogenic-induced fires (e.g. arson or industrial trials) that occur within or near urban centers can result in wide ranging and widely dispersed toxics. Smoke and related airborne particulates cause respiratory problems, and often produce caustic alkali compounds when in contact with water and body fluids. In the built environment in particular, suspended particulate matter can be a significant source of inhalable, and ingestible toxic metals including Pb, Cr, etc., as well as organic contaminants such as PAHs (Pleil et al., 2006; Plumlee et al., 2013; Kristensen et al., 2014). In addition, burned soil can release pathogens (e.g. soil fungus) that can cause disease, including well-documented cases of Valley Fever (Plumlee et al., 2013).

5.2.7. Terrorist acts

While one may envision any number of terrorist acts that could impact the urban environment, the collapse of the World Trade Towers in the 9/11 attack is the one well documented example. Dispersed dust and debris from the collapse comprised a mixture of construction materials and derivatives, including asbestos, gypsum, glass and glass fibers, calcium hydroxide, and toxic metals such as Pb, Cr$^{6+}$, Cu, and Zn (Plumlee et al., 2013). Fires associated with the collapse also emitted organic contaminants including PAHs. The combined effects of these released compounds are now associated with adverse health effects in those who were involved in rescue and cleanup or who live in the vicinity. Although the Fukushima incident was caused by an earthquake-related tsunami, this also provides context for the possible effects of a terrorist-inspired nuclear contamination event. Dispersal of radioactive material into local water and food supplies is one immediate source of ingestion for humans and animals, and this would impact a large population in an urban environment. Widespread dispersal of radionuclides, such as $^{131}$I and $^{137}$Cs, by emissions into the atmosphere have been documented throughout North America and Europe (e.g. Masson et al., 2011; Christoudias and Lelieveld, 2013), demonstrating that such an event could significantly impact urban centers over a very large geographic area. Modeling of the potential for radiological contamination, including fallout from an improvised nuclear device, has been investigated for urban environments in efforts spearheaded at the Idaho National Lab and the US EPA (Demmer et al., 2014).

5.2.8. Earthquakes

Earthquakes with epicenters in or near urban areas, especially in underdeveloped countries and/or unprepared cities, can result in significant dust generation resulting from building collapse. Similar problems can arise as described for collapse of the World Trade Center, with potential direct impacts on humans including clogged air passages, and longer-term effects including illnesses such as pulmonary problems, as well as environmental contamination with toxic metals and compounds released from construction materials. Other related effects such as fire, damage to energy generating facilities, leaks in fossil fuel distribution or storage facilities, as well as compromised drinking water supplies can result in contamination of the environment and pose human health risks (e.g., Noji, 2014). In addition, as with fire, it has been demonstrated that earthquake-induced landslides can cause release of soil fungi; outbreaks of Valley Fever have been clearly associated with the 1994 Northridge earthquake (Jibson, 2002; Floret et al., 2006).

Predicting contamination from former or current land use is an important new effort that is aided by GIS and modeling. Schulz et al. (2009) modeled Pb, Hg and As transport during a fictitious flood event, comparing urban areas to surrounding agricultural areas; urban areas were largely unchanged in this model, but the authors suggest this could have been driven by the imposed boundary conditions. For example, the 2008 Great California Shakeout Scenario (Porter et al., 2011) simulated likely impacts of a magnitude 7.8 earthquake with an epicenter at the southern end of the San Andreas Fault, near Los Angeles. In addition to the immediate loss of life and infrastructure, a wide range of adverse effects to the environment and human health are predicted based on this disaster scenario, including potential releases of hazardous materials (e.g. chlorine and ammonia; Eguchi and Gosh, 2008) from both physical damage as well as fires that could impact petroleum refineries, wastewater treatment plants, chemical manufacturing plants and storage facilities, and other industrial complexes (Eguchi and Gosh, 2008; Plumlee et al., 2012). In the case of earthquakes, GIS analysis and modeling linking severity of shaking and extent industry and infrastructure to population distribution is key to risk and hazard assessment as well as mitigation. More of these kinds of efforts could help planners and managers in reducing human exposure to harmful urban-derived contaminants associated with other types of natural or anthropogenic disasters.

6. Relating urban geochemistry to human and environmental health and policy

Decades of research into human disease have revealed a host of environmental factors influencing human health, both negatively and positively. Cities are at the epicenter of human—environment interactions, marked by high population density, high concentrations of fixed and mobile sources of human-produced or enhanced emissions, high traffic volumes, and frequent occurrence of industrial operations co-located in proximity to human habitation. These factors have resulted in a number of clear case-study examples of negative impacts of urban geochemistry on human health. Perhaps the most pernicious and well-studied of these is Pb poisoning, where the human-produced sources and the environmental cycling of Pb have been reasonably well-constrained (Rabinowitz and Wetherill, 1972; Hamester et al., 1994), and the human health impacts have been well documented (Pirkle et al., 1998). A clear cause-effect relationship for Pb has resulted in substantial and highly effective mitigation actions, although the limitations in current practices focusing on one particular source (degrading Pb-based paints in older homes; Lanphere et al., 1998) have masked somewhat the widespread problem of highly elevated soil Pb in urban areas (Mielke and Reagan, 1998; Laidlaw and Filippelli, 2008; Zahran et al., 2013).

Other examples highlight potential links between geochemistry and human health in urban areas, including: metals and renal disease (Brup, 2003), Hg and PCBs in consumed fish and neurological impacts (Grandjean et al., 2010), and organic solvents and chemicals in soils and water and cancers (Lynge et al., 1997). For
other components that might have human health impacts, many are poorly studied and/or study has been restricted largely to either the geochemistry realm or the human health realm. An example is the role that particulate matter plays in pulmonary disease (asthma, chronic obstructive pulmonary disease), as the primary environmental drivers of pulmonary exacerbations certainly includes particulate matter (Goss et al., 2004). It remains unclear which particulate size class is a main driver, what the relationship is between particulate matter and ozone, and what implications exist for ultra-fine particulates (e.g., less than 0.1 μm) to react deep in the alveolar portion of lungs. Initial studies (e.g. Verma et al., 2014) suggest that smaller particles may be associated with higher levels of toxicity.

Several other concerns are emerging that have strong implications for urban geochemistry and health. One of these concerns health risks due to synergistic effects of exposure to inorganic and organic chemical mixtures, a long standing research topic (e.g., Safe, 1990; Abdrahamanov, 1997; Burkart and Jung, 1998; Carpenter, et al., 2002). Although a target of multiple grant calls, we know little about health impacts of environmental “mixtures” of materials commonly found in cities and the synergistic or antagonistic effects of these mixtures (e.g., Mielenke et al., 2000; Filippelli et al., 2015). In addition, urban environments have other types of stressor such as wastewater and automobile/industrial emissions that could influence the nature of these synergistic effects (e.g., Zein et al., 2015; Mauderly, 1993). Also, our relatively recent identification of detectable amounts of pharmaceuticals in urban wastewater and stream systems (Kolpin et al., 2002) is sparking research into ecological and human health implications. Nanoparticles of various compositions are now widely utilized in manufacturing of consumer products—their release into the environment and subsequent uptake mechanisms by humans is not well-understood, and the health implications of this uptake are unknown (e.g., Dale et al., 2015). Further, microplastic pollution from industrial manufacturing sources and the breakdown of larger plastic litter has been studied extensively in marine environments (Ryan et al., 2009), McCormick et al. (2014) found abundant microplastics colonized with pathogenic bacteria in an urban stream sampled downstream from a wastewater treatment plant effluent in Chicago, USA. Microplastics originate from personal care products and synthetic fibers and are not removed by wastewater treatment processes, and urban streams are likely an important source of microplastic pollution in the Great Lakes, USA (McCormick et al., 2014).

A long history of environmental regulations and consumer protections have certainly been important in reducing population-level and individual exposures to harmful pollutants, although these protections are not internationally applied, with developing countries often more heavily affected by the harmful impacts of pollution than developed countries (Cohen et al., 2005). Thus, one aspect of mitigation (minimizing new harmful exposures) has been successfully applied to active sources and is credited for reducing such harmful urban chemicals as NOx, SOx and particulates from combustion sources, Pb from gasoline, paint, and solder, and other point sources of contamination to air, water and soil. But many of these chemicals have a long environmental half-life, and the legacy of past, unregulated emissions remain a strong characteristic of the modern urban environment. Mitigating geochemical legacies in cities is a challenge not accepted, both because of tangible reasons such as cost and practicality and intangibles such as environmental injustice (Filippelli and Laidlaw, 2010). Furthermore, mitigation requires clear justification, and we simply do not yet know enough about the cycling and impacts of a number of the chemical factors listed above to have them rise above the pre-cautionary level of concern.

Human health data are measured at a scale and frequency far exceeding geochemical data, which is largely project-focused (spatially and temporally confined) rather than continuous monitoring. For example, in many urban metropolitan populations in the developed world, there is locational, demographic, and basic health data (weight, height, blood pressure, etc.) of practically every individual in that city. These data might not always be current, but in general, it is present in some form. Recent protections have ensured privacy of citizens in many countries, and this privacy extends to details about demographics and health status. While these protections protect individual rights, similar mechanisms for extracting data — even non-identifiable data — that could provide enormous insight into scientific processes and greatly improve public health are not as well developed. The same is true in developed countries where health data are compartmentalized and difficult to access due to concerns over privacy. So, although there is a wealth of information (tens of millions of pieces of data in a typical city of 1 million) available concerning the health and wellbeing of citizens, the capacity for integrating these data with environmental or other metrics is significantly limited or controlled.

On the environmental side, geochemical data are plentiful, but typically collected, archived, and analyzed for targeted research or regulatory reasons, mostly not directly related to human health. Furthermore, these studies or projects are typically focused on a small set of variables of interest, and collected from a particular medium (e.g., soil, or water, or air). Thus if one wanted to construct a master layer of heavy metals in city soils, one would have to seek out individual project-specific data sets, select for those that relate soils only, populate the layer with these various data, and likely produce a map with little spatial or analytical coherency (Fig. 2).

Collectively, the issues with human health data protection coupled with spatial and contextual limitations of geochemical data makes for very limited abilities to construct meaningful epidemiological relationships between health and the environment. Often, the best that can be done is to aggregate the health data by census tract or zip code, hope that there is adequate geochemical data in the right medium to reasonably populate the geochemical layers, and then do regression analyses between the two. The limitations are clear, and not easily overcome. However, we feel strongly that a new data platform that combines urban geochemical, epidemiological, and land use/land change data would greatly enhance our understanding of the relationship between urban habitation and health.

Managing urban environments to adequately support large populations is a challenge that must rely on a variety of disciplines to inform policy makers. Factors influencing sustainable policy decisions encompass many topics, including altered biogeochemical cycles, human health initiatives, the use of geochemical baseline measurements, geologic hazards, as well as complex socioeconomic factors (Albanese and Cicchella, 2012; Hoornweg and Freire, 2013). Every aspect of the urban environment leaves a geochemical signature in the air, water and soil, which are considered our common-pool resources and critical to sustaining urban populations (Lyons and Harmon, 2012; Wong et al., 2012). Currently, institutional efforts for managing these environmental services do not adequately incorporate urban geochemistry into an urban policy framework (or, it could be argued, science in a general sense). However, continued research and awareness are advancing the role of geochemistry by contributing to strategies aimed at improving human and ecological health in urban areas. Here we discuss those contributions and recommend an expanded role for the discipline.

The diversity of topics included in this review makes clear that interdisciplinary communication is critical — urban geochemists
can bridge some of the gaps that exist between environmental policy and health policy spheres. There have been successes in this effort, which guide the way to more comprehensively incorporating geochemical processes into policy fronts. One example relates to the Clean Air Act of 1970 and revisions thereafter, and subsequent regulations controlling the use and remediation of Pb in the environment (e.g., Settle and Patterson, 1980; Mielke, 1999; Yohn et al., 2004). Long-recognized as a human health concern (Gilfillan, 1965), reducing Pb exposure became a public health concern in part due to the groundbreaking work of Clair Patterson and others, which ultimately contributed to Pb being removed from concern in part due to the groundbreaking work of Clair Patterson and others, which ultimately contributed to Pb being removed from use in the USA (Filippelli et al., 2016). This policy resulted in a significant decline in Pb exposure and a public health success of the highest level. Continued funding went into remediation of the remaining acute sources of Pb exposure—largely, degraded Pb-based paints on older homes. These efforts also yielded benefits, but now that population Pb levels have decreased, we are seeing newer sources emerge as the greatest risks to health—namely, urban soils contaminated with legacy Pb from multiple sources (Filippelli and Laidlaw, 2010). Contaminated soils and the dust produced from them continue to create exposure problems, largely impacting urban populations of color and lower socio-economic status. A shift in awareness to soils has occurred at the same time as an explosion of interest in urban gardening, providing a unique example to use gardening as a learning platform for the cycling of Pb in cities (Clark et al., 2008). Additionally, the awareness of the dust source of Pb to children (Laidlaw et al., 2005; Zabran et al., 2013) has modified some testing protocols in health clinics, which are implementing programs to note the neighborhood and age of the child tested, and the time of year that testing has taken place to more successfully identify children at risk.

Geochemists can contribute to restoration goals by providing quantitative measurements that indicate the scale and nature of the human influence in urban areas. Baseline data that assess deviation from pre-urban reference conditions include various geochemical parameters (e.g., Hg and Pb) and can be used to guide policy as with The European Union Water Framework Directive (WFD). The WFD legislation mandates “good ecological quality” of European surface water resources by 2015 based on ecological function and as compared to historical reference conditions that are often gleaned from paleolimnological studies, since historical data are more often than not unavailable (Bennion and Battarbee, 2007). Previously mentioned case studies summarized by Johnson et al. (2011) provide good examples of baseline geochemical data for a number of cities that could be used to inform policy decisions. Moreover, geochemical records evaluate differences in urban environment signatures that can facilitate sustainable policy decisions; old v. new cities, aging infrastructure impacts, city size, transportation history/preference, etc. by comparing unique policy needs (Albanese and Cicchella, 2012; Revi et al., 2014). For example, older cities may not be designed with recent sustainable building mandates and may have vacated properties, like brownfields or residential properties, which have a different geochemical signature than newer ‘greener’ cities with better mass transportation and modern infrastructure (Thornton et al., 2008; Wong et al., 2012). Isotopic measurements can appraise energy consumption and be integrated with atmospheric carbon composition (CO₂ and δ¹³C) to further elucidate sources of contamination (fossil fuel combustion or local photosynthesis/respiration), which can be used to support mitigation policies of emissions (Pataki et al., 2007; Newman et al., 2008; Gorski et al., 2015; Moore and Jacobson, 2015).

Environmental policy and management decisions in urban ecosystems are dictated by a myriad of factors including economic health, education level of population and cultural influences that collectively impact the urban signature. Tasked with researching and understanding the dynamics of urban biogeochemical systems, geochemists can engage with a wide range of decision makers and increase societal awareness of geochemical alterations (further fueling sustainable policies) (Hoornweg and Freire, 2013; Revi et al., 2014). An emerging concept of ‘Earth System’ governance focuses on human behavior in the ‘Anthropocene’ and explores how we view our political systems (Biermann, 2007, 2014). It is valuable to consider the concentrated effects of global climate, biodiversity loss, and interdependency on ecosystem services that will continue to mount as urban populations grow (McGranahan and Satterthwaite, 2003; Satterthwaite, 2007; Revi et al., 2014). One strategy for linking geochemists with politicians is to engage urban geochemists with health researchers, stakeholders, urban planners...
and policy makers to incorporate the above mentioned geochemical resources (health impacts and baseline data) for decision making with an emphasis on the local scale of urban governance (Menegat, 2002). An overall migration of science-based policy would allow urban geochemists to more avidly participate in developing policy for sustainable urban environments in the future.

7. The path forward

The path forward in the field of urban geochemistry and for the IAGC Urban Geochemistry Working Group involves understanding both the science of urban geochemistry and the challenges in developing a new sub-discipline. Recently, Tanner et al. (2014) posed two challenges in developing the sub-discipline of urban ecology: 1) how can urban ecology contribute to the science of ecology? And, 2) how can urban ecology be applied to make cities more livable and sustainable? One could easily replace “ecology” with geochemistry and ask the same questions.

In the first question, we ask what can be learned from studying urban systems (as opposed to solid earth or aquatic systems, for example) that aids in our quest to understand the chemical dynamics of the overall earth system. Urban environments are considered as dynamic, complex hot spots that drive environmental change at local to global scales, and thus are an important component of the overall earth system (Grimm et al., 2005, 2008; Lyons and Harmon, 2012). Urban geochemistry contributes to the science of geochemistry by understanding the principles controlling chemistry and chemical dynamics in urban environments. These principles can be advanced by: 1) Developing standardized sampling methodologies such as has been done in the EuroGeoSurveys’ Urban Geochemical Mapping projects (Demetriades and Birke, 2015a, 2015b), 2) comparing geochemical signatures in cities with different physical, economic, and sociological conditions across the world, 3) quantifying biogeochemical cycles in these cities through transport modeling in various media such as aerosols and water (e.g., Cermak and Takeda, 1985; Lv et al., 2015; Chen et al., 2016; Beisman et al., 2015), 4) examining undeveloped systems adjacent to the urban areas measured in points #1 and #2 to understand the degree and scale of urban influence, and 5) integrating legacy geochemical processes with current trends to predict the impact of changes in climate, water supplies, growth rates, industrialization rates, policy, and culture.

The second question is more applied — how can our geochemical investigations serve and benefit society? We are primarily interested in the geochemistry/biogeochemistry of urban systems because we want to maintain or improve both human and ecosystem health, sustain ecosystem services, and minimize natural resource wastage. In other words, how can geochemists aid in the development of more sustainable cities?

Following an ecosystem/ecoservices approach, it is important to recognize the effects that urbanization has on ecosystem function: habitat loss and fragmentation, loss of species diversity and richness, altered hydrology, and changes in the flow of energy and nutrients (Alberti, 2005). Similarly, human amplification of geochemical processes, especially in urban areas, has greatly affected most of the elements in the periodic table (Sen and Peucker-Ehrenbrink, 2012). The field of urban geochemistry is uniquely positioned to quantify changes to ecosystem functioning in urban environments through the five approaches listed above.

Another challenge facing urban geochemistry research is the need to interact with individuals from other disciplines and training backgrounds such as social scientists, city managers and planners, public health practitioners, epidemiologists, and environmental and civil engineers (Kaushal et al., 2014). The structuring of interdisciplinary groups can be daunting and initially have many disincentives and barriers, but as Tanner et al. (2014) point out, “broad multifaceted collaborations have made remarkable progress in understanding the holistic social-biophysical-ecological processes of urbanization.”

A diversity of cross-disciplinary approaches could both strengthen and broaden our science, and the two NSF-funded urban LTER sites in the United States are an example of this idea. However, the diversity of research approaches needed to solve the complexities of urban geochemical problems (e.g., the interfaces of health, social, basic and applied sciences) leads to a fourth challenge — funding. Funding for interdisciplinary research can be complicated because many funding agencies such as the NSF, EPA, and National Institutes of Health (NIH) have targeted missions that can preclude holistic funding.

The IAGC Urban Geochemistry Working Group is well positioned to take on these challenges. We propose a structure similar to other highly successful IAGC Working Groups, such as Water-Rock Interaction, to help address the first two challenges. The third challenge, cross-disciplinary conversation, will be facilitated by encouraging participation by urbanists from other disciplines to biennial Working Group meetings, the location of which will change in order to focus on a diversity of urban settings (e.g., decaying, rebounding, new). The Working Group will continue to host special topic sessions at national and international earth science meetings such as the relatively new yearly session on Urban Geochemistry at the annual Geological Society of America meeting, and the annual Goldschmidt Conference. At these sessions and at Working Group meetings, we will invite non-geochemists to speak and share their ideas for potential collaboration. The Working Group may be in a position to help create novel solutions to this funding challenge by fostering multidisciplinary collaborations that use specific agency funds for targeted research that, taken as a whole, address more comprehensive studies.

The consensus from Columbus meeting was that urban geochemistry is an important component within the domains of geochemistry and earth systems science. The keen interest in this subject is demonstrated by the frequent requests from publishers to develop special issues for journals or books (e.g., Long and Lyons, 2015; Lyons and Harmon, 2012). The importance of urban geochemical research will grow in the future because urban environments are expanding in both spatial and population scales. There are over 130 countries in which 50% of the population resides in urban environments, and over 50 countries with urban populations above 80%. Less than 30 of the over 200 countries that supplied data have a zero or a negative urbanization growth rate (United Nations, 2015). Human and ecosystem health will be at continued risk at global scales from disrupted biogeochemical cycles that affect our air, water, soil, and food.

Shneider (2009) provides an interesting perspective on the possible four stages in the development of a scientific discipline that we might apply to urban geochemistry. In stage one, new objects and phenomena are introduced as subject matter for the new discipline. During stage two, methods and techniques are introduced. Stage three is concerned with the application of new research methods to objects and/or phenomena. In the fourth stage, scientific knowledge developed during the first stages are maintained and passed on and critical revisions are made in the role of a discipline or sub discipline, within the greater scientific context. The boundaries between stages or the lengths of time at a particular stage are not well defined. For urban geochemistry, stage one is well passed and justified as shown in this paper. A major contribution of this paper to urban geochemistry might be the developments that occur in stage two. With the growing numbers of publications related to urban geochemistry we might suggest that urban geochemistry is moving into stage three.
Finally, after synthesizing the discussion of the five themes outlined in this article, we offer two perspectives on the science of urban geochemistry. The first is a definition of urban geochemistry that builds on the work of Thornton (1991): Urban geochemistry as a scientific discipline provides valuable information on the chemical composition of environments that support large populations and are critical to human health and well-being. Research into urban geochemistry seeks to 1) elucidate and quantify the sources, transport, transformations, and fate of chemicals in the urban environment, 2) recognize the spatial and temporal (including legacies) variability in these processes, and 3) integrate urban studies into global perspectives on climate change, biogeochemical cycles, and human and ecosystem health. The second is an overarching question, building on the work of Lyons and Harmon (2012) that attempts to link all the research endeavors of urban geochemistry: How do the developmental stage, development history, and state of economic growth influence biogeochemical cycles in urban environments? This question itself cannot be written as a hypothesis because it is not falsifiable, but hopefully it will stimulate constructive hypotheses to help guide urban geochemistry research.

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