Road sediment, an underutilized material in environmental science research: A review of perspectives on United States studies with international context

Matthew Dietrich[1], Michael J O'shea[2], Reto Gieré[2], and Mark P. S. Krekeler[3]

[1]Indiana University - Purdue University Indianapolis
[2]University of Pennsylvania
[3]Miami University

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Abstract

Road sediment is a pervasive environmental medium that acts as both source and sink for a variety of natural and anthropogenic particles and often is enriched in heavy metals. Road sediment is generally understudied in the United States (U.S.) relative to other environmental media and compared to countries such as China and the United Kingdom (U.K.). However, the U.S. is an ideal target for these studies due to the diverse climates and wealth of geo-chemical, socioeconomic, demographic, and health data. This review outlines the existing U.S. road sediment literature while also providing key international perspectives and context. Furthermore, the most comprehensive table of U.S. road sediment studies to date is presented, which includes elemental concentrations , sample size, size fraction, collection and analytical methods, as well as digestion procedure. Overall, there were observed differences in studies by sampling time period for elemental concentrations, but not necessarily by climate in the U.S. Other key concepts addressed in this road sediment review include the processes controlling its distribution, the variety of nomenclature used, an-thropogenic enrichment of heavy metals, electron microscopy, health risk assessments , remediation, and future directions of road sediment investigations. Going forward, it is recommended that studies with a higher geographic diversity are performed that consider smaller cities and rural areas. Furthermore, environmental justice must be a focus as community science studies of road sediment can elucidate pollution issues impacting areas of high need. Finally, this review calls for consistency in sampling, data reporting, and nomenclature to effectively expand work on understudied elements, particles, and background sediments.

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Road sediment, an underutilized material in environmental science research: A review of perspectives on United States studies with international context

Matthew Dietrich[1], Michael J. O'Shea [2], Reto Gieré[2,3], Mark P.S. Krekeler[4,5]

[1]Department of Earth Sciences, Indiana University – Purdue University Indianapolis, Indianapolis, IN, USA

[2]Department of Earth and Environmental Science, University of Pennsylvania, Philadelphia, PA 19104-6316, USA

[3] Center of Excellence in Environmental Toxicology, University of Pennsylvania, Philadelphia, PA 19104-3616, USA

[4] Department of Geology & Environmental Earth Science, Miami University, Oxford, OH 45056, USA

[5]Department of Mathematical and Physical Sciences Miami University-Hamilton, Hamilton, OH 45011, USA

Abstract: Road sediment is a pervasive environmental medium that acts as both source and sink for a variety of natural and anthropogenic particles and often is enriched in heavy metals. Road sediment is generally understudied in the United States (U.S.) relative to other environmental media and compared to countries such as China and the United Kingdom (U.K.). However, the U.S. is an ideal target for these studies due to the diverse climates and wealth of geochemical, socioeconomic, demographic, and health data. This review outlines the existing U.S. road sediment literature while also providing key international perspectives and context. Furthermore, the most comprehensive table of U.S. road sediment studies to date is presented, which includes elemental concentrations, sample size, size fraction, collection and analytical methods, as well as digestion procedure. Overall, there were observed differences in studies by sampling time period for elemental concentrations, but not necessarily by climate in the U.S. Other key concepts addressed in this road sediment review include the processes controlling its distribution, the variety of nomenclature used, anthropogenic enrichment of heavy metals, electron microscopy, health risk assessments, remediation, and future directions of road sediment investigations. Going forward, it is recommended that studies with a higher geographic diversity are performed that consider smaller cities and rural areas. Furthermore, environmental justice must be a focus as community science studies of road sediment can elucidate pollution issues impacting areas of high need. Finally, this review calls for consistency in sampling, data reporting, and nomenclature to effectively expand work on understudied elements, particles, and background sediments.

Keywords: Road sediment, road dust, urban pollution, heavy metals, electron microscopy

1. Introduction

1.1 Overview

Road sediment is a pervasive and mobile environmental medium, which is present in every urban, suburban, and rural community. Its physical properties and chemical composition are affected by multiple sources, retention and redistribution processes, as well as sediment transport and chemical mobilization processes. Here, we summarize several important studies and developments focused within the United States (U.S.) over the last several decades regarding sediment transport, geochemical characterization, risk assessment, and remediation of road sediment. The literature reviewed for this work forms the basis of road sediment research thus far in the U.S., but we include international sources to provide essential context and acknowledge that this review is not exhaustive worldwide. Furthermore, to retain focus for this review we primarily discuss inorganic metal(loid) compounds, although we recognize the importance of organic contaminants in road sediment, such as polycyclic aromatic hydrocarbons (PAHs), among numerous others.

Road sediment is an underutilized environmental medium relative to water, air, or soils for studying pollution, particularly within the U.S. Regarding soil, water and air pollution in the U.S. a basic Web of Science search of articles in August of 2021 indicated that there are tens of thousands of articles relating to these media. Conversely, using the terms “road dust” / “road sediment” / “street sediment” globally returns 743, 357, and 35 articles, respectively, and for the U.S. specifically returns 131, 58, and 10 articles, respectively, and at least 125 unique articles. This documents the lack of investigations on road sediment relative to other environmental media, especially in the U.S. compared to well-studied countries such as China (approximately 600 studies). Other areas of the world such as Europe have also extensively studied road sediment (e.g., Charlesworth et al., 2003; Dean et al., 2017; Padoan et al., 2017; Teran et al., 2020; Zglobicki et al., 2019), with the United Kingdom (U.K.) in particularly studying road sediment more than the U.S. (Haynes et al., 2020), despite its much smaller geographic area. Although search terms likely vary, this demonstrates globally that the U.S. has much room to grow regarding road sediment research, especially considering the large population and geographic area of the U.S.

Despite its underrepresentation, road sediment is valuable for pollution research, as it is a publicly accessible medium, which is composed of natural and anthropogenic materials. Additionally, road sediment is useful for spatially quantifying and relating pollutants of concern to elevated health risks, particularly in areas that are socioeconomically disadvantaged. For example, in predominantly African American and low-income portions of Hamilton, OH (Flett et al., 2016) and Gary, IN (Dietrich et al., 2019), road sediment contains high levels of metal pollutants such as Pb or Mn. Critically, nearly 500,000 children less than 6 years old in the U.S. have elevated blood Pb levels (BLLs $\geq$5 μg/dL) (Hauptman et al., 2017) and African American children are disproportionately affected by Pb poisoning in the U.S., illustrating racial disparities in pollution exposure (e.g., Yeter et al., 2020). The exact relationship between Pb content in road sediment and BLLs in children is not quantified. However, the systematic distribution of Pb in road sediment identifies probable primary and secondary paths of environmental exposure. Furthermore, through geospatial analysis, the relationships of pollutant concentrations in road sediment with health, demographic, and socioeconomic survey data can potentially be assessed.

The U.S. is a useful locale for holistic review of road sediment primarily due to the wide variety of climates that are represented and a well-documented set of environmental rules and regulations. Additionally, the variety of cities, ranging from older, post-industrial cities to newer, developing cities, has a broad history of potential for research studies. Furthermore, there is a wealth of information present in the U.S. such as census data and detailed environmental, industrial, and city records. All of this can be used to determine the health-relevant context of road sediment studies and provide opportunity for future health-based studies on road sediment. There is also ample geologic, geochemical, and climate data constraints that exist nationwide, which can further inform studies. Such context enables a large variety of hypotheses to be developed and be readily tested. Finally, due to the diversity of investigations, research in the U.S. can provide useful case studies that can be applied worldwide.

Although road sediment pollution research in the U.S. has much room to grow, there have been multiple advances in the last several years, including source apportionment through scanning electron microscopy (SEM), chemical mass balance approaches, larger geographic sampling representation, health risk assessment, multivariate statistical approaches, and spatial geographic information system (GIS) analyses (e.g., Fiala and Hwang, 2021; O’Shea et al., 2020; Dietrich et al., 2019; Lloyd et al., 2019; Dietrich et al., 2018; Flett et al., 2016). This follows alongside general worldwide advances in road sediment research such as investigations involving SEM, health risk assessment, and assessing adsorption behavior (e.g., Jayarathne et al., 2018; Jayarathne et al., 2019; Teran et al., 2020; Zhang et al., 2020). Thus, this review aims to expand on previous reviews of road sediment pollution (Haynes et al., 2020; Hwang et al., 2016; Loganathan et al., 2013; Roy et al., 2022) to describe research accomplishments thus far and what the future research directions should be. The goals of this review are to 1) emphasize the importance of road sediment as an environmental medium, 2) elucidate the status of road sediment research in the U.S. with international context, and 3) identify the primary outstanding scientific questions that should be addressed by researchers in the future and prioritize these major questions.

1.2 Nomenclature

One challenging aspect of conducting this review is addressing the diverse and non-uniform nomenclature that exists on the topic (Table 1). The exact origin of the term we label "road sediment" is unclear. Indeed, a wide variety of studies offer different names when referring to the same medium. One of the first wide-scale studies of the material was with the U.S. EPA (Pitt and Amy, 1973), where, among others, the term "street surface contaminants" was used. However, studies in the late 70s utilized other nomenclature such as Farmer and Lyon (1977), whose Glasgow study used the term "street dirt." Subsequently, many studies began to apply the term "street dust" or "urban street dust", beginning with a series of studies from the United Kingdom (e.g., Day et al., 1975; Duggan and Williams, 1977; Harrison, 1976; Harrison, 1979; Fergusson and Ryan, 1984). These terms were also used in an Illinois, U.S. study during the same era (Solomon and Hartford, 1976). The research by Ferguson and Ryan (1984) on "street dust" represents one of the first global examinations of the medium, as samples from London, New York City, Halifax, Christchurch and Kingston were analyzed. The use of the names "street dust" and "urban street dust" have continued internationally to this day (e.g., Li et al., 2001; Charlesworth et al., 2003; Tanner et al., 2008; Zheng et al., 2010; Tang et al., 2013; Lu et al., 2014; Dean et al., 2017; Zglobicki et al., 2019; Bartholomew et al., 2020; Teran et al., 2020).

While less common, there are other studies such as an Illinois study by Hopke et al. (1980) who coined the phrase "urban roadway dust." This name is similar to the terms "road dust" or "urban road dust" which have been utilized by numerous international (e.g., Liu et al., 2007; Amato et al., 2009a; Wei and Yang, 2010; Shi et al., 2011; Zannoni et al., 2016; Zhao et al., 2016; Bourliva et al., 2017; Jayarathne et al., 2019) and U.S. studies (Kalenuik and Deocampo, 2011; Deocampo et al., 2012; O'Shea et al., 2020).

Other references to this medium include "urban sediment" (Selbig et al., 2013), "street particles" (e.g., Lau and Stenstrom, 2005), and "road-deposited sediments" (e.g., Sutherland et al., 2000; Sutherland and Tolosa, 2000; Sutherland, 2003; Andrews and Sutherland, 2004). Similarly, the terms "street sediment" and "road sediment" have been used in the past (e.g., Dietrich et al., 2018; Dietrich et al., 2019; Flett et al., 2016; Irvine et al., 2009; LeGalley et al., 2013; LeGalley and Krekeler, 2013; Zibret and Rokavec, 2010).

Ultimately, all of these names refer to the same environmental medium, which accumulates on the street surface and represents the local natural and anthropogenic environment. Interestingly, later studies often use a different term than the studies they refer to. The actual term used is almost always defined in the introduction by the authors, but we suggest that moving forward, a single unified term is utilized to avoid confusion. We have selected to use the term "road sediment" in this review rather than "street sediment," as "road" seems to be a more encompassing term, where multi-lane highways and gravel lanes are both roads. "Street" seems to have the more restricted connotation of a paved road of medium size. Although this seems a minor point, by committing to a singular term moving forward, library work will become more efficient, minor issues in the review process may be reduced for researchers, and more uniform communication to researchers outside of this topic may occur. If future regulations were to evolve in the U.S., a codified single term such as "road sediment" would be pragmatic.

2. Results & Discussion

2.1 Processes controlling road sediment distribution

There are numerous processes and mitigating factors that control and impact the nature of road sediment distribution. These processes can be generally grouped into inputs, redistribution and modification, retention, and outputs, and these categories have numerous hypothetical subprocesses or factors that may vary from location to location (Table 2). These factors undoubtedly explain the large degree of material variations that are observed in road sediment, even within the same region of a city (e.g., LeGalley and Krekeler, 2013; Flett et al. 2016). Factors relating to these hypothetical processes, which are discussed in the peer-reviewed literature over time, are presented.

2.1.1 Road sediment stormwater runoff

One of the first comprehensive studies on the physical transport of road sediment was performed by Sartor and Boyd (1972) for the U.S. Environmental Protection Agency (U.S. EPA). This study provided information that established approaches for future work focused on water runoff as an important road sediment transport mechanism. This work proposed an exponential equation for particle transport based on experimentally "simulated rainfall" stormwater runoff observations, which can be described as:

$$N_C = N_0(1 - e^{-krt}) \tag{1}$$

where N_c is the amount of material (at a particular grain size) in g/ft^2 removed during a rainfall event with a time duration in minutes (t), rainfall intensity in in/hr (r), and a proportionality constant (k) in hr/in*min that changes based on the properties of the street surface but is largely unrelated to particle size and independent of rainfall intensity (Sartor and Boyd, 1972). N_o is the initial amount of road sediment material in g/ft^2 subject to removal from the rainfall runoff. Equation (1) provided the basis for updated stormwater runoff equations used in recent research, with slight refinements applied, such as substituting a capacity factor (Cf) for N_o, which is how easily a specified rainfall intensity will transport particles such as road sediment, and a fraction wash-off (Fw) term for N_c, which represents a ratio of the wash-off load to the initial particle load (e.g., Egodawatta et al., 2007; Haddad et al., 2014).

Following Sartor and Boyd (1972), pioneering urban stormwater modeling was conducted by researchers such as Alley and Smith (1981). A large amount of empirical research in the U.S. Pacific Northwest was also performed in the 1980s, assessing sediment runoff from roads into nearby streams (e.g., Bilby, 1985; Bilby et al., 1989; Duncan et al., 1987; Reid and Dunne, 1984). Some main findings of this work in the Pacific Northwest were that paved roads transport less sediment to streams than unpaved roads and that more heavily used roads yield more sediment runoff than non-traversed roads (Reid and Dunne, 1984); small streams can capture a large portion of road sediment runoff, with the finest sediment transported downstream (Duncan et al., 1987); and that road sediment is flushed rapidly and because it tends to be relatively fine in grain size compared to streambed gravel, it has little overall effect on streambed gravel composition (Bilby et al., 1989).

In the 1990s, Sansalone's research group performed analytical work exploring urban road sediment runoff and the chemical characteristics of that runoff (e.g., Sansalone and Buchberger, 1997a; Sansalone and Buchberger, 1997b; Sansalone et al., 1998). Essentially, these authors found that in urban settings, "first-flush" events (where a disproportionately large quantity of sediment or an element is transported via stormwater runoff in the initial stages of the hydrograph) were commonplace for solid particles (Sansalone et al., 1998; Sansalone and Buchberger, 1997a) and several dissolved elements such as Zn, Cd and Cu (Sansalone and Buchberger, 1997a). Additionally, their work showed that the road sediment particles in runoff had specific surface areas (SSA) much greater than would be expected for spherical particles (Sansalone and Tribouillard, 1999). These features thus affect physical transport processes and possible sorption of metals. In fact, Sansalone and Buchberger (1997b) showed how following both snow- and rain-fall events, the concentrations of Cu, Zn and Pb increased with decreasing particle size of the road washout sediment and that this particle size effect was directly related to SSA. Other important findings were that 1) very small road sediment particles (2-8 μm) were quickly flushed during high runoff conditions; 2) high runoff events are mass-limited, because there is often little available dissolved or suspended road sediment mass for stormwater runoff transport; and 3) when runoff is low, sediment transport becomes flow-limited because of a higher vehicle/runoff volume ratio and abundant road sediment mass (including the dissolved load) available for transport (Sansalone et al., 1998).

This notion of "first flush", presented by Sansalone's research group, has also been extensively applied internationally in stormwater management. Some researchers found this effect on highways (e.g., Gupta and Saul, 1996; Sansalone and Buchberger, 1997b; Lau et al., 2002; Ma et al., 2002), whereas others did not (Barrett et al., 1998). Li et al. (2005) found that approximately 40% of road sediment particles were mobilized with the first 20% of water volume from runoff.

2.1.2 Particle size and particulate metal runoff

A series of studies in Hawaii assessed the mobility and transport of road sediment in the early 2000s, led by R. A. Sutherland. Their work catalogued road sediment as a critical source of pollutants, which led to the degradation of local water bodies, including Manoa Basin

(Sutherland and Tolosa, 2000). They reported enrichment of elements such as Pb, Sn, and Zn, and that these elements were primarily related to vehicle pollution. To better understand elements enriched in this material, road sediment from Oahu, Hawaii was analyzed through an optimized four-step sequential extraction (Sutherland et al., 2000). They documented that Al, Co, Fe, Mn, and Ni were likely lithogenic-pedogenic in nature. They also noted that Zn could be mobilized with decreasing the pH in water. Furthermore, in a later study, Sutherland size-fractionated the road sediment samples and described particles that are < 63 µm accounted for a majority of the medium's mass, including total Pb present (Sutherland, 2003). However, at least one study demonstrated that metal mass loading may be greatest in larger particle size fractions primarily owing to a greater mass of midrange-coarser particles (Sansalone and Tribouillard, 1999). This example is complex, as the metal concentrations in midrange-coarser particles may be high, yet the bulk concentrations of metals may still be greatest in the smallest size fractions.

In a later investigation in Oahu, Andrews and Sutherland (2004) observed that several trace metals increased in concentration downstream as the river traversed residential, urban and commercial traffic areas. This may be related to the high traffic in these areas and the subsequent transport of polluted road sediment throughout the watershed. Similarly, more recent international studies found that land use and traffic can impact particle build-up and total particle loads (e.g., Gbeddy et al., 2018; Liu et al., 2016; Jayarathne et al., 2019). Road sediment transported by runoff degrades the quality of receiving water and impacts aquatic lifeforms (e.g., Liu and Sansalone, 2007; Marsalek et al., 2004).

Turer et al. (2001), building upon previous work (e.g., Sansalone and Buchberger, 1997a; Sansalone and Buchberger, 1997b; Sansalone et al., 1998), assessed the concentrations of pollutant metals in urban soils along highways in an effort to compare runoff and soil in Cincinnati, Ohio. They described high concentrations of metals in the top 15 cm of soil compared to background concentrations and that metal concentrations decreased with increasing depth. Furthermore, through sequential extractions, they reported that contaminants were predominantly not associated with soluble organic matter and that most Pb, Zn, Cu, and Cr were not exchangeable. Important work by Li et al. (2006) documented that most particles from highway runoff in West Los Angeles were < 30 µm in diameter. Furthermore, they observed that over 90% of particles were < 10 µm in diameter. Thus, because smaller particle size fractions have been generally shown to have elevated levels of metals compared to coarser particles (e.g., Characklis and Wiesner, 1997; Evans et al., 1990; Lee et al., 2005; Sansalone and Buchberger, 1997b; Sutherland, 2003), runoff may have a pronounced effect on particulate metal transport.

2.1.3 Road sediment resuspension

2.1.3.1 Resuspension overview

Resuspension can be either an atmospheric term, or a sedimentological term in aqueous settings. Resuspension is a complex process and may facilitate metal transport into ecosystems through a variety of mechanisms, such as physical dispersion of fine particles into water and air. Especially in aqueous resuspension, it is probable that adsorption and desorption of dissolved metals and organic molecules present in the system would occur. For example, resuspension is a well-recognized process for certain pollutants in aquatic environments such as polyaromatic hydrocarbons, polychlorinated biphenols (e.g., Latimer et al., 1999), and mercury (e.g., Kim et al., 2006), and as an important overall process in lakes (e.g., Bloesch, 1995). However, in this review, we will focus on the context of atmospheric resuspension, even though aqueous resuspension of road sediment through runoff processes also occurs.

2.1.3.2 Detailed road sediment particle resuspension studies

Prior to the late 1980s, research into particle resuspension from roads was sparse (e.g., Nicholson, 1988). Up until then, the main research on particle resuspension was completed by Sehmel (1973; 1976) who quantified particle resuspension rates with ZnS and demonstrated that resuspension rates from roads dropped over time from the original tracer ZnS particles, and that particle resuspension was lower in a cheat grass (vegetated road) area compared to asphalt roads. Additionally, Sehmel (1973) observed that vehicles driven at increased speeds through the ZnS tracer created the greatest particle resuspension on an asphalt road.

Beginning in the late 1980s, investigations of road sediment resuspension included improved quantitative apportionment of the contributions from road sediment to atmospheric particulate matter and more detailed work on the effects of vehicular traffic on resuspension. Research such as that by Nicholson et al. (1989) demonstrated that just one passing vehicle can resuspend a significant amount of road sediment. They found more resuspension at greater vehicle speeds in general and that larger silica particles (within four particle size groups, each with average particle diameters of 4.5, 9.8, 15.0, and 19.5 µm, respectively, and with a particle density of ~1000 kg/m^3 for porous silica) preferentially resuspended in general. Nicholson and Branson (1990) had similar results, but also importantly found that for the larger particle size groups in their study (defined as 9.5, 12, and 20 µm average particle size groups), particle resuspension was similar whether a vehicle drove over the road sediment or adjacent to it, illustrating that particle resuspension on roads can be induced relatively easily.

Several studies in the 1990s helped quantify the proportion of road sediment in the atmosphere through chemical mass balance and direct source analysis. Through chemically analyzing primary organic aerosol sources in the Los Angeles, U.S. area as well as using the source emission rates, road sediment was attributed to ~16% by mass of the total fine aerosol organic carbon emissions (Hildemann et al., 1991). This research was built upon by Schauer et al. (1996), who used a chemical mass balance approach to conclude that road sediment contributed significantly to atmospheric fine particulates in multiple southern California cities. Lankey et al. (1998) looked at Pb particulates in the California South Coast Air Basin and, using findings from previous research, stated that over half of particulate Pb by mass is deposited on or near roadways close to the particulate source, ~33% by mass is transported by wind outside of the basin, and <10% by mass is deposited on other surfaces within the basin. Using Pb road sediment resuspension estimates from the 1989 California Air Resources Board and a mass balance approach, Lankey et al. (1998) also estimated that about 43% of the Pb in the atmosphere (by mass) was from road sediment resuspension. However, they acknowledged that the percent contribution of resuspended Pb from soil to the atmosphere was unknown and that their resuspended road sediment estimate therefore included Pb from soils and roads.

Historically (i.e., 1950s-1980s), laboratory studies of particulate atmospheric resuspension were limited, but there were still several applications of laboratory work examining important controls on particle resuspension such as adhesion (e.g., Nicholson, 1988). Recently though, on-site laboratory road sediment resuspension experiments have gained utilization through “mobile” laboratories, particularly in the cases of vehicles with sensors or on-site resuspension chambers brought directly to roadways (e.g., Rienda and Alves, 2021). The ability of mobile laboratories to conduct sampling and analysis of road sediment resuspension directly on roadways helps bridge the disconnect between field-based and laboratory simulation studies. For more detailed information regarding recent developments in road sediment resuspension measurement methodologies, the reader is led to a comprehensive review by Rienda and Alves (2021).

Recent studies have identified resuspended road sediment and non-exhaust traffic emissions as sources of atmospheric pollution (e.g., Lough et al., 2005; Sabin et al., 2006; Zhao et al., 2006; Thorpe and Harrison, 2008; Amato et al., 2009a; Amato et al., 2009b; Bukowiecki et al., 2010; Amato et al., 2011; Harrison et al., 2012; Pant and Harrison, 2013; Zhao et al., 2016; Meza-Figueroa et al., 2018; Sommer et al., 2018). However, the exact impact of different resuspension mechanisms on particle transport is unclear, and therefore particle resuspension should be further explored in the urban environment.

2.1.4 Urban soil resuspension

Urban soils are commonly geochemically related to road sediment, particularly near roadways, and are often transported to roadways via resuspension. Past investigations have noted that road sediment was approximately 57-90% soil-derived by mass (Hopke et al., 1980; Fergusson and Ryan, 1984) whereas other studies have continued to explore soils as a sink for road sediment (e.g., Li et al., 2001; Padoan et al., 2017; Shi et al., 2008; Wei and Yang, 2010). The transport of resuspended soils to the atmosphere, and then to road sediment, may depend on the spatial patterns of vehicular and lawn mowing induced turbulence, soil moisture, and seasonal variation (e.g., Hosiokangas et al., 2004; Kuhns et al., 2001; Laidlaw et al., 2012; Lough et al., 2005; Nicholson et al., 1989). Additionally, other investigations support this hypothesis (e.g., Lenschow et al., 2001; Davis and Birch, 2011).

As road sediment and soils can serve as both sources and sinks to one another, and may do so repeatedly, it is important to acknowledge this relationship and its complexity. Lead is an element that has especially been evaluated in the context of urban settings. Generally, road sediment Pb contamination has been understudied compared with soil Pb contamination (e.g., U.S. EPA, 1996; U.S. EPA, 1998; Laidlaw and Filippelli, 2008; Mielke et al., 2011; Lusby et al., 2015; Filippelli and Taylor, 2018; Frank et al., 2019) in the U.S., despite their similarities. A recent U.S. review of Pb concentrations in environmental media, including soils, outlined that mean Pb concentrations were nearly three times higher in residential urbanized areas compared to residential non-urbanized areas (Frank et al., 2019). The same has been demonstrated for road sediment, where Pb was higher in concentration in urbanized areas compared to less urbanized areas near Boston, MA (Apeagyei et al., 2011). The exact hotspots of Pb contamination can be hard to find if there are no clear emission sources and sinks (e.g., Zahran et al., 2013; Filippelli et al., 2015; Laidlaw et al., 2017; Mielke et al., 2019; Filippelli et al., 2020). Identifying hotspots of Pb and other contamination is important although, as studies have found soil and dust particles smaller than 10 µm are highly susceptible to resuspension and thus Pb and other pollutants may be blown into homes (e.g., Lepow et al., 1974; Archer and Barratt, 1976; Sayre and Katzel, 1979; Bornschein et al., 1986; Fergusson, 1986; Davies et al., 1987; Farfel and Chisolm, 1990; Al-Radday et al., 1993; Kutlaca, 1998). These transported particulates can come from a combination of road sediment and nearby soils. In addition to particle size, traffic plays an important role in road sediment particulate resuspension, as most Pb deposited from vehicles may be within 50 m of the roadside in soil and decreases with distance from the road (e.g., Laidlaw and Taylor, 2011).

2.1.5 Seasonality

Seasonality is an important variable, which impacts road sediment and urban soil resuspension. There has been some work on the seasonal variation of atmospheric Pb concentrations, where authors generally observed summertime highs of atmospheric Pb in various U.S. cities and states (Billick et al., 1979; Edwards et al., 1998; Green and Morris, 2006; Melaku et al., 2008; Paode et al., 1998; U.S. EPA, 1995; Yiin et al., 2000). Authors have suggested that blood Pb seasonality may be a product of soil and dust resuspension increasing in certain months of the year (e.g., Filippelli et al., 2005; Laidlaw et al., 2005). Ultimately, blood Pb tends to peak in the summer and autumn (Laidlaw and Filippelli, 2008). To better understand the seasonality of Pb soil resuspension from multiple regions, Laidlaw et al. (2012) studied the resuspension of Pb-bearing soils as a source of Pb in the atmosphere in Pittsburgh, Birmingham, Chicago, and Detroit. They found that resuspended soils and Pb aerosols were related in Pittsburgh (R^2=0.31, p<0.01) from April 2004 to July 2005, in Birmingham from May 2004-December 2006 (R^2=0.47, p<0.01), in Chicago from November 2003-August 2005 (R^2=0.32, p<0.01), and in Detroit from November 2003-July 2005 (R^2=0.49, p<0.01). Furthermore, as also reported by previous inquiries, there were seasonal peaks in the summer and fall where resuspended soil and atmospheric Pb were at their highest concentrations. Ultimately, seasonal and temporal variation of road sediment must be consistently measured and assessed (e.g., Laidlaw et al., 2005; Laidlaw and Filippelli, 2008; Laidlaw, 2010; Filippelli et al., 2020).

Although seasonality has a strong impact on road sediment and urban soil resuspension, a directly related component of seasonality, climate, does not seem to have a large influence on road sediment concentrations of Cr, Pb, Zn and Cu in the U.S. (Fig. 1). Based on multidimensional scaling and Aitchison distance, these metal concentrations seem to differ more based on the year of sampling as opposed to the region they were sampled in. This points to climate/region having a lesser effect on metal concentrations in road sediment compared to changes in pollutant sources over time such as leaded gasoline. The distinction between pre- and post-1990 data was made in order to illustrate possible differences between early road sediment studies and those where sampling occurred after the near complete phase-out of leaded gasoline in the U.S. Most, but not all leaded gasoline was phased out in the U.S. by 1986, with ~5-6 million metric tons of Pb added to gasoline up to that point (Laidlaw et al., 2012 and the references therein). Thus, samples collected prior to 1990 were closer to peak Pb gasoline usage in the U.S. and were higher in Pb (Fig. 2). However, this does not discount seasonality and climate playing important roles in resuspension and distribution of the bulk contents of road sediment, such as resuspension into the atmosphere or into homes.

2.2 Anthropogenic enrichment of heavy metals and metalloids in road sediment

A primary concern with road sediment is the occurrence of toxic metal(loid)s at concentrations that can have potential negative environmental impacts and detrimental human health effects. While other works in the past have compiled tables of international road sediment metal(loid) concentrations (e.g., Flett et al., 2016; Hwang et al., 2016; O'Shea et al., 2020), the present review has compiled an extensive list of nearly all major road sediment heavy metal(loid) analyses completed in the U.S. over the last 50 years (Table 3; Fig. 2). This summary identifies different methodologies employed in road sediment research thus far, such as collection method, grain size fractionation, and sample digestion method. Furthermore, this summary illustrates the geographical extent of road sediment research in the United States (Fig. 3) and is the most up-to-date comprehensive dataset for heavy metal and metalloid concentrations in U.S. road sediments (Table 3). This dataset also serves as a useful tool for understanding general changes in pollution over time in the U.S. from regulatory implementation (pre- and post-leaded gasoline usage), as well as which elements (i.e., Hg) have been understudied thus far.

When comparing to recent road sediment studies in China, it is evident that the enrichment of several metals relative to upper continental crust (UCC) is broadly similar in both the U.S. and China (Fig. 2), although it is noted that general lithogenic background material will differ slightly between countries. While the countries are separated geographically, their metal concentrations in road sediment are comparable, suggestive of similar general sources and metal enrichments even if climate and history may differ. Importantly though, it is noted that the China post-1990 Cd values more closely resemble the U.S. pre-1990 Cd values, and the China post-1990 Pb values are closer to the post-1990 U.S. values than U.S. pre-1990 Pb values (Fig. 2). These two heavy metals highlight both differences and similarities in industrial growth and environmental pollution regulation between the U.S. and China. Specifically, Cd concentrations in China may be reflective of the large amount of Cd released from fertilizers, pesticides, and the metal production industry in the last 2-3 decades, more closely mirroring the rapid industrial growth China experienced post-1990 relative to some metals such as Cu and Zn, which are sourced primarily from vehicular waste (e.g., Jiang et al., 2016). However, Pb road sediment enrichment in China may closely resemble the decline seen in U.S. road sediment because of similar emphasis on leaded gasoline phase-out beginning in the 1980s, with complete elimination of leaded gasoline by 2000 in China (e.g., Wang et al., 2006).

Although a detailed global comparison of road sediment metal enrichment is outside the scope of this study, it is noted that a recent review by Roy et al. (2022) compiles continental comparisons of road sediment metal concentrations, and states that in Europe there also appears to be a trend of lower Pb concentrations in more recent studies of road sediment, possibly linked to the modern usage of unleaded gasoline.

Road sediment is commonly enriched in several metals relative to background reference materials that have undergone minimal anthropogenic perturbation, such as UCC (Fig. 2). Multiple studies have also used various reference materials such as minimally perturbed background sediments in close vicinity of the study area instead of UCC to illustrate the anthropogenic footprint in road sediment (e.g., Sutherland and Tolosa, 2000; Dietrich et al., 2019). Some elements in road sediment commonly linked to anthropogenic sources include Cd, Cr, Cu, Pb, and Zn. Specific anthropogenic sources from vehicles, households, industry, and commercial activity include, but are not limited to: vehicle traffic and coal combustion for Cd (e.g., Zgłobicki et al., 2018); house siding and brake wear for Cu (e.g., Davis et al., 2001); house siding and tire wear for Zn (e.g., Davis et al., 2001); steel facilities (e.g., Dietrich et al., 2019) and lead chromate ($PbCrO_4$) particles in road paint for Cr (e.g., White et al., 2014; O'Shea et al., 2021a); while Pb has sources including $PbCrO_4$ (e.g., White et al., 2014), lead wheel balancing weights in vehicles (e.g., Ayuso and Foley, 2020; Hwang et al., 2016), other Pb-bearing paint components (e.g., Tchounwou et al. 2012), or past deposition of leaded gasoline that may be resuspended as soil particles in the environment (e.g., Laidlaw et al., 2012). Thus, potential anthropogenic sources of heavy metals are numerous in urban settings.

Sediments such as glacial till in the midwestern U.S. (Barnes et al., 2020) or sands from arid regions in the western U.S. (Oglesbee et al., 2020) may also be useful background materials if they are likely source materials in the area. However, complications can arise if there are multiple potential sources of background contribution with different geochemical signatures. If this is the case (i.e., glacial till juxtaposed to nearby exposed deposits of ultramafic rocks), then it is recommended that comparisons of the "contaminated media" be made against either multiple potential background sources (e.g., Dietrich et al., 2019), or a mixing model be employed that estimates a theoretical background sample with reasonable proportions of potential sources based on distances to these sources and their relative abundances in the area. Additionally, it is recommended that several background samples collectively be used as reference to road sediment or other contaminated media. Specifically, at least three background samples will enable a reasonable standard deviation (1σ) to be calculated, thus constraining the geochemical variability within background material, which can be reflected through reporting contamination indices with 1σ variability instead of only the raw, computed values.

Other common ways to assess anthropogenic pollution in road sediment, besides a simple ratio of road sediment/background material, are through "enrichment factors" and "geoaccumulation index" calculations. The enrichment factor is a common way to distinguish pollution from natural contributions through using a reference element to normalize the potential pollutant of interest (e.g., Loska et al., 1997; Chen et al., 2007; Çevik et al., 2009). For example, in the equation:

$$EF = \frac{(\frac{M_{Sample}}{Al_{Sample}})}{(\frac{M_{Ref}}{Al_{Ref}})} \tag{2}$$

the enrichment factor (EF) is calculated through first utilizing the concentration of the potential contaminant of interest in the sample (M_{Sample}) and the concentration of Al or another reference element in the sample (Al_{Sample}). Then, the concentration of the potential contaminant of interest in the background or reference material (M_{Ref}) and the concentration of Al or another reference element in the background or reference material (Al_{Ref}) can all be used to quantitatively assess the degree of pollution in a road sediment sample. The reference material can be shale or UCC, but it is recommended that the reference material be representative of geogenic sourcing to the study area. The reference element or "tracer" should not be strongly affected by the same processes as the element of interest. Oftentimes Al and Fe are used because of their abundance in the environment (e.g., Çevik et al., 2009; Chen et al., 2007; Loska et al., 1997).

The geoacumulation index (I_{geo}) is another way to compare an anthropogenically affected sample to an unaffected background sample. It was originally developed by Müller (1969) and is still widely used by scientists to assess potential pollution in sediments (e.g., Chen et al., 2007; Çevik et al., 2009; Wei and Yang, 2010). The I_{geo} equation can be represented as:

$$I_{geo} = log_2\left(\frac{C_{sample}}{1.5C_{background}}\right) \tag{3}$$

where C_{sample} is the concentration of the element of interest in the road sediment sample, whereas $C_{background}$ is the concentration of that element in the background or reference sample. The value of 1.5 is used to help correct for possible lithogenic variation in the background concentration (e.g., Çevik et al., 2009). Positive I_{geo} values are indicative of some anthropogenic contribution to the sample.

While there are classification schemes regarding "how polluted" a sample may be based on EF and I_{geo} values that road sediment and sediment studies in general often use (e.g., Çevik et al., 2009; Trujillo-González et al., 2016), it is cautioned that these can be largely arbitrary. The true assessment of pollution is usually site- and element-specific, particularly because some elements may be more easily mobilized than other elements, sites may have undocumented geologic "background" contributions, and certain sites may be more sensitive to pollution. This concern over traditional usage of indices such as I_{geo} or *EF* has been raised recently (Hopke and Jaffe, 2020), particularly because of the abundance of more advanced statistical and analytical capabilities now available, which can better quantify anthropogenic enrichment. For example, Haynes et al. (2020) recommends principal component analysis (PCA) to help differentiate between elements that are likely of geogenic origin, and thus can be used for background calculations such as the enrichment factor. Thus, it is encouraged to not simply rely on one contamination index ratio to assess anthropogenic enrichment at a site, but instead several different factors, such as taking into account multiple samples with a range of data and considering all potential background geologic material as feasible.

2.3 Particulate matter (PM) in the context of road sediment

The grain size of road sediment is critical to understand source, transportation mechanisms, and potential exposure to the medium. Pollutant particles may exist in the silt and sand fraction, or even larger particles as debris or debitage from vehicles or objects, such as the road surface. However, of special concern are particulate matter ≤10 μm in aerodynamic diameter (PM_{10}) and particulate matter ≤ 2.5 μm in aerodynamic diameter ($PM_{2.5}$). Both size fractions have been extensively studied as they are inhalable and potentially health relevant (e.g., Kastury et al., 2017). Generally, the finer the particles, the more readily they can be inhaled and the deeper they penetrate the respiratory system. When inhaled, PM_{10} can reach the tracheo-bronchial region whereas larger particles (still less than 100 μm) may be lodged in the naso-pharyngeal region (e.g., Newman, 2001; Kastury et al., 2017). Particles in the $PM_{2.5}$ category may reach the alveolar region, and particles smaller than 0.1 μm may reach the lung interstitium and extrapulmonary organs (e.g., Nemmar et al., 2013).

Often, road sediment samples will be size fractionated in order to separate the above categories and better understand their risks. Common techniques to study these road sediment particles include scanning electron microscopy (SEM) for the microscale and transmission electron microscopy (TEM) for the nanoscale, both of which have been recently advocated for as a tool in road sediment research (Haynes et al., 2020).

2.3.1 Microscopic investigations of road sediment particles

2.3.1.1 SEM investigations

Scanning electron microscopy is a key environmental materials characterization technique as it can provide particle size, texture, and chemical composition information on metal pollutants in road sediment. This is complex, however, as the user must have experience in discerning natural materials derived from local geological materials and soils from potential anthropogenic particles. Interpretations of materials can be complicated by the presence of multiple minerals or materials in the sample of interest. For example, due to electron beam interactions (for energy-dispersive X-ray (EDX) spectroscopy)), surrounding particles and substrate may contribute to the X-ray spectra for a potential pollutant particle, obscuring the pollutant signal. One related issue is in determining whether a given particle is a metal, oxide, or an aggregate of both. One approach is to utilize backscatter imaging and assess contrast, as metals will always be brightest or brighter than their oxide equivalents. We note that, unlike the geologic literature where there are organized “atlases” of mineral and rock SEM data, there are no comprehensive SEM data sets for road sediment. Such a compilation is beyond the scope of this paper but would be of high utility for numerous researchers.

Electron microscopy has not been used as extensively in the study of road sediment compared to typical bulk chemistry techniques such as inductively coupled plasma-optical emission spectroscopy (ICP-OES), because many past assessments primarily focused on elemental concentration. SEM investigations of road sediment show a wide range of particle size and types. Commonly encountered particle types fall into the broad categories of metals, technogenic spherules, simple minerals or mineral analogs, and tire-wear particles. Investigations of road sediment particles using SEM-EDX include those of Teran et al. (2020) and Gunawardana et al. (2012). Gunawardana et al. (2012) found that tire-wear particles were prominent pollutants, supporting previous research (e.g., Adachi and Tainosho, 2004). Using SEM and EDX, Teran et al. (2020) primarily interpreted metal-rich particles to be derived from steel production and tire and brake pad wear, and clearly identified differences between urban and rural road sediments. This was similar to work done by Dietrich et al. (2018), whose later SEM work also identified anthropogenic inputs of local steel mills to road sediment (Dietrich et al., 2019). Dietrich et al.’s work built on pollution identification work such as that of Flett et al. (2016) and Legalley and Krekeler (2013). Other studies utilized SEM and EDX to quantitatively characterize road sediment morphology based on composition (e.g., Jayarathne et al., 2018). Furthermore, they also interpreted the sources of road sediment pollution.

2.3.1.1.1 SEM investigations of lead phases, particles, and aggregation

Lead and Pb-rich phases are of high interest owing to the toxicity of these materials. Road sediment often contains Pb-rich particles (e.g., Pb, PbO, $PbCrO_4$), which may be large (several tens of µm) and mechanically rounded, or µm-scale aggregates composed of particles <1 µm to the nm-scale (e.g., Dietrich et al., 2019; LeGalley and Krekeler, 2013). Qualitatively, larger Pb particles tend to be more rounded, whereas smaller Pb particles tend to be more euhedral. Through entropic processes alone, one would expect fragmentation in a road sediment setting over time. Fragmentation is an important natural process in general, and recent comprehensive models suggest that there are inherent distributions of polyhedral shapes that arise from fragmentation (Domokos et al., 2020) The work of Domokos et al. (2020) indicates that for rocks, the average shape of fragments is cuboid.

Aggregate textures of Pb-rich particles documented in Dietrich et al. (2019) and in LeGalley and Krekeler (2013) are challenging to interpret but leave open the question as to whether some micro- to nanoscale Pb particles may be aggregating into larger particles through time. O'Shea et al. (2021b) investigated road sediment and soil from the Fishtown area of Philadelphia using SEM. Their results indicate that Pb particles are pervasive in both road sediment and soils, and that most Pb particles are approximately 0.1 to 10 µm in average diameter. However, many of these Pb particles also occur as irregular subrounded aggregates that are commonly 20 to 60 µm in average diameter.

Lead, or particles that contain Pb that are aggregates of µm and nm particles are of concern as they may shed smaller particles ($PM_{2.5}$, nanoparticles) simply through transport. Whether the textures observed by LeGalley and Krekeler (2013), Dietrich et al. (2019) and O'Shea et al (2021b) are simply inherent textures of larger Pb particles or are result of an aggregation process is unclear. Orthokinetic aggregation has been observed in geological metal systems (e.g., Saunders and Schoenly, 1995) yet this mechanism is implied to be primarily for colloidal systems (Hansen et al., 1999). The energy levels required for aggregation of this type seem to be low in road sediment environments. Although nanoparticle aggregation has been studied in the context of medicine and toxicology (e.g., Zhang, 2014), it appears that there are no studies on the aggregation of Pb metal particles in road sediment. The details of Pb particle dynamics and evolution should be studied more extensively in road sediment and in closed laboratory systems to determine the extent of the above observations and refine the understanding of processes relating to particulate Pb.

An additional key Pb phase observed by SEM in road sediment is $PbCrO_4$, which was widely used as pigment in yellow traffic paint (e.g., O'Shea et al. 2021a; White et al., 2014). White et al. (2014) investigated $PbCrO_4$ yellow traffic paint from locations in Hamilton, OH and found that these particles existed as micrometer scale aggregates of sub-micrometer to nanoscale crystals. O'Shea et al. (2021b) also observed some particles which are interpreted to be at least in part degraded $PbCrO_4$ yellow traffic paint from the Fishtown area of Philadelphia, similar to $PbCrO_4$ paint textures observed by White et al. (2014). White et al. 2014 suggested at least some of these particles dissolve, although O'Shea et al. (2021a) have shown experimentally that silica coatings applied to $PbCrO_4$ pigment may inhibit its dissolution. Thus, the extent of dissolution and the role $PbCrO_4$ nanoparticles play in Pb pollution in road sediment should be investigated further, as environmental processes interacting with $PbCrO_4$ may be complex. Whether or not $PbCrO_4$ still occurs widely in extant traffic paint, historic traffic paint, and adjacent soils and environments should be investigated broadly in the U.S. SEM surveys of traffic paint for $PbCrO_4$ are straightforward and should be undertaken in each state to assess the prevalence of $PbCrO_4$.

2.3.1.1.2 SEM investigations of zinc, copper, and other particles

Zinc and Zn-Cu alloys are commonly encountered particle type in road sediment (e.g., Flett et al. 2016; Dietrich et al., 2018). These particles usually exhibit a hackly fractured surface and texturally appear somewhat pitted, owing to oxidation or dissolution. These particles are commonly interpreted as being derived from anti-corrosion coatings of vehicles or potentially debitage from galvanized guardrails or other galvanized sources, however, other sources such as housing may also contribute (Davis et al., 2001). There are no clear comparatives to assess whether zinc observed in SEM is metal or an oxide, however comparisons of zinc oxide derived from the passive oxidation of batteries provide some constraint. Zinc oxide derived from the oxidation of zinc powder in batteries tends to be euhedral and prismatic in texture (Barrett et al., 2011), whereas particles interpreted as zinc metal are irregular in shape, have hackly fractures common of metals, and may or may not show inclusions.

Other metallic particles in road sediment are common, as LeGalley and Krekeler (2013) observed Ni fibers tens of µm in length as well as Cr-rich metallic shards through SEM observations. Additionally, they observed aggregates of W-rich material that was either W metal or tungsten carbide (WC). O'Shea et al. (2021b) identified fragments of steel, which was interpreted to be the very common ASTM A36 steel.

Technogenic spherules are a well-recognized pollutant component in the environment (e.g., Lue et al., 2016; Magiera et al., 2013; Magiera et al., 2011) and are also a common particle type encountered in road sediment. These particles are characteristically round but are diverse in composition and texture. Common textures observed include smooth glassy surfaces, glassy textures serving as a matrix for crystalline phases such as spinels or magnetite, and phases that appear to be dominated by crystalline material (LeGalley and Krekeler, 2013; Dietrich et al., 2018; Dietrich et al., 2019). LeGalley and Krekeler (2013) observed and interpreted images of technogenic spherules in different states of weathering, where some degraded spherules appeared to be altering to clay minerals. LeGalley and Krekeler (2013) also showed textures of some technogenic spherules that exhibited mechanical abrasion. Owing to the spatial association with a coal plant and comparison to textures documented in the literature, LeGalley and Krekeler (2013) interpreted these particles to be coal fly ash derived. Dietrich et al. (2019) observed a range of technogenic spherules from several 10s of μm to the nm-scale and concluded that these were derived from coal or steel processing. In that study it was also noted that many spherules contained detectable manganese.

2.3.1.2 TEM investigations

Few road sediment studies have utilized TEM because the application of this technique is time-intensive and because it is most commonly used for characterizing specific nanoparticles, minerals, or synthetic materials rather than for bulk assessment. However, TEM studies are common in PM_{10} and $PM_{2.5}$ research to characterize contaminants and emissions (e.g., Chen et al., 2004; Gieré et al., 2006), and to study their health effects (e.g., Wang et al., 2015).

LeGalley and Krekeler (2013) used TEM on road sediment and described pollutant metals adsorbed to clay particles. Specifically, they observed that Cu, Zn, Ni, and Hg likely remobilized, potentially in the street, and were adsorbed onto clay surfaces. Arrington et al. (2019) looked at a limited number of samples from Gary, Indiana and found spherule textures, oxides, and metal particles containing Pb, Mn, Ni, Cr at the nanoscale. The TEM work by O'Shea et al. (2021b) on Fishtown samples from Philadelphia suggests that Pb may have dissolved, remobilized and adsorbed or reprecipitated onto particles on the nano-scale. Their work also documents examples of $PM_{2.5}$ or nanoparticles that contain Pb. Collectively these textures indicate that Pb is reduced in particle size to some degree and also appears to have dissolved and reprecipitated or have adsorbed to mineral surfaces. The textures suggest that Pb is mobile physically and chemically in this complex system. Paltinean et al. (2016) used TEM along with XRD and SEM-EDX to document the presence of nano-sized quartz and clay particles in resuspended road sediment.

Ideally, TEM work should involve comparative or background materials to better understand the nature and influence of naturally occurring PM_{10} and $PM_{2.5}$. Such work aids in the interpretations of the sources of pollutant particles broadly and potentially helps distinguish the form and nature of metal content observed on natural Fe and Mn oxyhydroxides and those that may be remobilized in road sediment, thus providing constraints on geochemical and sediment transport processes. Where feasible, TEM should be included in future road sediment studies to ascertain the nature of nanoparticles, whether pollutant metals have adsorbed or mineralized, and to determine the form and potential source of metal pollutants. It is not feasible to do large numbers (e.g., n = 20 or 30) of samples for detailed TEM work, but it is suggested that ideally 1 representative background sample and three representative road sediment samples be investigated to complement every 20 to 30 study samples.

2.4 Toxicity, pollution spatial heterogeneity, and element mobility in the context of road sediment

Road sediment has been internationally recognized as a potential health risk to humans through multiple routes of exposure, including inhalation, dermal contact leading to chemical absorption, and ingestion (e.g., Ferreira-Baptista and De Miguel, 2005; Shi et al., 2011; Du et al., 2013; Bian et al., 2015; Men et al., 2018). Human exposure to road sediment is often explored due to its elevated metal contents relative to other media such as soil (Shi et al., 2011). Ingestion is typically the highest-risk pathway of road sediment (e.g., Dietrich et al., 2019; Shi et al., 2011). Children typically have higher ingestion rates than adults, primarily due to a greater frequency of hand-to-mouth activities (e.g., Needleman, 2004; Ko et al., 2007; Shi et al., 2011; Stewart et al., 2014), and are therefore at a higher risk for interaction with road sediment. In fact, children may ingest up to 60 g of soil each day and they are vulnerable to pollutants due to their developing bodies and brains (e.g., Van Winjen et al., 1990; Calabrese et al., 1997).

2.4.1 Potential health risks associated with metals found in road sediment

Several elements with typically elevated concentrations in road sediment relative to background materials, such as Cd, Cr, Cu, Pb, Zn, and Hg, can have adverse effects on humans if consumed in excess. However, it is noted that primarily chronic, long-term exposure is of greatest concern with road sediment and thus not acute exposure, and many metals often occur as various compounds in the environment with varying toxicity. Cd has been labeled as a carcinogen by the IARC (International Agency for Research on Cancer) and is a non-essential metal that has been linked to adverse effects on the skeletal (i.e., lowering bone density, Itai-Itai disease), respiratory, and reproductive systems, and causes kidney damage (e.g., Godt et al., 2006). Lead exposure can also be problematic, adversely affecting the central nervous system (e.g., Tchounwou et al., 2012 and the references therein). Additionally, studies have shown that chronic Pb exposure can harm the kidneys, negatively impact vitamin D metabolism, and that elevated blood Pb levels in children are linked to diminished IQ levels and growth inhibition (e.g., Tchounwou et al., 2012 and the references therein). Chromium toxicity is inherently tied to the oxidation states of Cr. Hexavalent chromium (Cr(VI)) has been linked to a variety of health effects such as ulcers, adverse respiratory effects, and renal damage, with multiple agencies considering Cr(VI) a human carcinogen (e.g., Pavesi and Moreira, 2020; Tchounwou et al., 2012). However, Cr(III) compounds appear to be comparatively less toxic (Tchounwou et al., 2012), as Cr(III) has also been documented as an essential micronutrient, although there is some recent debate surrounding this point (Pavesi and Moreira, 2020 and the references therein). Although road sediment tends to accumulate in oxidizing surface environments, previous work on road sediment from an urban center found Cr(III) to be the predominant species, likely because of Cr(III) being prevalent in basic oxygen furnace steel slag and other industrial by-products captured in urban road sediment (Byrne et al., 2017). Thus Cr(VI) may not be as prevalent in road sediment in certain urban environments, although specific speciation studies are needed to discern this. However, Cr(VI) may still be introduced to road sediment from a variety of anthropogenic sources such as tanneries or steel facilities (e.g., Tchounwou et al., 2012; Welling et al., 2015), or $PbCrO_4$-based road paint in some areas (e.g., LeGalley et al., 2013; O'Shea et al. 2021a).

Mercury is studied much less in road sediment compared to other elements such as Pb and Zn (Fig. 4). Mercury is a highly toxic metal, which has many potential adverse health effects resulting from low-dose exposure, including but not limited to decreased muscular strength, memory loss, decreased fertility, and compromising the immune system (e.g., Zahir et al., 2005). Although Hg is highly toxic, most road sediment studies in the U.S. thus far have likely omitted Hg from their analyses because more specialized instrumentation is often needed (e.g., cold-vapor atomic absorption spectroscopy (AAS)) as opposed to the routine ICP and AAS techniques often employed (Table 3). Importantly, Hg in road sediment may be increasingly mobilized following stormwater runoff to aquatic ecosystems, where methylation of Hg may occur.

Both Cu and Zn are common metals in road sediment and, unlike other metals discussed, are well-documented essential micronutrients, where deficiency in uptake of either metal can result in adverse health outcomes for both humans and other organisms (e.g., Gaetke and Chow, 2003; Plum et al., 2010). However, chronic excessive intake of Cu can result in toxic effects such as liver cirrhosis, particularly because the liver is the first organ to accumulate Cu (Gaetke and Chow, 2003). Zinc toxicity is fairly low, with the main concerns of overexposure related to chronic elevated exposure, which inhibits Cu uptake (Plum et al., 2010). There is more concern related to Zn deficiency in diets as opposed to Zn toxicity (Plum et al., 2010).

While some studies did not report high carcinogenic risks from elements in road sediment, they did note significant non-carcinogenic risks (e.g., Bartholomew et al., 2020). Lead is often the most hazardous element present in road sediment (e.g., Jayarathne et al., 2018). As mentioned above, ingestion is the most dangerous pathway for interaction (e.g., Ferreira-Baptista and De Miguel, 2005). Studies have demonstrated that oral bioaccessibility must be taken into account to understand real health risks of pollutants, particularly for Pb (Elom et al., 2014). While inhalation is less common, and more often studied with PM, key studies have explored the inhalation of PM and dust with simulated lung fluids such as Gamble's solution (e.g., Caboche et al., 2011), in-vivo lung fluids (e.g., Stebounova et al., 2011) and neutral-pH synthetic epithelial fluid (e.g., Dean et al., 2017; Kastury et al., 2017).

2.4.2 Spatial variability of potentially health-relevant pollutants

Previous assessments have outlined very extensive spatial variability of metal contamination in road sediment, which results in some locations being high risk whereas other areas are not (e.g., Decampo et al., 2012; O'Shea et al., 2020). However, in general, exposure to metals in urban settings is higher than in suburban settings (Shi et al., 2011). Furthermore, urban road sediment may have finer-sized particles than in non-urban settings and thus may present higher risks from elevated levels of metals (Pb, Cd, Cu, Zn, Ni, and Cr) (e.g., Shi et al., 2011). For risk assessment, previous studies indicated that size-fractionation is crucial because the finest size fraction generally has the highest concentration of bioaccessible metals (e.g., Padoan et al., 2017). In regard to land use, a recent study reported that Pb bioaccessibility was highest in residential areas and lowest in gardens, whereas Cd bioaccessibility from road sediment was highest in parks and residential areas (Zhang et al., 2020). Another similar study described the highest risk from metals (Pb, Cu, Zn, and Ni) in road sediment in tourism areas as opposed to residential, education, or high traffic-density areas (e.g., Wei et al., 2015), whereas a different assessment reported the highest metal (Pb, Cu, Zn, Co, V, Al, Ni, Cr, Cd) concentrations in industrial areas (e.g., Li et al., 2013). Indeed, a better knowledge of the spatial distribution of road sediment pollution and urban pollution in general is crucial to better understanding the risk it presents to human populations (e.g., O'Shea et al., 2020; O'Shea et al., 2021c).

2.4.3 Environmental mobility of elements found in road sediment

Another important aspect of element toxicity, and directly related to bioaccessibility, is environmental mobility. Duong and Lee (2009) utilized partial sequential extraction and described that in the carbonate and exchangeable fractions, the most mobile element was Cd, followed by Zn, Pb, Cu and Ni, respectively. Other road sediment studies also documented Cd to be the most mobile element, followed by Zn, Pb, and Cu (e.g., Li et al., 2001). Similar results were detailed by Yildirim and Tokaliglu (2016). However, a different study in Shiraz, Iran outlined that the most mobile elements were Pb and Hg, followed by Zn, Mn, Cu, Sb, Ni, Cr, and Fe (Keshavarzi et al., 2015).

A factor that may influence element mobility is particle size (e.g., Jayarathne et al., 2019; Jayarathne et al., 2018). Adsorption capacity may be lower for coarser particles compared to finer particles, which impacts mobility based on size fractionation (e.g., Jayarathne et al., 2019). The type of particle must also be taken into account. Furthermore, previous investigations found that land use and antecedent dry days influenced the variability associated with the adsorption of Zn, Cu, Pb, Cd, Cr and Ni where initial dry days after storms had a stronger influence on adsorption compared to later dry days for all land use types (Jayarathne et al., 2018). Overall, multiple factors such as sediment mineral contents, grain size, element speciation, and desorption/sorption reactions collectively affect element mobility. Variation in road sediment element mobility is important to document and research, because of possible adverse effects on nearby ecosystems and waterways.

2.5 Health risk assessment of road sediment pollution

A variety of risk assessment models have been employed to better understand the hazard of human exposure to road sediment. Common calculations include non-carcinogenic hazard quotients (HQ) and carcinogenic risk (e.g., Zhang et al., 2020). Other calculations include the average daily dose (ADD) (mg/kg/day) of metals from soil ingestion, inhalation and dermal contact (U.S. EPA, 1989; U.S. EPA, 1996). A potential ecological risk index (RI) for non-human biological organisms, originally developed by Hakanson (1980), is also commonly used with sediments (e.g., Bian et al., 2015; Men et al., 2018; Zhao and Li, 2013). HQ and hazard index (HI) are widely used in road sediment studies to assess human health risk (e.g., Bourliva et al. 2016; Chen et al., 2019; Dietrich et al., 2019; Du et al. 2013; Zheng et al., 2010), and will thus be discussed in greater detail here. Additionally, because of inconsistencies regarding carcinogenicity documentation for several inorganic elements and frequent lack of known chemical speciation in road sediment, a discussion of carcinogenic health risk assessment will be avoided here (e.g., slope factor implementation).

HQ values are based on calculations of average daily ingestion exposure (E_{ing}), chronic/subchronic inhalation exposure (EC_{inh}), and dermal absorbed dose (DAD), where HI is the summation of HQ values for ingestion, inhalation, and dermal exposure, with each individual exposure pathway represented as "i" (Eqs. 4-7) (U.S. EPA, 1989, 2001, 2004, 2009):

$$HQ_{ing} = E_{ing}/RfD \tag{4}$$

$$HQ_{inh} = EC_{inh}/(RfC_i \times 1000\,\frac{ug}{mg}) \tag{5}$$

$$HQ_{derm} = DAD/RfD_{ABS} \tag{6}$$

$$HI = \Sigma \mathrm{HQi} \tag{7}$$

HQ values >1 represent a greater potential for adverse health effects to occur in the human body, whereas an HI value >1 represents an increased likelihood that non-carcinogenic adverse health effects will occur (U.S. EPA, 1989, 2001). While some studies of environmental media tend to calculate HI through summation of HQ values for each element (e.g., Ogunlaja et al., 2019; Roy et al., 2019), we caution against this, as additive effects of various inorganic elements can greatly complicate risk evaluation. Reference doses (RfD or RfC_{i} for inhalation) are essentially a baseline of exposure to compare against and are specifically an estimation of maximum acceptable exposure that will likely not lead to the development of adverse health effects (U.S. EPA, 2002). The reference doses are very important variables in determining risk assessment, because misuse can easily lead to an over- or under-estimation of risk. Thus, even if researchers are using the same risk assessment equations, input of variables' values into the equations must be explicitly defined, as this can confound comparative risk assessment between studies. Furthermore, caution must be employed when doing health-based risk assessment with bulk concentrations of potentially harmful heavy metals and metalloids, because speciation and bioavailability play an important role in determining risk. In general, research has shown road sediment to particularly affect the respiratory system when resuspended into the air, although it is suggested that more holistic health-based risk assessment studies are needed to understand the impacts road sediment may have on human health (Khan and Strand, 2018). This is particularly important because risk assessment studies of road sediment typically find ingestion to be a major exposure pathway as opposed to inhalation (e.g., Dietrich et al., 2019; Ferreira-Baptista and De Miguel, 2005; Shi et al., 2011), but often do not examine what specific health effects are induced by road sediment exposure.

2.6 Road sediment metal pollution source apportionment

Metal pollution source apportionment in road sediment studies commonly involves multivariate statistical methods such as PCA, factor analysis, and hierarchical clustering (e.g., Mummullage et al., 2016; O'Shea et al., 2020) to better differentiate potential sources of heavy metal pollutants. Additionally, tools such as SEM and metal isotope analysis have also been applied to help support pollutant source interpretations (e.g., Teran et al., 2020; Sutherland et al., 2003). While these attempts in source apportionment are important for gaining insight into the best possible mitigation and remediation measures for metal pollution, the ubiquitous nature of metal pollution in many urban settings makes definitive sourcing increasingly difficult.

Whether it be statistical interpretations of metal pollutants in road sediment via bulk chemistry, or metal isotopic ratios (e.g., for Pb), it is important not to oversimplify assumptions of pollution sources. For example, it has been shown that using only one multivariate statistical method for pollution source apportionment can result in deficiencies (Mummullage et al., 2016), and that there can be significant overlap in Pb isotopic ratios within one urban system (e.g., Dietrich and Krekeler, 2021). Thus, it is imperative that multiple analytical techniques or more sophisticated statistical models (i.e., Bayesian mixing models) be utilized to better differentiate pollution sources in road sediment, or that the relative uncertainty is clearly documented. One simple example is that of Cu and Zn, which can be grouped together in various forms of multivariate factor analysis, but can come from one of two primary, yet vastly different sources in the urban environment—vehicles (tire/brake wear) or housing (roof/siding) (Davis et al., 2001).

2.7 Remediation efforts for road sediment pollution

Environmental regulation within countries can have a large effect on pollution concentrations in environmental media, such as road sediment. However, while environmental regulations in the U.S. over the past 40-50 years have reduced levels of metal input into the environment, such as Pb (e.g., Hwang et al., 2016), recent studies of road sediment in the U.S. have still found concentrations of several heavy metals, including Pb, Zn, and Mn, greater than background values and sometimes approaching levels of health concern (e.g., Dietrich et al., 2019; O'Shea et al., 2020) (Table 3; Fig. 2). Thus, even with enhanced regulation, many of these metals still pose a threat to both humans and other biota either through stormwater runoff or particle resuspension into the atmosphere. However, it is promising that in studies involving U.S. road sediment collected post-1990, Cd and Pb concentrations are clearly lower than concentrations reported by studies where sampling was conducted pre-1990 (Fig. 2). Samples collected prior to 1990 were closer to peak Pb gasoline usage in the U.S. and were higher in Pb (Fig. 2). Hwang et al. (2016) also observed the same temporal differences for Pb in their comprehensive global road sediment analysis, as did Haynes et al. (2020). While our observed differences may in part be due to better analytical accuracy in modern studies and some spatial variation, a large proportion of the heavy metal decline is likely attributed to better regulation, remediation, and technological advancements. Thus, moving forward, enhanced technology (i.e., green roofs, less metal wear from vehicles), remediation efforts, and regulations that limit pollution from sources such as vehicles or industrial facilities will hopefully aid in further lowering the anthropogenic footprint of various metals and metalloids in road sediment, with the aim of reaching concentrations closer to "background" values.

A number of studies have investigated the impacts of street sweeping and vacuuming to remove road sediment particles as a pollution remediation tool. These quantitative studies generally reported that coarser particles were more efficiently removed and that vacuum assistance, in combination with flushing can be effective (e.g., Amato et al., 2009; Ang et al., 2008; Clark and Cobbins, 1963; Duncan et al., 1985; Minton et al., 1998; Pitt and Amy, 1976; Pitt, 1979; Sartor and Boyd, 1972; Selbig and Bannerman, 2007). Amato et al. (2012, 2013) documented that after rain events, the mobile dust load (road sediment particles <10 µm), on average, returned nearly to pre-rain PM_{10} concentrations after 24-72 hours. A focus of research has been on the impact of street sweeping on air quality, testing a variety of methods and parameters (e.g., Chou et al., 2007; Chow et al., 1990; Düring et al., 2007; Gertler et al., 2006; Kantamaneni et al., 1996; Karanasiou et al., 2011; Karanasiou et al., 2012; Kuhns et al., 2003). Overall, many studies did observe a reduction of atmospheric particles, typically PM_{10}, from road sediments after either the application of brooms, vacuums, water flushing, sweepers or a combination of techniques. However, the results were not uniform and were tested using a variety of methodology. Ultimately, for high loadings, the use of vacuuming followed by washing is recommended (Airuse, 2013). However, washing may adversely impact stormwater quality and there may therefore be a trade-off between preserving air quality or water quality. Lastly, the removal of solids from the finest fraction, in an early dry state, is expected to assist in stormwater mitigation measures to reduce the amount of released metals into metropolitan environments (Jayarathne et al., 2018). Therefore, street sweeping must be considered.

Green roofs have potential for PM removal from the atmosphere (e.g., Speak et al., 2012; Yang et al., 2008), which would otherwise end up in road sediment. Additionally, trees in urban settings show promise for removing PM from the atmosphere (e.g., Bealey et al., 2007), thus reducing road sediment loading. While the high cost of implementation for green roofs serves as a barrier for development (Yang et al., 2008), the higher availability of surfaces for green roofs in urban settings as opposed to limited space for tree planting makes it desirable for future development (e.g., Yang et al., 2008). Additionally, green roofs reduce stormwater runoff (e.g., Ahiablame et al., 2012 and the references therein), which influences road sediment particle transport.

Vegetated highway medians in Texas, U.S. have been shown to be effective in reducing road sediment runoff to waterways (Barrett et al., 1998). Vegetative swales in general have good capacity for lowering stormwater runoff pollution (e.g., Ahiablame et al., 2012 and the references therein). Retention ponds, including bioretention systems, and permeable pavements have also led to a reduction of stormwater runoff parameters such as total suspended solids (TSS) (e.g., Ahiablame et al., 2012 and the references therein), which would include road sediment pollution particles and thus help protect aquatic systems. This area has been well investigated (e.g., Gupta and Saul, 1996; Sansalone and Buchberger, 1997b; Lau et al., 2002; Ma et al., 2002), and Li et al. (2006) recommended that capturing the first 20% of runoff, by volume, would potentially remove 40% of the total particulate load (from calculated particle mass). This would remove a majority of the metals investigated, and thus lead to reduced deposition in stormwater. Additionally, when preparing stormwater treatment protocols, an author recommended that particle size data be measured and accounted for as different metals were found to be associated with different particle size ranges (Tuccillo, 2006); as an example, Pb and Cr were primarily associated with the >5 μm size fraction.

Lastly, pollution remediation directly at the source is needed to reduce the impacts of road sediment pollution. This essential strategy is emphasized by Hwang et al. (2016), who noted phasing out of leaded gasoline as an example of Pb reduction in road sediment, which we also notice in our literature review of U.S. road sediment (Fig. 2). However, Pb is still being emitted to the environment through means such as leaded wheel weights in vehicles (e.g., Ayuso and Foley, 2020; Hwang et al., 2016) and $PbCrO_4$ road paint (e.g., LeGalley et al., 2013; White et al., 2014; O'Shea et al., 2021a). While steps have been taken to reduce the prevalence of leaded wheel weights in the U.S., only nine states currently ban its use (Ayuso and Foley, 2020). $PbCrO_4$ derived from yellow traffic paint is also still prevalent in many urban environments, even if application has been discontinued in certain areas in recent years. Thus, further removal of Pb from road sediment and the environment can be achieved through continued phasing-out of leaded wheel weights and $PbCrO_4$ paint. Additionally, while agreements between the U.S. EPA and automobile industry aim to reduce Cu, Cd, Pb, and other materials in brake pads through a series of initiatives up to 2025 (Hwang et al., 2016 and the references therein), Zn is largely sourced from tire wear (e.g., Davis et al., 2001; Hwang et al., 2016). Thus, improvements in tire technology to reduce tire degradation may help diminish Zn input into road sediment and the environment, although the usage of Zn as a vulcanization agent will likely persist in tires for the foreseeable future and the proportion of Zn from tire wear will likely increase as exhaust emissions are reduced.

3.1 Future directions

Owing to the complexity of sources and processes involved in the redistribution/modification, retention, accumulation, and evolution of road sediment (Table 2), there is great need for future investigations. This includes simply more investigations to assess both the variation and the extent of road sediment pollution, as well as specific studies to apply techniques to answer fundamental questions. While several recommendations to improve road sediment research are alluded to above, important future research directions are explicitly discussed below. Several are in agreement with recent recommendations at a global scale by Haynes et al. (2020), such as closer examination of other pollutants like polycyclic aromatic hydrocarbons (PAHs), better characterization of "background" concentrations, and greater usage of microscopy as a technique to categorize road sediment.

3.1.1 Increase the number and diversity of studies

Studies in more diverse settings are needed. For example, it was clearly shown that there are limited road sediment studies in the Mountain West region (WY, UT, CO) of the U.S. (Fig. 3), which is also reflected in minimal Pb studies of other environmental media in the U.S. Mountain West such as water, soil and air (Frank et al., 2019). New investigations should not only evaluate large and mid-sized cities, but also small towns and rural sites. In addition, future investigations should juxtapose demographically diverse sampling areas to help elucidate possible environmental justice issues. Doing so will enable a better understanding of the variation in pollutant materials, will help analyze the extent of elevated concentrations of hazardous elements, and foster fair development of environmental policies across a spectrum of settings. Furthermore, these investigations should have a wide geographic or physiographic spread. Ideally, in the U.S., there should be at least a few studies representing each state. By conducting such investigations throughout the U.S., a rigorous analysis to determine how road sediment compares and relates to overall community health will be possible. The context of road sediment studies provides an opportunity for broader community involvement such as through community science initiatives. This can potentially serve to generate interest in the environmental health of communities, including those that may be socioeconomically disadvantaged or lack access to adequate health resources. Due to the diverse industrial history, climate, and social dynamics throughout the U.S., study locations in the U.S. can serve as effective roadmaps to assess pollution in many different international locations that have analogous industrial history and variation in socioeconomic status. Such approaches will not be straightforward, and a standardized approach for sampling and reporting road sediment data in a community should be developed, as standardized approaches are essential in developing a central system where data can be synthesized and shared effectively (Frank et al., 2019).

3.1.2 Detailed geogenic and pedogenic background studies

There is a major need for extensive investigations that produce well characterized background geological (geogenic) and pedological (pedogenic) material. Such investigations by their nature should be data-intensive and describe the variation of bedrock and soils in sufficient detail. Examples of such investigations include Barnes et al. (2020) and Oglesbee et al. (2020), where a major component of the environment is characterized. Establishing background has always been a challenge in environmental investigations, but it is particularly challenging in road sediment pollution studies owing to numerous sources, settings (e.g., urban vs. rural), and processes involved. We suggest that detailed studies of background geogenic and pedogenic materials continue and be made a priority for key U.S. locations such as Gary, IN, Hamilton and Middletown, OH, and Philadelphia, PA, as well as future locations throughout the world where researchers may carry out multiple studies over time.

3.1.3 Investigation of apparently underrepresented inorganic (Hg, Tl, radiological and asbestos) components.

Compared to all other metals, Hg is an underrepresented analyte in U.S. road sediment (Fig. 4), which is surprising given the overall ubiquity of Hg in the environment and the very well-established toxicity of Hg (e.g., Zahir et al., 2005). We strongly advocate for detailed investigations of the bulk concentration and form of Hg in road sediment, with particular attention to the spatial distribution of potential sources, such as coal power plants and heavy manufacturing sites.

Tungsten is also a metal that is very much underreported in road sediment studies yet has long been recognized for its use in machining and numerous other aspects of industry (Rieck, 1967). LeGalley and Krekeler (2013) observed particles interpreted to be either W metal or WC that were micrometer to near nanoparticle in scale in road sediment of Hamilton, OH. Tungsten is potentially of interest in road sediment near industrial areas owing to its use in tooling, cutting and abrasion. Shepard et al. (2007) and Shepard et al. (2012) investigated the distribution of W in Fallon, Nevada in the context of leukemia clusters.

There are also metals/metalloids that are not commonly analyzed in road sediment that may be of high interest geographically or in site-specific contexts. These include Tl, which is well recognized for its toxic properties and was used extensively as a pesticide (Clarkson, 2001). Arsenic is also an element of major concern in general, but specifically in chicken-farming communities (Burros 2006; Sambu and Wilson 2008) and older brass-manufacturing sites (Garelick et al., 2009; Reddy, 2016) due to the pervasive use of As. When possible, it is recommended that researchers analyze for all possible contaminants as the exact environmental history of sites are often not known. Not all buildings or sites have an open or accessible history, and for example, radioactive pollutants can exist and be unrecognized (e.g., Foley and Floyd, 1990). Certainly, settings exist where past radiological contaminants were known to occur on a site but may not have been investigated off the site, where release may have occurred during transportation of the radiological material (e.g., Pourcelot et al., 2011; Gieré et al., 2012).

One particle type that appears to not be studied extensively in the context of road sediment is asbestos. Asbestos was used extensively in automotive brake pads and linings and other parts (Van Gosen, 2008). The nature and extent of asbestos in road sediment appears to not be studied in the U.S. Although this material has largely been removed from current production of automotive parts, there are older components likely in use that contain asbestos. Asbestos most likely is of more significance in older sediments. However, we note that there may be localized sources of asbestos in road sediment that may be associated with past construction, asbestos production, asbestos removal and product transportation. Asbestos in road sediment is perhaps the most uncertain pollutant component and there is great need to understand its presence in historic and current contexts as road sediment can be remobilized.

Buck et al. (2013) conducted a survey of naturally occurring asbestos in areas of southern Nevada with a total of 43 samples. This study was primarily a SEM investigation supported by other techniques that detected actinolite asbestos. Six samples were obtained from dirt roads and one sample from a vehicle tire. All samples of rock, soil, dust and car tire contained fibrous asbestos. This asbestos is derived from natural sources such as the Miocene Plutons in the McCullough range, Black Hill and Boulder City. The region that is potentially impacted by this naturally occurring asbestos includes Boulder City, Henderson, and the greater Las Vegas area, owing to recognized dust storms (Buck et al., 2013). It is known that asbestos fibers become airborne through both natural erosion processes and human actions that produce dust, such as mines, quarries, roads, and outside activities (Bauman et al., 2013). Bauman et al. (2015) documented that, compared with the United States as well as other Nevada counties, southern Nevada had a significantly higher proportion of malignant mesotheliomas that occurred in young individuals (<55 years in age) as well as in women. Bauman et al. (2015) concluded that the presence of naturally occurring asbestos in southern Nevada contributes to mesothelioma in the region.

The nature and extent of occurrence of asbestiform actinolite in road sediment throughout the southern Nevada area is not quantified. Asbestos surveys of road sediment may however provide further constraints on this specific issue of naturally occurring asbestos in the region. Broader investigations of road sediment near locations where asbestos products have been processed may also be of interest to document dispersal and assess possible health risks.

3.1.4 Improving geochronology of road sediment

The question of road sediment age is a major one, as this relates to the parameter of retention and the potential for an understanding of legacy contaminants, such as Pb from gasoline. Numerous techniques are available that should be applicable, including ^{137}Cs, ^{210}Pb, and optically-stimulated luminescence methods (e.g., Appleby, 2008; Madsen and Murray, 2009; Arias-Ortiz, 2018). To our knowledge, there are no quantitative studies on the age distribution of road sediment. Thus, how long pollutants persist in the environment within road sediment in different settings is unclear. We suggest researchers move forward in this area using comparative approaches, such as short sedimentary cores of thick accumulations of road sediment in potholes, drainage or other features as much as possible. This, combined with parallel investigations of adjacent sediment, pond or catchment cores could potentially capture a near equivalent environmental record. Such environments may be challenging to identify and may not be common or widespread. Disturbance and resuspension are processes that likely complicate chronology. Ideally, investigations would link road sediment and traditional environmental media not only in geochronological methods, but in bulk chemical analysis as well as electron microscopy and isotopic methods. Gary, IN is just one example of a potentially ideal location for such work based on the results of Dietrich et al. (2019) and the numerous small waterbodies immediately east of their study area.

3.1.5 Closer examination of barite

Barite is common in some road sediment samples and usually occurs as discrete multi-µm to sub-µm subhedral crystals, as identified via SEM or TEM (e.g., LeGalley and Krekeler, 2013; Yang et al., 2016). Multiple sources of barite exist in road sediment. It is recognized that the combustion of coal and diesel and the incineration of waste releases barium particulates to the atmosphere (ATSDR, 2007). Other sources of barite include automobile brake and clutch components (USGS, 2019) as well as commonly used pigments (Zhou et al., 2015). Barite may be useful for estimating contributions of different pollutant reservoirs, provided chemical fingerprints of barite can be elucidated. Such investigations would not be without technical challenges, primarily because of the small particle size of barite, which may limit or prohibit some isotopic analyses such as S isotopes by secondary ionization mass spectrometry (SIMS). However, mineral separation and concentration of barite may yield reasonable Sr isotope and trace element signatures for this mineral. Provided reservoir samples can be investigated, this may be promising for comparative analysis in proper context.

Barium concentrations are much less than UCC in glacial till (about half, Barnes et al., 2020) and lower than UCC in some sand dunes samples (Oglesbee et al., 2020). Thus, although Ba may be depleted in bulk road sediment composition in some locations relative to UCC, Ba may still have some anthropogenic sourcing because of Ba-depleted background sources. Generally though, Ba is of lesser concern than other components of road sediment such as Pb, Zn, Cu, and Cr.

3.1.6 Nanoparticles in road sediment and multi-analytical studies

Nanoparticle pollution is a growing concern (e.g., Gao et al. 2015; Zhiqiang et al. 2000) and connections of nanoparticles to transportation systems is also recognized (Kumar et al. 2011a; Kumar et al., 2011b). The nature, distribution and processes associated with nanoparticles in road sediment are not well defined. Assessing the nature and extent of nanoparticles in road sediment will better delineate processes and relationships such as pollution source apportionment, pollutant relationships to clays, the behavior of nanoaggregates, the mobility of heavy metals on roads, and their morphology.

Future work should also consider utilizing isotopes, microscopy, and multifactor analyses (i.e., PCA) in tandem to source the pollutants found in road sediments. Specifically, while Pb isotopes have been useful for pollutant source apportionment in road sediment (e.g., LeGalley et al., 2013; Sutherland et al., 2003), recent advancements in Zn and Cu isotope analyses can aid in connecting road sediment with associated sources such as tires, road paint and combustion processes (e.g., Borrok et al., 2010; Dong et al., 2017; Souto-Oliveira et al., 2019). Recent work in Europe has further shown the utility of detailed, multi-analytical approaches when trying to understand cycling of road sediment and other materials in the environment (e.g., Gaberšek and Gosar, 2021; Kelepertzis et al., 2021). These combined approaches, as well as nanocharacterization, would make road sediment pollution sourcing more reliable and quantitative in future research endeavors.

In summary, nano- and microparticles in road sediment are understudied despite their clear risks to human health due to their potential for resuspension and inhalation. Both TEM and SEM are underutilized tools that could assist with pollution source apportionment and elucidate the links between atmospheric PM and road sediment. Additionally, SEM and TEM should be more heavily utilized alongside other geochemistry analytical techniques such as ICP-MS/ICP-OES and high-resolution mass spectrometry for metal isotopes.

3.1.7 PAHs and other contaminants of potential concern in road sediment

Thousands of persistent organic pollutants (POP) are recognized (e.g., Jones and de Voogt, 1999), which may also end up in road sediment. Some examples of these pollutants include important classes of POP chemicals, and many are families of chlorinated (and brominated) aromatics, including polybrominated diphenyl ethers (PBDEs) and organochlorine pesticides (e.g., DDT and its metabolites, toxaphene, chlordane, etc.), polychlorinated biphenyls (PCBs), polychlorinated dibenzo-p-dioxins, and furans (Jones and de Voogt, 1999). Other organic pollutants are also well recognized including benzothiazoles (e.g., Kloepfer et al. 2005 Seiwert et al. 2020), benzene, toluene, ethylbenzene, and xylene (BTEX) (e.g., Lovley, 1997), chlorinated solvents (e.g., Rivett et al. 2006), and dyes (Cymes et al., 2021), among many others (e.g., Nzila, 2013).

One organic molecule pollutant group of particular concern is polycyclic aromatic hydrocarbons (PAHs). PAHs are a class of stable organic molecules made up of two or more fused aromatic rings. In urban environments, PAH concentrations are elevated in dusts deposited on impervious surfaces, including road surfaces and roofs (Boonyatumanond et al., 2007, Zhao et al., 2009). Sizeable quantities of PAHs can be transported into local surface waters or retention ponds, thus representing a considerable risk to aquatic life (Schiff et al., 2003). Sources of PAHs in road runoff include lubricating oils and exhaust from diesel and gasoline vehicles, as well as tire-and road-wear particles (e.g., rubber, asphalt, bitumen). It is long recognized that road traffic is a major source of PAHs (e.g., Benner et al., 1989). Contribution of PAH sources to the environment varies. For example, Christensen and Arora (2007) investigated seven box cores of sediment approximately 13 cm in depth from central Lake Michigan for PAH apportionment and found 45% of PAHs were derived from traffic, 20% from wood burning, and 35% from coke oven emissions. Certain PAHs are well established as being carcinogenic and are U.S. EPA priority pollutants. The U.S. EPA has listed 16 PAHs as priority pollutants (U.S. EPA, 2021).

The topic of organic molecule pollutants in the context of road sediment is inherently complex, as road sediment can be a source and sink. Thus, a separate, detailed review of organic pollutants in the context of road sediment is warranted. Specifically, enough information appears to exist for PAHs that the diversity, concentration, and evolution of PAHs in road sediment should be looked at systematically. It is recommended that PAHs be reviewed in detail, followed by other groups of organic pollutants. Doing so will enable researchers to view the pollutant through the lens of source, degradation, adsorption and other properties.

4. Summary

Road sediment in the U.S. is an understudied medium compared to air, water, and soil. A body of literature exists on this topic, where there is a range of basic to advanced understanding of inputs, redistribution/modification, retention, and outputs. Furthermore, there is a wide variety of sampling and analytical methodology that has been employed. Due to the large breadth of diversity in social, industrial, and climate parameters within the U.S., advancements of road sediment research can prove informative for other regions around the world. Despite this, the U.S. is poorly geographically represented in road sediment research, with rural areas and regions such as the Mountain West being relatively neglected thus far.

Key techniques such as electron microscopy have been employed to assess anthropogenic particles in road sediment but currently remain underutilized compared to common bulk chemical techniques. Electron microscopy has extensive potential to provide detailed pollutant characterization information. Overall, electron microscopy should be combined with common techniques such as bulk chemistry analyses in addition to isotopic analyses and mineral phase determination. Some studies have already effectively used these techniques to provide insights into the sourcing and behavior of pollutants in road sediment.

Given the vast array of road sediment studies covered in this review, we call for unity in future road sediment research sampling methodology and data reporting, recently emphasized by international researchers as well (Haynes et al., 2020). Ensuring that studies are readily comparable and utilize similar methods for collection, size fractionation, analytical analysis, and risk assessment will help the research field advance. The same can be said for utilizing one term to refer to this medium rather than the variety that has been used in the past, as this will narrow literature searches.

Several heavy metal(loid)s of significant human and environmental concern such as Hg and As are vastly underreported in U.S. road sediment literature, thus offering a future research gap that should be filled. Organic pollutants are also understudied and there should be future focus on numerous organic compounds as well with priority to the most toxic. A key component of assessing pollution is also the detailed identification of proper background material in order to properly contextualize results. Furthermore, we recommend the careful determination and application of health risk assessment models in addition to metal(loid) enrichment calculations.

There is a great need for interdisciplinary involvement in road sediment studies and tremendous opportunities exist for collaboration with epidemiologists, public health professionals, biologists, materials scientists, geologists, and community scientist projects. Only through technological advancements, transparent and consistent terminology and methodology, and interdisciplinary holistic research approaches will future studies in this field both prosper and improve pollution remediation efforts.

Acknowledgements

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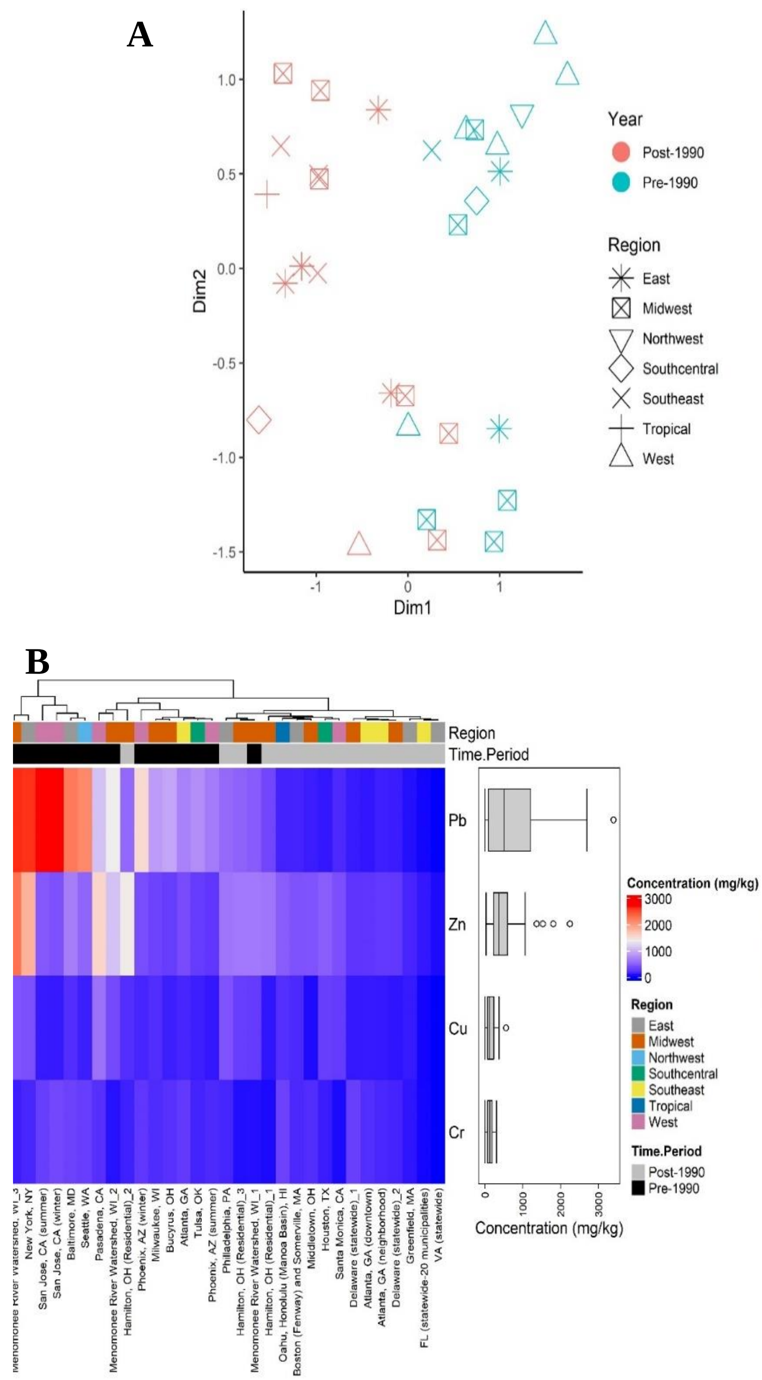

Figure 1: (A) Multidimensional scaling (MDS) plot using Aitchison distance to plot similar points closer together based on Cr, Pb, Zn and Cu. Data is from Table 3, and studies were only included if all four elements were measured and reported (n = 31). O’Shea et al. (2021b) was omitted because it was a subset of Philadelphia, PA, which was more broadly sampled in O’Shea et al. (2020). Samples labeled according to approximate climatic region in the U.S. (tropical = Hawaii), and identified according to the timing of sampling as well. (B) Heatmap of element

concentrations in mg/kg for all samples used in the MDS plot. Dendrograms are given for groupings on both axes, using complete linkage and Euclidean distance. Boxplots showing distributions of element concentrations are also provided (the boxes represent the interquartile range (IQR) of 25-75 percentiles of data, the horizontal line within the box represents the median, and the whiskers represent 1.5 times the IQR). Notice Pb concentrations noticeably decreasing from left to right across the heatmap, matching a change in sampling time period from pre-1990 to post-1990 samples.

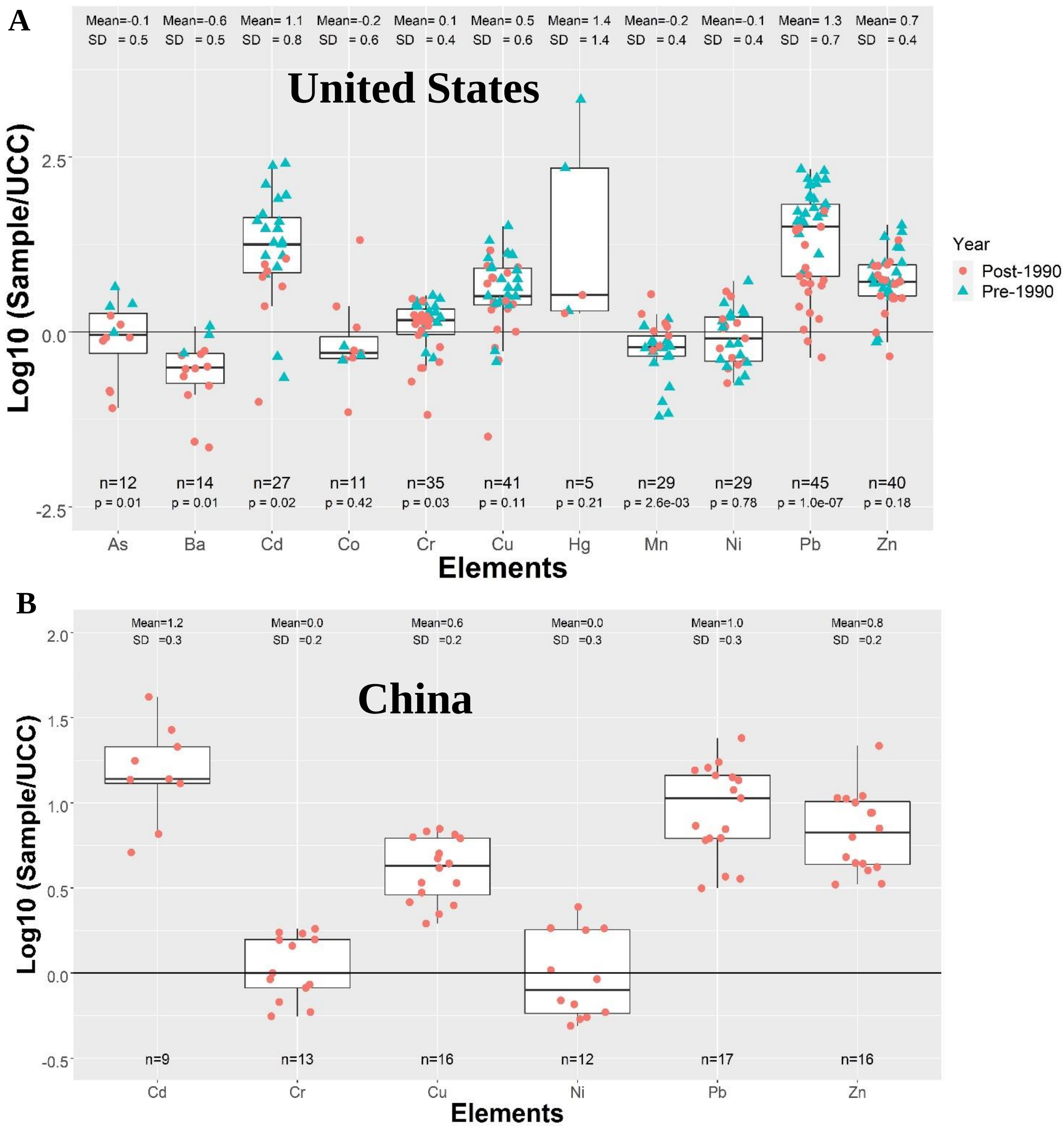

Figure 2: (A) Box-and-whisker summary plot of Table 3 normalized to upper continental crust (UCC) (Rudnick and Gao, 2003). The median Pb value of the range given in Franz and Hadley (1981) was used. Sutherland et al. (2003) and Andrews and Sutherland (2004) were excluded to avoid overrepresentation of Hawaii road sediment data from the same time period in the same location, as well as O'Shea et al. (2021b) because of more comprehensive Philadelphia data already gathered from O'Shea et al. (2020), but otherwise all reported U.S. road sediment data is included. (B) Box-and-whisker summary plot of select road sediment data from China post-1990

(Bi et al., 2018 and the references therein; Men et al., 2018 and the references therein; Pan et al., 2017 and the references therein; Wei and Yang, 2010 and the references therein). All values above 0 in both figures are enriched relative to UCC. The boxes represent the interquartile range (IQR) of 25-75 percentiles of data, the horizontal line within the box represents the median, and the whiskers represent 1.5 times the IQR. Number of samples (n), means, standard deviations, and two-sample t test p-values between the post- and pre-1990 sample groupings of the log normalized values for U.S. data are also provided.

Figure 3: Approximate locations of U.S. road sediment studies conducted thus far, where each point represents one study, except for multiple studies in Hawaii (n = 5), Philadelphia, PA (n = 2), Hamilton, OH (n = 3), and Urbana, IL (n = 2).

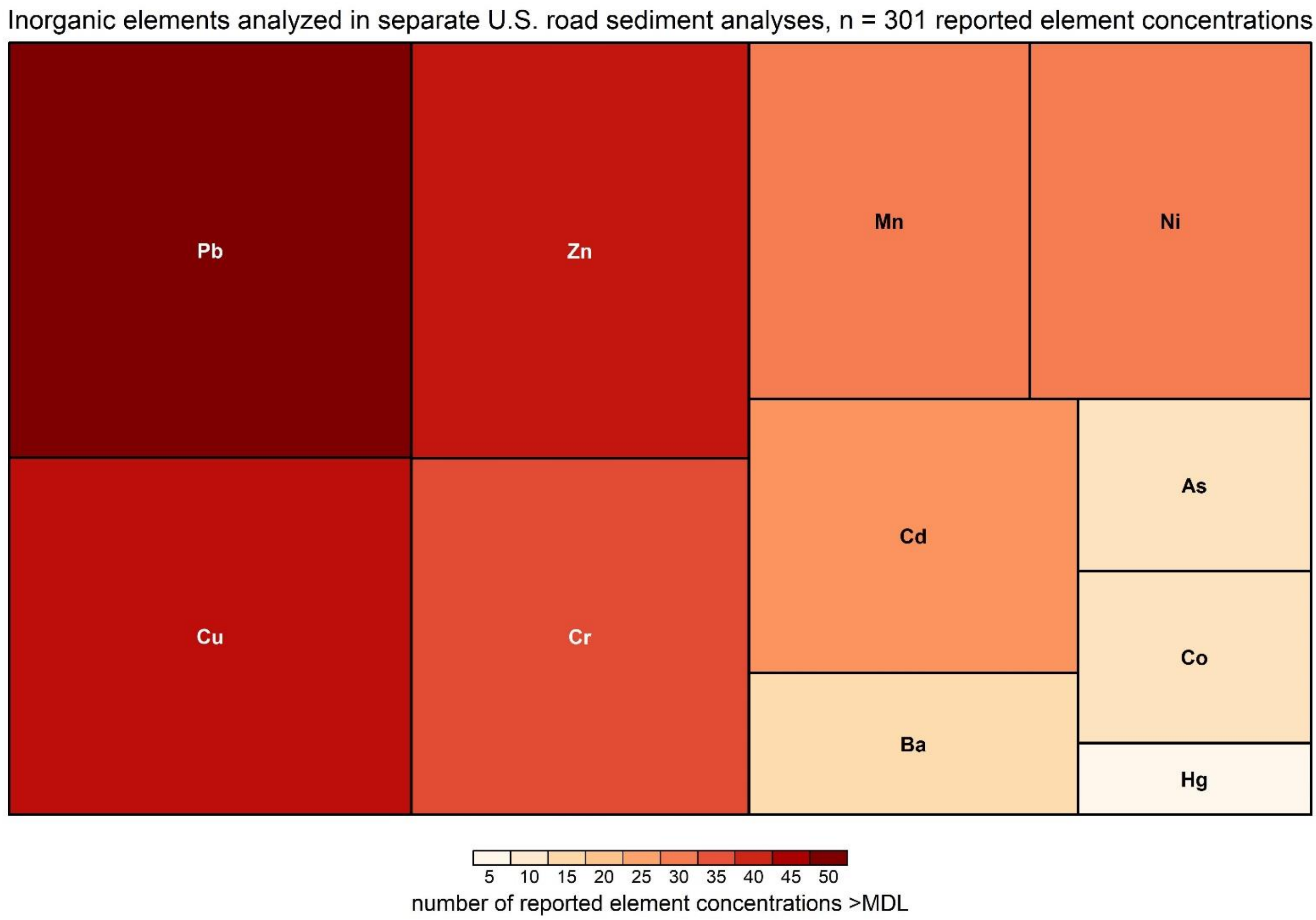

Figure 4: Treemap depicting the proportions of elements > method detection limit (MDL) from each separate analysis within Table 3, shown through the size of each rectangle. Total sample size (n) refers to the total number of elements analyzed and reported within the entire Table 3.

Table 1: Examples of nomenclature utilized in previous studies to describe road sediment.

Term	Example References
Street surface contaminant	Pitt and Amy, 1973
Street dirt	Farmer and Lyon, 1977
Street dust/urban street dust	Bartholomew et al., 2020; Charlesworth et al., 2003; Day et al., 1975; Dean et al., 2017;Duggan and Williams, 1977; Fergusson and Ryan, 1984; Harrison, 1976; Harrison, 1979; Li et al. , 2001; Lu et al., 2014; Solomon and Hartford, 1976; Tang et al., 2013; Tanner et al., 2008; Teran et al., 2020; Zglobicki et al., 2019; Zheng et al., 2010
Urban roadway dust/urban road dust/ road dust	Amato et al., 2009; Bourliva et al., 2017; Deocampo et al., 2012; Jayarathne et al., 2019; Kalenuik and Deocampo, 2011; Hopke et al., 1980; Liu et al., 2007; O'Shea et al., 2020; Shi et al., 2011; Wei and Yang, 2010; Zannoni et al., 2016; Zhao et al., 2016
Urban sediment/street sediment/road sediment	Dietrich et al., 2018; Dietrich et al., 2019; Flett et al., 2016; Irvine et al., 2009; LeGalley et al., 2013; LeGalley and Krekeler, 2013; Selbig et al., 2013; Zibret and Rokavec, 2010
Road-deposited sediment	Andrews and Sutherland, 2004; Sutherland et al., 2000; Sutherland and Tolosa, 2000; Sutherland, 2003
Street particles	Lau and Stenstrom, 2005

Table 2: A summary of the major processes and related factors or subprocesses that control the nature and distribution of road sediment.

Processes controlling road sediment distribution			
Inputs	Redistribution/ Modification	Retention	Outputs
Atmospheric Deposition Aerodynamic sorting Photoreactions	**Air/ Wind Resuspension** Aerodynamic sorting Photoreactions Moisture	**Vegetation** Abundance Height Leaf density Geometry	**Street Sweeping** Frequency Efficiency
Vehicular Spall and Deposition Corrosion Abrasion Exhaust	**Vehicular Transport and Redeposition** Adhesion to tires and surfaces Vehicle size Vehicle speed Ambient precipitation	**Road Surface Modification** Coatings (tar and chip) Road treatment mineralization	**Vehicular Entrainment** Adhesion to tires and surfaces Traffic density Vehicle size Vehicle speed Weather conditions
Building Spall and Release Material type Material ages Roof and drainage design	**Construction/ Modification** Abrasion Mechanical produced debris	**Road Abandonment or Closure** Selective vehicle type Total or occasional	**Atmospheric Winnowing/Transport** Wind speed Humidity Geometry of buildings
Road Materials (as a source) Aggregate composition Metals (e.g., V) in asphalt Paint pigments	**Road Treatments** NaCl / $CaCl_2$ Sand Coal wastes Liquid wastes (historic)	**Surface and Infrastructure Features** Permeable pavement Sewers Curbs Swales	**Surface Runoff** Amount Frequency Rate
Surface Flow Overland flow Direct precipitation	**Surface Flow** Amount Frequency Rate	**Potholes** Dimensions Spatial distribution Growth rate	**Stormwater Sewer** Geometry Degree of maintenance Flow capacity
Soil Deposition Slumping Erosion Winnowing	**Street Sweeping** Abrasion Mechanical sorting Aerodynamic sorting	**Pavement Fractures** Dimensions Spatial distribution Growth rate	**Construction/ Modification** Removal Cutting or drilling

Table 3: Summary table of road sediment studies within the United States that contain reportable concentrations of heavy metal(loid)s. Studies included are only those that sampled stationary road sediment, including street sweeping samples (no water runoff samples included). Concentrations are in mg/kg (ppm) and are arithmetic means unless specified.

(Attached Excel File)

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