Critical Review

Use of Riparian Spiders as Sentinels of Persistent and Bioavailable Chemical Contaminants in Aquatic Ecosystems: A Review

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Abstract: Aquatic ecosystems around the world are contaminated with a wide range of anthropogenic chemicals, including metals and organic pollutants, that originate from point and nonpoint sources. Many of these chemical contaminants have complex environmental cycles, are persistent and bioavailable, can be incorporated into aquatic food webs, and pose a threat to the health of wildlife and humans. Identifying appropriate sentinels that reflect bioavailability is critical to assessing and managing aquatic ecosystems impacted by contaminants. The objective of the present study is to review research on riparian spiders as sentinels of persistent and bioavailable chemical contaminants in aquatic ecosystems. Our review of the literature on riparian spiders as sentinels suggests that significant progress has been made during the last two decades of research. We identified 55 published studies conducted around the world in which riparian spiders (primarily of the families Tetragnathidae, Araneidae, Lycosidae, and Pisauridae) were used as sentinels of chemical contamination of lotic, lentic, and estuarine systems. For several contaminants, such as polychlorinated biphenyls (PCBs), Hg, and Se, it is now clear that riparian spiders are appropriate sentinels. However, many contaminants and factors that could impact chemical concentrations in riparian spiders have not been well characterized. Further study of riparian spiders and their potential role as sentinels is critical because it would allow for development of national-scale programs that utilize riparian spiders as sentinels to monitor chemical contaminants in aquatic ecosystems. A riparian spider sentinel program in the United States would be complementary to existing national sentinel programs, including those for fish and immature dragonflies. Environ Toxicol Chem 2022;41:499–514. © 2021 SETAC

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INTRODUCTION

Aquatic ecosystems around the world are contaminated with a wide range of anthropogenic chemicals, including metals and organic pollutants, that originate from point and nonpoint sources (Chen & Driscoll, 2018; Kolpin et al., 2002; Nisbet & Sarofim, 1972; Swackhamer et al., 2004; US Environmental Protection Agency Urban Environmental Program, 2002; Zhou

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et al., 2020). Many of these chemical contaminants have complex environmental cycles, are persistent and bioavailable, and can be incorporated into aquatic food webs. Some contaminants such as Hg and polychlorinated biphenyls (PCBs) biomagnify, thereby reaching elevated concentrations in predators feeding at high trophic positions (Kelly et al., 2007; Lavoie et al., 2013; Walters et al., 2016). Aquatic ecosystems with food webs contaminated by high concentrations of chemicals can pose a threat to the health of wildlife and humans (Ankley et al., 2021; Berninger & Tillitt, 2019; Mergler et al., 2007; Scheuhammer et al., 2007; Sunderland et al., 2019).

The hazard posed by chemical contaminants to wildlife and human health is determined in part by their bioavailability;

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however, determining the bioavailability of contaminants from measurements of concentrations in water or sediment can be difficult (Phillips & Segar, 1986 but see Lydy et al., 2014). Monitoring contaminants in water and sediment can be challenging from an analytical perspective because contaminants occur at low concentrations, are variable over space and time, and have the potential for matrix interference during analyses. In addition, the concentration of a contaminant in water or sediment may not be reflective of its bioavailability. The degree to which a contaminant is bioavailable and eventually bioaccumulates in biota is a function of the qualities of the contaminant, the organism, and the environmental conditions under which the organism and contaminant interact (Newman & Unger, 2003). Determining chemical concentrations in the tissues of sentinel species (species that accumulate contaminants in their tissues without significant adverse effects) overcomes some of the challenges associated with determining the bioavailability of chemicals in the environment. Use of sentinel species to monitor contaminants can be more straightforward from an analytical perspective (i.e., more concentrated chemicals integrated across space and time, and lower possibility for matrix interference) and provides a direct measure of contaminant

bioavailability (Beeby, 2001; Phillips & Segar, 1986). Therefore, identifying appropriate sentinels that reflect bioavailability is critical to assessing and managing aquatic ecosystems impacted by contaminants.

Although terrestrial spiders have been used for decades as sentinels to assess the bioavailability of chemicals that directly contaminate consumers in terrestrial environments (Larsen et al., 1994; Migula et al., 2013), terrestrial spiders feeding in riparian habitats (hereafter referred to as riparian spiders) also have the potential to help explore how chemicals in aquatic environments cross ecosystem boundaries and contaminate terrestrial consumers (Kraus et al., 2020). Riparian spiders depend on nutrient and energy subsidies from aquatic environments (Baxter et al., 2005); however, there can be a "dark side" to these subsidies when they are associated with persistent and bioavailable chemical contaminants (Sullivan & Rodewald, 2012; Vander Zanden & Sanzone, 2004; Walters et al., 2008). Aquatic contaminants are transported from aquatic systems by adult emergent insects (e.g., dipterans) that are consumed by terrestrial predators such as riparian spiders (Figure 1). Pioneering studies in the early 2000s (e.g., Du Laing et al., 2002; Walters et al., 2008) pointed out that riparian spiders have many traits

FIGURE 1: A generalized diagram depicting the movement of biomagnifying contaminants from aquatic to terrestrial ecosystems. Key ecological and anthropogenic processes appear in italics. Thickness of arrows represents increasing contaminant concentrations. Insect emergence from aquatic to terrestrial ecosystems is depicted by a dashed line connecting larval and adult stages. Representatives of riparian spiders with two different hunting strategies (web builders and ground dwellers) are shown. Web‐building spiders typically derive a higher proportion of the diet from aquatic food webs than ground-dwelling spiders (Supporting Information, Table S1). The relative amount of a given contaminant originating from aquatic versus terrestrial sources in a spider's diet will vary by the unique cycle of the contaminant and the ecology of the system (e.g., the amount of insect emergence). In general, contaminants that have high bioavailability in aquatic habitats, are highly bioaccumulative, and exhibit biomagnification, will be readily transferred from aquatic ecosystems to terrestrial predators (as illustrated in the image). Contaminants that are not bioavailable in aquatic systems or that are lost during metamorphosis and emergence are not transferred in high concentrations to terrestrial predators.

TEXTBOX 1

Spiders (order Araneae) are a diverse group of organisms whose members have unique natural histories that should be considered when utilizing riparian spiders as sentinels of aquatic contamination. In this textbox we highlight important characteristics of four spider families commonly utilized in contaminant studies ([A] Tetragnathidae, [B] Araneidae, [C] Lycosidae, and [D] Pisauridae). Supporting Information, Table S1, contains additional details about these four families, along with references to supporting literature. Photo credits: (A) R. Otter. (B) Paul Reeves Photography, Shutterstock. (C) Regina K. Dale. (D) Michael Benard, Shutterstock.

that make them effective sentinels of aquatic to terrestrial contaminant flux. For example, spiders in the families Araneidae, Lycosidae, Pisauridae, and Tetragnathidae that live in riparian habitats: (1) are found on all continents except Antarctica, (2) are relatively sedentary, (3) can specialize in the consumption of aquatic insects, (4) typically occur in high densities (which allows for sample replication), and (5) are "labeled" by persistent, bioavailable chemicals exported from aquatic systems (Du Laing et al., 2002; Walters et al., 2008; Textbox 1 and Supporting Information, Table S1).

The objective of the present study is to review research on riparian spiders as sentinels of persistent and bioavailable chemical contaminants in aquatic ecosystems. Based on a literature search, we first overview studies using riparian spiders as sentinels of aquatic contamination. We then evaluate each study's contribution to the understanding of factors that facilitate the interpretation of sentinel data. Specifically, we overview progress made in understanding factors related to:

(1) the suitability of riparian spiders as sentinels, (2) the biological determinants of assimilation and excretion rates in riparian spiders, (3) community, ecosystem, or landscape ecology factors influencing contaminant accumulation, and (4) the application of riparian spider sentinels in aquatic contaminant monitoring. Our results highlight the potential use of riparian spiders as sentinels in national‐scale monitoring programs focused on assessing chemical contaminants in aquatic systems.

MATERIALS AND METHODS

We focused our review on publications that use riparian spiders as sentinel accumulator species (hereafter referred to as sentinel species), defined by Beeby (2001) as "species that serve to map the bioavailable fraction in an ecosystem by retaining the pollutants in their tissues." Using this definition, any publication that examined contaminants in riparian spiders was included in the review. We did not include studies that used

riparian spiders solely as monitor species, defined as "measuring the impact by an impairment of their function/performance" or indicator species defined as "indicating the scale of pollution by their absence or presence" (Beeby, 2001). We limited the scope of our review to field studies that used riparian spiders as sentinel species to assess the transfer of contaminants from aquatic to terrestrial ecosystems. We did not include studies that used spiders as sentinels of contaminant cycling solely within terrestrial environments (e.g., Maelfait & Hendrickx, 1998; Marc et al., 1999; Migula et al., 2013; Yang et al., 2016) or laboratory studies of the ecotoxicology of spiders (e.g., Hendrickx et al., 2003; Jung et al., 2005; Migula et al., 2013; Wilczek, 2017; Wilczek et al., 2008).

Next, we used a multi‐tiered approach to identify published studies written in English for inclusion in this review. We first searched our personal libraries for relevant studies. We then performed a Web of Knowledge (Thompson Reuters) database search in December 2020 for articles published from 1864 to the present using the keyword combinations: (contamina* and spider*) and (contamina* and riparian food web*). The "*" insured that any variant of the stem (e.g., contaminant, contaminated, etc.) was included in the results. If we determined that a publication met our criteria for inclusion in this review, we then examined the papers cited within the reference section of the publication as well as all the papers that have cited the publication (papers citing the relevant publication were identified using Google Scholar). We continued searching in this manner (examining papers in reference sections of each relevant study as well as papers citing relevant studies) until we no longer identified publications that met our criteria. The papers that met our criteria for inclusion in this review are overviewed in Supporting Information, Table S2. We acknowledge that focusing on studies written in English may have biased our results toward countries and regions where scientists typically publish in English.

We determined how each study contributed to the field's understanding of factors that facilitate the interpretation of sentinel data (Tables 1 and 2; factors adapted from Beeby, 2001). Beeby (2001) developed a list of characteristics associated with ideal sentinels (hereafter referred to as suitable sentinels) as well as the biological determinants of contaminant assimilation and excretion by sentinels (see tab. 1 and 2 in Beeby, 2001). Some of the characteristics of suitable sentinels are associated with spider natural history; however, these characteristics are not frequently assessed directly by the studies we focused on for this review. Because an understanding of spider natural history is critical to incorporating them into ecotoxicology research, we discuss natural history characteristics of riparian spiders in Supporting Information, Table S1, along with an overview of selected natural history literature and commentary on how these characteristics are relevant to contaminant monitoring. After reviewing the literature on riparian spiders as sentinels of aquatic contaminants, we determined that several studies addressed ecological factors that influence contaminants in spiders. Ecological factors—especially at the community,

ecosystem, or landscape level that could influence contaminant accumulation in sentinels—have not been well documented in earlier reviews of sentinels (Beeby, 2001; Phillips & Segar, 1986). Therefore, we added several ecological factors to Table 2 that may influence contaminants in riparian spiders. Finally, we categorized studies according to the primary application(s) of riparian spider sentinels (Table 3). We determined that published studies used riparian spider sentinels of aquatic contamination in at least four ways, including: (1) monitoring for aquatic contaminants in terrestrial ecosystems, (2) monitoring riparian spider contamination levels in response to disturbance, (3) conducting ecological risk assessments, and (4) assessing levels of contamination in previously unmonitored aquatic ecosystems. The definitions/ criteria that we used to determine if a published study should be included in Tables 1 through 3 are provided in Supporting Information, Table S3.

RESULTS AND DISCUSSION

Overview of studies using riparian spiders as sentinels of aquatic contamination

Our keywords yielded 782 publications in a Web of Knowledge database search. From these and our personal libraries, we identified 55 publications in which riparian spiders were sentinels of aquatic contamination (Figure 2 and Supporting Information, Table S2). The earliest study that met our criteria was published in 1973. Since the late 1990s, the cumulative number of papers published on riparian spiders as sentinels has increased exponentially (Figure 2). From 1998 to 2015, an average of 1.4 papers were published per year; however, during the past 5 years (2016–2020) an average of 4.6 papers have been published per year (Figure 2). Studies of riparian spiders as sentinels have been conducted in 12 countries with most (65%) in the United States (Figure 3A). Research has been conducted primarily in lotic systems (53% of studies), followed by lentic systems (36% of studies), and estuaries (10% of studies; all discussed in Supporting Information, Table S2). Studies have been nearly evenly distributed between point and nonpoint sources of contamination (58% and 42% of studies, respectively), with most studies examining Hg, a suite of trace metals, PCBs, and Se (Figure 3B and Supporting Information, Table S2). Finally, studies have involved 13 spider families, with the majority of work focusing on the families Tetragnathidae, Araneidae, Lycosidae, and Pisauridae (Figure 3C and Supporting Information, Table S2).

Suitability of riparian spiders as sentinels of aquatic contamination

As the field has developed, many studies have assessed whether riparian spiders have characteristics that make them suitable sentinels of aquatic contamination (Table 1). These studies focused on determining if contaminants are retained in the tissue of the spider, defining the route of exposure to the contaminant (e.g., from diet, sediment/soil, or water),

TABLE 1: Studies (identified by number) that have assessed the suitability of riparian spiders as sentinels of aquatic contamination TABLE 1: Studies (identified by number) that have assessed the suitability of riparian spiders as sentinels of aquatic contamination

Koch et al., 2020; 50, Naslund et al., 2020; 51, ćCetinić et al. 2021; 52, Hannappel et al., 2021; 53, Kraus et al., 2021; 54, Ku et al., 2021; 55, ćPrevišić et al., 2021.

Linyphiidae, Salticidae. Contaminant abbreviations: Hg = mercury; PCBs = polychlorinated biphenyls; Se = selenium.

Spider family name abbreviations: Ara = Araneidae; Lyc $=$ Lycosidae; Pis = Pisauridae; Tet = Tetragnathidae; Other = Gnaphosidae, Phoicidae, Theridiidae, Agelenidae, Oxyopidae, Clubionidae, Clubionidae,

determining if there is correspondence between concentrations in spider tissues and ambient environmental concentrations, determining if this correspondence is consistent across space and time, and determining if spiders are insensitive to the contaminant (Table 1). Below, we review what has been discovered about each of these characteristics.

Retention of contaminant in tissues

One of the first steps in determining the suitability of riparian spiders as sentinels of aquatic contamination is to determine if contaminants are retained in the tissues of spiders. The retention or bioaccumulation of Hg, PCBs, and Se in the tissues of riparian spiders has been demonstrated repeatedly (Table 1). The algal toxin microcystin (Moy et al., 2016), organic contaminants including per‐ and polyfluoroalkyl substances (PFAS), along with several pharmaceuticals and endocrine‐disrupting compounds (Koch et al., 2020; Previšić et al., 2021; Richmond et al., 2018), and some metals including Cd, Cu, Cr, Zn, and Pb have been less well studied but have been found to be retained in the tissues of riparian spiders (Beaubien et al., 2020; c´Cetinić et al., 2021; Du Laing et al., 2002; Hendrickx et al., 2004; Kim & Kim, 2016; Kraus et al., 2021; Maelfait & Hendrickx, 1998; Otter et al., 2013; Ramirez et al., 2011; Schipper et al., 2008; Tojal et al., 2002; Torres & Johnson, 2001; Zhang et al., 2009). This suggests that riparian spiders may be suitable sentinels for these contaminants as well (but see c ´Cetinić et al., 2021; Kim & Kim, 2016; Kraus et al., 2021; Torres & Johnson, 2001). In contrast, other metals, including Ag, Al, As, Ba, Co, Cs, Fe, Mn, Mo, Ni, Sb, Sr, Ti, V, and tributyltin c'(Cetinić et al., 2021; Hepp et al., 2017; Kraus

et al., 2021; Laws et al., 2016; Otter et al., 2013; Torres & Johnson, 2001) and radionuclides of Cs and Ur (Anderson et al., 1973; Bergmann & Graça, 2020) are not retained in the tissues of riparian spiders or are found at concentrations lower than concentrations in their prey items, which may limit the utility of riparian spiders as sentinels of these chemicals.

Defined route of exposure

Several studies have attempted to define the route of exposure to riparian spiders by measuring contaminant concentrations in media (e.g., diet, sediment/soil, and water) and conceptualizing exposure pathways. Riparian spiders are primarily exposed to Hg, PCBs, and Se through the consumption of contaminated emergent aquatic insects, as indicated by studies using stable isotopes of N, C, and Hg as ecological tracers of diet (Abeysinghe et al., 2017; Alberts & Sullivan, 2016; Bartrons et al., 2015; Kwon et al., 2015; Ortega‐Rodriguez et al., 2019; Raikow et al., 2011; Rodenhouse et al., 2019; Speir et al., 2014; Tsui et al., 2012; Walters et al., 2008, 2010), correlations between spider contaminant levels and those of insect prey (Naslund et al., 2020; Pennuto & Smith, 2015; Tweedy et al., 2013; Wyman et al., 2011), and analysis of prey items in webs (Chaves‐Ulloa et al., 2016). Studies of microcystin, pharmaceuticals, PFAS, and some metals have also used stable isotopes and correlations of riparian spider–prey contaminant concentrations to confirm insect prey as the primary source of these contaminants (Kim & Kim, 2016; Koch et al., 2020; Moy et al., 2016; Richmond et al., 2018) but these toxins and contaminants have been less well studied. We note that a recent laboratory study (Aziz et al., 2020) suggests that soils can be an important source of

	Hg				PCBs			Se			Other contaminants		
	Tet	Ara	Lyc	Other	Tet	Ara	Pis	Tet	Ara	Other	Tet	Ara	Other
Biological determinants													
1. Diet	17, 19, 22, 34 33, 34, 43		23, 43	12, 23, 25, 33, 43	9, 11	9, 11, 13	- 9				31		47, 49
2. Age/size	52	27, 52	52	52								14	
3. Season/year			26								31	14	
4. Sex	42												
5. Taxonomic differences	33, 34, 43, 46, 52	52	34, 46, 23, 37, 43, 52	23, 33, 37, 43, 52	11, 36, 11, 36, 41	41							5,6
Ecological determinants													
1. Distance from shore		39	23, 39	23, 25, 39, 45		13							
2. Physicochemical and hydrological factors	29, 33			33									4
3. Habitat and land cover/ land use	24, 28, 48							28, 48					
4. Interactions with other species	19												

TABLE 2: Studies (identified by number) assessing biological and ecological determinants of contaminant accumulation in riparian spider sentinels

References and study numbers, along with definitions of abbreviations, are given in Table 1 footnote. Factors in column 1 are defined and reviewed in the text and in Supporting Information, Table S3.

FIGURE 2: The total (histogram) and cumulative (line) number of publications using riparian spiders as sentinels of aquatic contamination. The cumulative number of publications has increased exponentially over the time period $1998-2020$ (y=1.69e $^{0.15\times}$; $R^2 = 0.98$).

contamination for some metals and this source should be explored for riparian spiders.

Correspondence between contaminant concentrations in riparian spiders and ambient levels in the environment

Correspondence between contaminant concentrations in riparian spiders and ambient levels in the environment or tissues of other sentinels (e.g., fish) has been qualitatively established for Hg, Se, PCBs, and pharmaceuticals (Abeysinghe et al., 2017; Gann et al., 2015; Gerson et al., 2020; Otter et al., 2013; Raikow et al., 2011; Richmond et al., 2018; Walters et al., 2018). For example, Gann et al. (2015) found that Hg concentrations in riparian spiders were elevated in lake habitats where fish tissue concentrations were also elevated, and Richmond et al. (2018) found that pharmaceutical concentrations in riparian spiders were highest at sites dominated by treated wastewater effluent (i.e., the primary source of pharmaceutical contamination). Some studies have established a correspondence between riparian spider tissues and ambient levels in aquatic ecosystems in a more quantitative way for Hg, Se, PCBs, and other metals (Cd, Cu, Pb, and Zn) (Kraus et al., 2017, 2021; Naslund et al., 2020; Schipper et al., 2008; Tweedy et al., 2013; Walters et al., 2010). For example, Walters et al. (2010) found a positive correlation between PCB concentrations in riparian spiders and sediment PCB concentrations, whereas Naslund et al. (2020) correlated Se concentrations in riparian spiders and Se concentrations in stream biofilm.

The correspondence between riparian spider tissues and ambient concentrations in the environment is not always straightforward. Some studies have reported correlations

FIGURE 3: Publications utilizing riparian spiders as sentinels of aquatic contamination as a function of (A) location, (B) contaminant, and (C) spider family. In panel (B) EDCs = endocrine disrupting compounds, PCBs = Polychlorinated biphenyls, Pharm = pharmaceuticals, and PFAS = Per- and polyfluoroalkyl substances. In panel (C) : Ara = Araneidae, Lyc = Lycosidae, Pis = Pisauridae, Tet = Tetragnathidae, and Unidentified = studies that did not identify spider families or examined multi-family composite samples. Total number of publications in (B) and (C) was >55 (i.e., greater than the number of publications reviewed in the present study) because some publications examined multiple contaminants and spider taxa. Map credits: ©2021 Mapbox; ©Open-StreetMap.

that are modulated by another factor; others were unable to establish correspondence (Alberts & Sullivan, 2016; Chaves‐ Ulloa et al., 2016; Du Laing et al., 2002; Hendrickx et al., 2004; Kim & Kim, 2016; Kraus et al., 2021; Maelfait & Hendrickx, 1998; Otter et al., 2013; Tojal et al., 2002; Zhang et al., 2010). For example, Chaves‐Ulloa et al. (2016) concluded that a relationship between Hg concentration in water and Hg concentrations in riparian spiders appears to exist but that it is modulated by dissolved organic carbon (DOC), whereas six different studies reported a lack of correlation between the concentrations of Cr, Cu, Cd, Zn, or Pb in soil/ sediment and the concentration in riparian spiders (Du Laing et al., 2002; Hendrickx et al., 2004; Kim & Kim, 2016; Maelfait & Hendrickx, 1998; Tojal et al., 2002), perhaps reflecting a disconnect between concentrations in soil/sediment and bioavailability for these metals (Maelfait & Hendrickx, 1998). Finally, Kraus et al. (2021) demonstrated limited correspondence between ambient trace metals in streams and concentrations in riparian spiders due to less efficient trophic transfer and high metamorphic loss of these metals by insect prey. Although it is clear that riparian spiders can reflect aquatic contamination for some chemicals—namely, persistent organic contaminants and some metals such as Hg more studies are needed to understand: (1) the quantitative relationships between riparian spider contaminant concentrations and concentrations in sediment, prey items, and other sentinels such as fish, especially for trace metals where correspondence between contamination and riparian spider tissue concentrations has not been consistently identified, and (2) the factors that may modulate correspondence between contaminant concentrations in riparian spiders and these media (i.e., sediment, prey items, and other sentinels). In addition, the concentration of contaminants in water and sediments may be more variable than in spiders, which integrate contaminant signals. The potential for encountering patchy distribution of contaminants in sediment and water should be taken into consideration when designing studies to assess correspondence between contaminant concentrations in riparian spiders and ambient levels in the environment.

Correspondence between contaminant concentrations in riparian spiders and ambient levels in the environment are consistent across space and time

Although a few studies have identified correlations between contaminant concentrations in riparian spider tissue and environmental media or even concentrations in other sentinels (e.g., Gann et al., 2015; Otter et al., 2013; Richmond et al., 2018; Walters et al., 2018), only one study (Kraus et al., 2021) assessed whether these relationships are consistent across space and time. Kraus et al. (2021) analyzed metal concentrations in water and riparian spider tissues from 18 streams. Despite observing a 2 to 3 order of magnitude gradient in the concentrations of 17 trace metals (e.g., Cd, Cu, Mn, Mo, Pb, Sb, Se, and Zn) in the water, for most metals, metal concentrations in spiders were low and largely invariant (Kraus et al., 2021). Kraus et al. (2021) concluded that decoupling between metal concentrations in stream water and riparian spiders can be attributed to losses of metals that occur during the metamorphosis of emergent aquatic insects (the primary prey of riparian spiders). As a result, Kraus et al. (2021) concluded that riparian spiders are unlikely to be useful sentinels for most trace metals examined in their study (but noted that Pb, Ba, Cd, Cu, and Mn may be exceptions to this generalization). Future work focusing on the relationship between contaminants in riparian spiders and aquatic environmental media could help determine whether predictable, consistent relationships exist between them.

Insensitive to the pollutant over the range of ambient concentrations

Most studies make the implicit assumption that there is no effect of the focal contaminant on riparian spiders; however, we only found three examples of studies that tested this assumption in a field setting. Wyman et al. (2011) found no effect of Hg at ambient concentrations at their field site on web building of araneid spiders. Maelfait and Hendrickx (1998) and Ramirez et al. (2011) reported a number of physiological and morphological changes related to Cd and Cu concentrations in riparian spider tissues. For example, Maelfait and Hendrickx (1998) found that riparian spiders with elevated concentrations of Cd in their tissues exhibited greater fluctuating asymmetry (i.e., differences between the two sides of bilateral organisms) and that sexually mature females were smaller and produced fewer but larger eggs than those at a reference site. We note that laboratory and terrestrial field studies that fell outside the scope of this review have assessed the toxicology of a number of contaminants on spiders and other arthropods (e.g., Hendrickx et al., 2003; Jung et al., 2005; Migula et al., 2013; Wilczek, 2017; Wilczek et al., 2008) but in general this topic has not been well studied. More studies are needed that address this issue, especially for those riparian spider taxa frequently utilized in sentinel studies.

Future studies on the suitability of riparian spiders as sentinels of aquatic contamination

In addition to the factors discussed above, determining the rate of equilibrium between the contaminant source and contaminant concentrations in riparian spiders is critical to determining their suitability as sentinels (Beeby, 2001). Although the rate of equilibrium of metal concentrations in tissues in one taxon of spider (Lycosidae, Pardosa sp.) has been determined in a laboratory experiment (Jung et al., 2005), the rate of equilibrium with the contaminant source has not been explicitly studied in riparian spiders in the field, and how quickly spiders respond to changes in contaminant concentrations is unknown. In the laboratory, the rate of equilibrium differed by metal but occurred on the scale of weeks for all metals studied (Jung et al., 2005). The rate of change may also be linked to prey consumption and growth rates as found for fish (e.g., Sandheinrich & Drevnick, 2016). Determining the rate of equilibrium for other contaminants would be required to determine how riparian spiders could be used as sentinels in situations when contamination levels may change, such as after a spill or contaminant remediation action.

Biological determinants of contaminant accumulation in riparian spiders

An organism's biology influences assimilation and excretion rates and ultimately contaminant concentrations (Beeby, 2001). A few biological determinants of contaminant accumulation in riparian spiders—including diet, age/size, seasonal/annual variation, sex, and taxonomic differences—have been studied. Below we review what has been discovered about each of these biological determinants of assimilation and excretion rates in riparian spiders (Table 2).

Diet. Diet is one of the most studied biological determinants of contaminant concentration in riparian spiders. In general, emergent aquatic insects are more contaminated with some chemicals, such as Hg and PCBs, than terrestrial insects (e.g., Ortega‐Rodriguez et al., 2019; Speir et al., 2014; Walters et al., 2010) and studies have used stable isotopes, analysis of insects in webs, or correlations between spider and prey to demonstrate that contaminant concentrations in riparian spiders are correlated with the proportion of emerging aquatic insects in their diet. This relationship between contaminant concentrations in riparian spiders and the proportion of emerging aquatic insects in their diet has been well established for Hg and PCBs (Ortega‐Rodriguez et al., 2019; Raikow et al., 2011; Speir et al., 2014; Sullivan et al., 2016; Tsui et al., 2012; Tweedy et al., 2013; Walters et al., 2010). Although there have been fewer studies, the same trend has been observed for Se, pharmaceuticals, and PFAS (Alberts & Sullivan, 2016; Koch et al., 2020). It should be noted that two studies have shown that terrestrial insects can expose riparian spiders to Hg (Bartrons et al., 2015; Kwon et al., 2015); more studies are needed to elucidate if terrestrial insects serve as important sources of Hg and other contaminants.

Age/size. The effect of riparian spider age/size has only been examined for metals. Negative correlations between Hg and size (Pennuto & Smith, 2015) and Cd and size/age class (Ramirez et al., 2011) have been reported, whereas positive correlations were reported between Hg and size in four riparian spider taxa (Hannappel et al., 2021). Hannappel et al. (2021) reported a lack of correlation between Hg and size in two riparian spider taxa, whereas Ramirez et al. (2011) reported a lack of correlation between size and Zn, Cu, and Cr in riparian spider tissues. Although age‐related bioaccumulation occurs in other sentinels (e.g., fish; Wiener et al., 2003), its importance in riparian spiders has not been well established. More studies involving additional taxa and contaminants are needed to understand the effect of riparian spider age/size on contaminant concentrations.

Season/year. Seasonal and annual variation in contaminant concentrations in riparian spiders have rarely been examined but three studies suggest that these factors could be important. Significant interannual differences in Cd, Cr, Cu, and Zn in spider tissue were identified by Ramirez et al. (2011) and hypothesized to be related to rainfall differences between years. Laws et al. (2016) found that the proportional contribution of chironomids to riparian spider biomass varied with month and that organotin concentration in spiders was correlated with the proportional contribution of chironomids to riparian spider biomass. Seasonal variation in Hg concentration that corresponded with fog prevalence, an important source of MeHg in northern California, was observed by Ortiz et al.

(2015). Further study of seasonal and annual variation in contaminant concentrations in riparian spiders is needed.

Sex. The potential effect of sex on contaminant concentrations in riparian spiders has only been examined in one study. Beaubien et al. (2019) found no difference in Hg concentrations between male and female tetragnathid spiders. Additional studies involving other contaminants and riparian spider taxa, especially those with pronounced sexual dimorphism such as some genera in the family Araneidae (Elgar & Fahey, 1996; Ubick et al., 2017), are needed to further explore the role of sex in riparian spider contaminant concentrations. This could be particularly important for designing longer term monitoring studies because spiders often need to be composited into a single sample to achieve mass requirements for analysis of some chemicals (e.g., PCBs).

Taxonomic differences. Twelve studies have statistically assessed taxonomic differences in contaminant concentrations (Abeysinghe et al., 2017; Bartrons et al., 2015; Beaubien et al., 2020; Hannappel et al., 2021; Hendrickx et al., 2004; Kraus et al., 2017; Ortega‐Rodriguez et al., 2019; Sullivan et al., 2016; Tavshunsky et al., 2017; Tojal et al., 2002; Walters et al., 2008, 2010; Walters et al., 2018). Among these, more than 80% (10 out of 12 studies) reported taxonomic differences in contaminant accumulation. These differences in contaminant levels are likely caused by taxon‐specific differences in diet (e.g., reliance on emergent aquatic insects and trophic position) and other biological traits (e.g., age/size and season) known to influence contaminant concentrations (as discussed previously). The most commonly collected taxa belong to the families Tetragnathidae and Araneidae. Two studies have reported greater contaminant concentrations in tetragnathids (Beaubien et al., 2020; Walters et al., 2018), three have reported equivalent concentrations (Abeysinghe et al., 2017; Hannappel et al., 2021; Kraus et al., 2017); and one reported that tetragnathid spiders had lower PCB concentrations than araneids consuming mostly emergent insects, but higher concentrations than a particular species of araneid (Mecynogea lemniscata) which consumes a greater proportion of terrestrial insects (Walters et al., 2010). These studies illustrate the importance of reporting the lowest taxonomic resolution of riparian spiders possible and taking taxonomic differences into account when selecting study species.

Future studies on biological determinants of contaminant accumulation in riparian spiders

To our knowledge, other potential biological determinants of contaminant accumulation in sentinels identified by Beeby (2001) have not been assessed in riparian spiders. These include physiological state (e.g., condition), feeding area exploited, population/genetic differences, and how the duration, level of exposure, or interactions with other contaminants may impact assimilation. Some of these factors, such as condition, are known to influence contaminant accumulation in other

sentinels, such as fish (Sandheinrich & Drevnick, 2016), and should be explored in riparian spiders.

Community, ecosystem, or landscape ecology factors influencing contaminant accumulation

Ecological factors can impact the transport and bioavailability of contaminants as well as the biomass and diversity of spiders and their prey items. As described in the following four paragraphs, a few studies have explored how ecological factors such as distance from shore, physicochemical and hydrological factors, habitat and land cover/land use, and interactions with other species influence contaminant concentrations in spiders (Table 2).

Distance from shore. A few studies have explored the effect of distance from the shore on riparian spider contaminant concentrations, which may be dependent on the specific contaminant and spider taxa being studied. For PCBs, Raikow et al. (2011) found that concentrations exhibited a strong decline beyond 5 m from the shoreline that corresponded to a decrease in emerging aquatic insects in riparian spider diets. For Hg, the effect of distance from the shore on riparian spider tissue concentrations is inconsistent. Rodenhouse et al. (2019) reported that Hg concentrations were significantly higher in "web spiders" collected <1 m from a stream channel than in those collected 75 m away; however, no difference was found for "litter spiders." Bartrons et al. (2015) observed that Hg concentrations in spiders at one lake were elevated at nearshore sampling sites and at sampling sites 200 m away, whereas Hg concentrations in spiders at another lake were elevated at sampling sites 200 m from shore relative to nearshore sampling sites. In another study, Howie et al. (2018) observed that spiders collected hundreds of meters from the contamination source had elevated concentrations of Hg. More studies are needed to understand how contaminant concentrations in the tissues of a given spider taxa change with distance from shore and how this is impacted by the dispersal of aquatic insects inland (Muehlbauer et al., 2014; Vander Zanden & Gratton, 2011), spider hunting strategy (Textbox 1 and Supporting Information, Table S1), spider home range (Supporting Information, Table S1), and terrestrial contaminant cycling (Tsui et al., 2019). Regardless, it is clear from meta‐analysis of stream ecosystems (Muehlbauer et al., 2014) that insect dispersal exhibits a negative exponential decay with distance from streams; thus, most aquatic insect biomass (50%) is concentrated near stream banks (1.3 m). Whereas some nontrivial amount of insect biomass can extend much farther (10s–100s of meters) into terrestrial habitats, these findings indicate that studies using spiders as sentinels of aquatic contamination should focus on nearshore taxa because riparian habitats are where the contaminant signal is most likely to be detected.

Physicochemical and hydrological factors. Some studies have demonstrated the potential influence of physicochemical (DOC and nutrients) and hydrological factors on contaminant

concentrations in riparian spiders. For example, Sullivan et al. (2016) reported that body burdens of Hg in riparian spiders were positively correlated with streamflow. Chaves-Ulloa et al. (2016) determined that the relationship between Hg concentrations in water and Hg concentrations in riparian spider tissues is modulated by DOC to such a degree that Hg concentrations in spider tissues were reduced in the presence of elevated DOC. Du Laing et al. (2002) found that chloride and cation exchange capacity of soils were correlated with Cd, Cu, and Zn in riparian spiders. Although the influence of physicochemical factors and hydrology on riparian spider contaminant levels has only been addressed in a few studies, these are known to strongly influence contaminant concentrations in aquatic taxa such as fish (Evers et al., 2007) and should be explored further for spiders.

Habitat and land cover/land use. A few studies have explored the influence of habitat and land cover/land use on contaminant concentrations in riparian spiders. Alberts and Sullivan (2016) found that riparian spider body burdens of Hg and Se were associated with land cover in the riparian zone and river distance downstream from urban areas. Gann et al. (2015) determined that riparian spiders collected from riverine and wetland habitats were more contaminated with Hg than those from open‐water habitats. Gerson et al. (2020) found that Se, but not Hg, concentrations in riparian spiders were correlated with the percentage of watershed area subjected to mining. Habitat and land cover/land use have been linked to contaminant levels in fish (Driscoll et al., 2007; Drenner et al., 2013; Eagles‐Smith et al., 2016; Evers et al., 2007) and should be explored further for riparian spiders.

Interactions with other species. We are only aware of one study that has explored how interactions with other species influence contaminant concentrations in riparian spiders. Tweedy et al. (2013) found that the presence of fish did not affect Hg concentrations in riparian spiders captured on the shorelines of small ponds because fish did not affect emergence of small, emergent insects—the primary prey of spiders. The influence of other species (including the microbiomes of spiders) on contaminant accumulation in riparian spiders needs further study.

Future studies on community, ecosystem, or landscape ecology factors influencing contaminant accumulation

Other ecological factors that could potentially influence contaminant concentrations in riparian spiders such as ecosystem of collection (e.g., lentic, lotic, or estuary) have never been assessed. Ecosystem type impacts contaminant accumulation within and across food webs (Lavoie et al., 2013; Walters et al., 2016); however, these effects are often best characterized through meta‐analysis of larger datasets that at present do not exist for riparian spiders. Additional research on ecological factors that influence contaminant accumulation will

allow meta‐analyses on a variety of topics, including the influence of ecosystem type.

Application of sentinels in contaminant monitoring

Published studies have used riparian spider sentinels in at least four ways, including to: (1) monitor the movement of aquatic contaminants to terrestrial ecosystems, (2) monitor spider contamination levels in response to a disturbance, (3) conduct ecological risk assessment, and (4) assess the level of contamination in previously unmonitored aquatic ecosystems (Table 3).

Monitoring for aquatic contaminants in terrestrial ecosystems

All studies in our review used riparian spiders to provide insight into the transfer of contaminants from aquatic to terrestrial ecosystems (Table 3). These studies have revealed that not all contaminants accumulate in riparian spider tissues (e.g., As, Hepp et al., 2017), and therefore are not readily transferred to terrestrial ecosystems. However, some contaminants including Hg, Se, and PCBs are retained in riparian spider tissues and, in some cases, riparian spiders have been used to demonstrate the potential exposure of terrestrial predators to these contaminants (Gann et al., 2015). As discussed above, some studies have shown that levels of contaminants in riparian spider tissues are correlated with contaminant levels in the environment (e.g., Abeysinghe et al., 2017; Gann et al., 2015; Gerson et al., 2020; Otter et al., 2013; Raikow et al., 2011; Richmond et al., 2018; Walters et al., 2010, 2018). If the relationships between chemical concentrations in riparian spiders and sediments, water, or other sentinels, such as fish or dragonflies, are consistent among sites, then the concentration of any given contaminant in spiders could be predicted from these other media and biota, and vice versa.

Monitoring contamination levels in riparian spiders in response to a disturbance

Riparian spiders have frequently been utilized to monitor changes in aquatic to terrestrial contaminant flux in response to disturbance (Table 3). Any study that monitored riparian spiders in reference and impacted sites subjected to a disturbance (e.g., contaminant spill or discharge, mining, and remediation) was categorized as such. Most studies in Table 3 have used riparian spiders as sentinels of change in areas exposed to a spill or other point sources of contamination. For example, Ku et al. (2021) monitored Hg in riparian spiders in an area impacted by a coal ash spill at 17 and 29 months after the disturbance. Ku et al. (2021) found that after 2 years, Hg concentrations in riparian spiders were not elevated relative to reference sites, which suggested that the coal ash had been buried beneath the sediment or that Hg in coal ash was not bioavailable. Walters et al. (2018) examined PCBs in araneid and tetragnathid spiders in the Ashtabula River, a Great Lakes Area of Concern, after remediation (dredging and removal of contaminated sediment).

Data collected on PCB concentrations in riparian spiders revealed ongoing contamination in an area that had undergone remediation and also revealed that PCBs found in riparian spiders likely originated from multiple sites within the Area of Concern (Walters et al., 2018).

Although riparian spiders show great potential for monitoring change in response to disturbance, and have been used in this way frequently (Table 3), more studies are needed to allow for better interpretation of results. For example, no field studies have determined the rate of equilibration between riparian spider and source contaminant levels. For most contaminants, it is unknown if the relationship between riparian spider and source contaminant levels is consistent across sites (Table 1). These data are critical to interpreting riparian spider contamination data collected from sites that have experienced a perturbation.

Ecological risk assessment

Riparian spiders are an important prey item for songbirds, especially during the breeding season when energy‐rich spiders are consumed by both adults and nestlings (e.g., Cristol et al., 2008; Walters et al., 2010). A few studies have conducted formal assessments of the risk posed to arachnivorous birds by contaminated riparian spiders. Walters et al. (2010) developed a risk assessment approach for PCB‐contaminated riparian spiders that was later extended by Gann et al. (2015) for Hg. Beaubien et al. (2020) developed an additional risk assessment technique for a large number of birds and heavy metal contaminants. Future studies could be expanded to incorporate arachnivorous bird species living in other regions of the world and other contaminants. Theoretically, the risk assessments that have been developed for songbirds could also be extended to other riparian spider predators such as amphibians. Riparian spiders have a great deal of potential in ecological risk assessment. A chapter by Otter et al. (2020) presents practical considerations for site managers and risk assessors to consider when including riparian spiders in their assessment protocols.

Assessing the level of contamination in previously unmonitored aquatic ecosystems

Finally, because a relationship between aquatic contaminant levels and contaminants in riparian spiders has been established for some contaminants, riparian spiders could be used to identify aquatic systems that were previously not known to be contaminated. To our knowledge, only one study has used riparian spiders as sentinels to identify a previously unmonitored aquatic system as contaminated. Pennuto and Smith (2015) monitored riparian spiders within and upstream of the Buffalo River Area of Concern in New York. They discovered high levels of Hg in riparian spiders within the Area of Concern and also in upstream areas where Hg levels had not been previously monitored in sediments or biota. They suggested that further monitoring and expansion of the Area of Concern boundaries may be needed. Riparian spiders are abundant and easy to sample; therefore, they have great potential for identifying previously unknown areas of aquatic contamination. However, using them to their full potential requires a better understanding of the correlation between contaminants in riparian spiders and contaminants in sediments, water, or other aquatic sentinels, which should also include an understanding of how these relationships vary among sites.

CONCLUSIONS

Our review of the literature on riparian spiders as sentinels suggests that significant progress has been made since pioneering studies during the late 1990s and early 2000s. For several contaminants such as PCBs, Hg, and Se, it is now clear that riparian spiders are appropriate sentinels but that spiders may have less utility as sentinels for some trace metals. Spiders show the most promise as sentinels for contaminants that have higher bioavailability in aquatic habitats relative to terrestrial habitats, are highly bioaccumulative, exhibit biomagnification, and are not lost during metamorphosis. However, many contaminants and factors that could impact chemical concentrations in riparian spiders have not been well studied and continued research is needed to fully understand spider sentinels. In addition, it is clear that spiders can be used to monitor the movement of aquatic contaminants to terrestrial ecosystems, assess aquatic contamination after a disturbance, and be incorporated into ecological risk analyses. Spiders may also have utility in assessing the level of contamination in previously unmonitored aquatic ecosystems; however, only one study has used spider sentinels in this way (Pennuto & Smith, 2015).

The present study also suggests that the field could benefit by incorporating additional spider families into future studies. As shown in Figure 3, tetragnathids have been the focus of most research on riparian spiders as sentinels of aquatic contaminants. Because tetragnathids are widely distributed and commonly found along the shorelines of stream, river, and lentic systems (Ubick et al., 2017), they could form the basis for larger‐scale, regional comparisons of contaminant flux among disparate aquatic–terrestrial ecosystems (Walters et al., 2008). However, tetragnathids also have limitations as sentinel species. Some species of tetragnathids require direct access to water (Gillespie, 1987; Supporting Information, Table S2) and therefore are not abundant in dry areas away from the water's edge. Also, the vegetation that tetragnathids require for construction of webs to capture emergent insect prey (Supporting Information, Table S2) may not be available along all shorelines (e.g., areas where vegetation is overgrazed by livestock or the shoreline is fortified with riprap armor to prevent flooding). The small body size of tetragnathids can be a limitation in situations where large masses of tissue (e.g., grams) are required to detect particular chemicals. In these situations, it is likely that other riparian spiders such as araneids that are found in trees or on human‐made structures (e.g., bridges and boat docks), ground‐dwelling

lycosids, or semi‐aquatic pisaurids may be available as sentinels (Textbox 1 and Supporting Information, Table S1). The wide distribution of these taxa and their abundance in riparian habitats mean that at least one spider family will be available for study around most water bodies (Supporting Information, Table S1). Establishment of correlative relationships between tetragnathids and other riparian shoreline spiders such as araneids, lycosids, and pisaurids (e.g., Kraus et al., 2017) would allow for estimation of contaminant concentrations in tetragnathid‐equivalent spiders by samples of other riparian spider taxa, as is now possible for fish (Wente, 2004) and immature dragonflies (Eagles‐Smith et al., 2020). This would facilitate development of national‐scale monitoring programs (described in the following paragraph).

Further study of riparian spiders is critical for the development of national‐scale programs utilizing riparian spiders as sentinels to monitor certain chemical contaminants that are bioavailable in aquatic ecosystems (e.g., PCBs, Hg, and Se). A national riparian spider sentinel program in the United States would be complementary to other national sentinel programs (e.g., for fish [Wente, 2004] and immature dragonflies [Eagles‐Smith et al., 2020]). Having multi‐dimensional sentinel programs for fish, immature dragonflies, and riparian spiders would provide regulators and scientists with multiple tools to monitor persistent bioavailable chemical contaminants in aquatic systems. Each type of sentinel has both advantages and disadvantages, which provides regulators and scientists with the ability to select among the most appropriate sentinels for the situation. For example, fish have revealed much about contaminants in aquatic systems (Wiener et al., 2003); however, they are labor-intensive to capture, are absent from temporary aquatic systems that periodically dry (Chumchal & Drenner, 2015), and often have migratory behavior and home ranges that could extend far beyond areas of contamination (e.g., Fry & Chumchal, 2011). Immature dragonflies occur at high densities in temporary aquatic systems that lack insectivorous fish (Wellborn et al., 1996); but are not as abundant and can be difficult to collect in systems with fish (Chumchal & Drenner, 2015). Riparian spiders are seasonally abundant and, although they can be readily collected in the warm part of the year where suitable habitat is available, many taxa in temperate climates are often impractical to collect during winter months (Aitchison, 1984; Foelix, 2011). Furthermore, sampling after dark (which is often the most productive sampling time to capture spiders) cannot be done safely at all sites. Eventual integration of fish, dragonfly, and riparian spider sentinel programs would provide a very powerful tool to monitor aquatic contaminants in a wide variety of aquatic systems and assess the risk of contaminants to human and wildlife health.

Supporting Information—The Supporting information are available on the Wiley Online Library at https://[doi.org](https://doi.org/10.1002/etc.5267)/10. 1002/[etc.5267](https://doi.org/10.1002/etc.5267).

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Data Availability Statement-All data are available in Supporting Information, Table S2.

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