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Mitigating Toxic Metal Exposure Through Leafy Greens: A Comprehensive Review Contrasting Cadmium and Lead in Spinach

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Key Points:

- The toxic elements targeted in the Food and Drug Administration Closer to Zero action plan behave differently in soils and in plant uptake
- Mitigation strategies to reduce exposure to toxic elements must consider the drivers of soil mobility and accumulation into edible tissues
- Health and nutrition factors that affect metal and metalloid bioavailability upon ingestion should also be considered

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Abstract

Metals and metalloids (hereafter, metal(loid)s) in plant-based foods are a source of exposure to humans, but not all metal(loid)-food interactions are the same. Differences exist between metal(loid)s in terms of their behavior in soils and in how they are taken up by plants and stored in the edible plant tissue/food. Thus, there cannot be one consistent solution to reducing toxic metal(loid)s exposure to humans from foods. In addition, how metal(loid)s are absorbed, distributed, metabolized, and excreted by the human body differs based on both the metal(loid), other elements and nutrients in the food, and the nutritional status of the human. Initiatives like the United States Food and Drug Administration's Closer to Zero initiative to reduce the exposure of young children to the toxic elements cadmium, lead, arsenic, and mercury from foods warrant careful consideration of each metal(loid) and plant interaction. This review explores such plant-metal(loid) interactions using the example of spinach and the metals cadmium and lead. This review highlights differences in the magnitude of exposure, bioavailability, and the practicality of mitigation strategies while outlining research gaps and future needs. A focus on feasibility and producer needs, informed via stakeholder interviews, emphasizes the need for better analytical testing facilities and grower and consumer education. More research should focus on minimization of chloride inputs for leafy greens to lessen plant-availability of Cd and the role of oxalate in reducing Cd bioavailability from spinach. These findings are applicable to other leafy greens (e.g., kale, lettuce), but not for other plants or metal(loid)s.

Plain Language Summary

Toxic metals like cadmium and lead in foods can be harmful to our health, especially for babies and young children who are more vulnerable due to their smaller size and rapid development. Leafy greens like spinach can absorb these metals from the soil but in different ways. In addition, how and where they accumulate in edible plant tissues also differs. This review uses spinach as an example to compare and contrast how cadmium and lead differ in how they move through soil and accumulate in plant foods. It also discusses practical pre- and post-harvest techniques to lessen human exposure to these metals that can be adopted by producers and consumers. Finally, it highlights future needs and research directions.

1. Introduction

The toxic metals cadmium (Cd), lead (Pb), and mercury (Hg) and the toxic metalloid arsenic (As) (hereafter, toxic metal(loid)s) occur naturally in the environment at levels that do not cause phytotoxicity but can pose human health risks through food consumption. Unlike excess nutrient metals (e.g., Zn) that when controlled to normal levels cause better plant growth and thus a visible response, achieving lower levels of toxic metal(loid)s in edible plants does not cause a change in a measurable plant growth parameter or appearance, thus necessitating confirmation via analytical means. The risk of toxic metal(loid) exposure to human health is based partly on the concentration and speciation of the metal(loid) and other nutrients in the edible portion of the food, the body size of the human, the amount of the food consumed, and the nutritional status of the human (United States Environmental Protection Agency, 1989). It is noteworthy to mention that regulatory standards target only total metal in foods, not their bioavailable amount. It is known that humans who eat varied diets with replete micronutrients and who have larger body sizes are at reduced risk of absorption of a toxic metal(loid) by the intestine (Reeves & Chaney, 2008). Due to their lower body size, limited diets, and rapid developmental growth, babies and young children are a subgroup who are at higher risk of consuming some toxic metal(loid)s in foods (Pokharel & Wu, 2023). A recent study of baby food products showed that products containing rice (*Oryza sativa* L.) tended to have higher levels of As, those containing sweet potato (*Ipomoea batatas* L.) and carrot (*Daucus carota* subsp. *sativus*) tended to have higher levels of Pb, and those containing leafy greens such as kale (*Brassica oleracea* L.) and spinach (*Spinacia oleracea* L.) tended to contain higher levels of Cd (P. J. Gray, 2023). In 2021, the United States Food and Drug Administration (FDA) announced the "Closer to Zero" action plan, which specifically targets foods intended for consumption by babies and young children to have levels of Cd, Pb, As, and Hg as close to zero as possible. To achieve this goal, it is important to consider (a) the root cause of the metal(loid) in the food in question, given that the four metal(loid)s differ greatly in their behavior in the environment and in their uptake into and storage in edible foods, and (b) how effective and practical the mitigation strategies are for the food industry. These considerations are important because interactions between the plant, contaminant, and environment can result in differing levels of each contaminant in different crops.

In this review, we will focus on spinach as an example of a plant-based food and Cd and Pb as the toxic metals to illustrate how different metals behave along the soil-plant-human continuum and how mitigation practices must consider each plant-metal pair individually. We focus on spinach because spinach is a component of foods intended for young children, Cd is known to accumulate to higher levels in leafy greens than in other foods (P. J. Gray, 2023), and recent work has shown that infants in the 6–24 and 24–60 months age range are at a higher risk of Cd exposure through food (Pokharel & Wu, 2023). To help propel achievable mitigation strategies for metal(loid) contamination in spinach, we review the literature on the presence of Cd and Pb in soils, their migration into or onto spinach leaves due to soil and/or plant factors, and exposure mitigation options for both pre- and post-harvest. We start with a brief overview of the human health impacts of these elements, as well as how they are found in natural and human-impacted environments. Throughout, we treat the two elements in parallel, highlighting their differences to emphasize that a one-size-fits-all approach to metal(loid) mitigation is unlikely to succeed due to various chemical and biological characteristics and mechanisms. Finally, in addition to the state of the science regarding mitigation, this review will also consider how each mitigation effort will affect the food yield and the cost-benefit to the food producers, as informed by stakeholder interviews.

1.1. Human Health Impacts of Cd and Pb

The primary routes of human exposure to Cd are via inhalation of contaminated dusts, from smoking, and in the consumption of Cd-rich plant materials. Once inside the body, high cumulative uptake of Cd interferes with processes involving other divalent cations such as calcium and can cause osteoporosis, renal tubular dysfunction, heart disease, and cancer (Faroon et al., 2013). With a long half-life of 18–33 years in the kidney cortex (Kjellström, 1971), Cd exposure can have lasting impacts on the human body long after exposure.

Like Cd, Pb is a heavy metal that is toxic to humans but is particularly threatening to young children. Increased blood Pb levels in young children are associated with impairment of neurological development (Abadin et al., 2007) and disproportionally affect children in urban areas who may be more highly exposed, and who also tend to be from minority groups and/or economically disadvantaged backgrounds (Clark et al., 2006). The primary routes of exposure to children are direct ingestion of soil and house dust, and from elevated Pb on or in some vegetables grown in Pb-rich soils (Attanayake et al., 2014; Brown et al., 2016).

1.2. Cd and Pb in Agricultural Soils

In general, metal(loid)s that naturally occur in the environment or are elevated in concentration due to human activity can be retained by soil through precipitation or sorption with various soil components including organic matter, clays, or hydrous Fe and Mn oxides. Thus, the soil can be considered as a pool of metals to plants, with the contaminant flow to plant roots being controlled by both plant and soil processes. Both Pb and Cd are not phytotoxic at levels typically encountered in soil, which is in contrast to the essential plant nutrients zinc, copper, and nickel, which can cause phytotoxicity when present in excess to what is needed for optimal plant growth (Chaney, 1980, 2015; McLaughlin et al., 1999).

Cd is naturally present in the environment but can be introduced to soils through human activity. There is a wide range of Cd in rocks from 0.001 to 90 mg kg^{-1}, with sedimentary rocks generally having higher Cd concentrations (McLaughlin et al., 1999). This wide range of crustal Cd leads to a wide range of soil Cd levels ranging 2–3 orders of magnitude. In the United States, typical soil Cd concentrations in agricultural soils range from 0.01 to 2.0 mg kg^{-1} (Holmgren et al., 1993), with the higher range either from Cd-rich parent material such as those developed on Monterrey shales in California or topsoils surrounding phosphate ore deposits, or from anthropogenic inputs. Anthropogenic sources include dispersed atmospheric deposition from fossil fuel combustion, production of iron and steel, mining, and smelting. In Japan, Cd-rich waters that drain Zn and Cd-rich mine tailings were used to irrigate rice paddies for decades and were later recognized as the cause of the widespread Cd poisoning and resulting *itai itai* disease in the mid-19th century (Friberg, 1971). Cadmium contamination of soil and rice was not identified as the cause of this disease until the late 1960s (Kobayashi, 1978). Direct inputs of Cd to soil include the use of most phosphate fertilizers, historical inputs of "low quality" Cd-rich biosolids, and Cd-rich manure or compost materials (Chaney, 2012; McLaughlin et al., 1999). Phosphate rock derived fertilizers are reported to contain Cd at levels ranging 2–200 mg kg^{-1} (Lee & Keeney, 1975; Mortvedt & Giordano, 1977; Mulla et al., 1980). Application of sewage sludge has historically elevated soil Cd in some areas (Berrow & Webber, 1972; Furr et al., 1976; McGrath et al., 1988; Sterritt & Lester, 1980), but industry action in the United States to limit Cd discharge in wastewater has resulted in less Cd in biosolids over time (Stehouwer et al., 2000). Results from national surveys of biosolids composition by the US-EPA show a strong reduction in Cd after imposition of both limits of Cd in biosolids and industrial waste discharge to sewers regulations. Before regulations were established, some biosolids contained in excess of 1,000 mg Cd kg^{-1} dry weight (e.g., Chaney et al., 1977). In the latest US-EPA survey of biosolids composition (unpublished), of 4900 samples with valid analyses, 4421 were below 5 mg kg^{-1} and 3283 were below 2 mg kg^{-1}. Some limestone deposits are also enriched in Cd, possibly also contributing to elevated soil Cd in some areas (Rambeau et al., 2010). For example, limestone recovered from Zn mine deposits may contain as high as 20 mg Cd kg^{-1} and 2,000 mg Zn kg^{-1} (Chaney, 2012). Cd-rich soil amendments generally result in Cd enrichment in the tillage depth of agricultural soils (Mulla et al., 1980; Page et al., 1986), whereas soils with naturally present Cd (e.g., soils developed on Monterey shales) may not show decreases with depth to at least 1 m.

Like Cd, Pb can also occur naturally in the environment but is introduced to soils through anthropogenic activity. Concentrations of Pb in U.S. agronomic soils generally range from 10 to 50 mg kg^{-1} with lower levels found in spinach growing regions of California and Arizona and higher levels found in the Pb belt region that encompasses the Missouri and Ohio river valleys, suggestive of industrial activity (Holmgren et al., 1993). The historic use of leaded gasoline has elevated soil Pb levels particularly in areas with high vehicle traffic (Datko-Williams et al., 2014; Schwarz et al., 2013). While the phase-out of leaded gasoline has lessened Pb inputs over time, there may still be health effects from residual dust from these historic inputs and other anthropogenic activities, particularly in urban centers (Mielke et al., 2011; Ye et al., 2022). Leaded paint on structures also increases soil Pb levels in surrounding soils to as high as 10,000 mg kg^{-1}. In studies of vegetables grown in New York City urban gardens, some samples of leafy vegetables were markedly enriched in Pb. However, fine soil particles were the source of the high tissue Pb based on analysis of other elements not accumulated by most plants (McBride et al., 2014). Pb-bearing soils used for urban gardens could be a source of Pb exposure, so extra care should be taken to remove Pb-bearing soil particles from vegetables grown in Pb-rich soils (Attanayake et al., 2014; Clark et al., 2006; McBride et al., 2014).

2. U.S. Spinach Production and Magnitude of Cd and Pb Concentrations

In the United States, 98% of all spinach production is in just four states, with California growing approximately 65% of all acres, and Arizona, New Jersey, and Texas comprising the balance (National Agricultural Statistics Service, 2021). As a cool-season crop, the climate of parts of California (i.e., Salinas Valley) is conducive to growing spinach year-round. The late fall/winter climate in the deserts of southern California, Arizona, and Texas also provides the cool conditions needed for a spinach crop, whereas spinach in New Jersey and surrounding areas is mainly grown in the spring and fall, while summer production (i.e., July) is not possible due to warm temperatures. Spinach production is labor intensive and requires hands-on weed management, clipped spinach handling, and bunched spinach harvests. Spinach is nutritious and provides a good source of antioxidants, folate, vitamins C, K, A, E, and B-6, iron, calcium, and potassium (Morelock & Correll, 2008).

Before discussing the range of concentrations of metals Cd and Pb in spinach, it is critical to understand reported units. In exposure assessments and in regulatory standards, the consumed plant part is expressed as the mass of metal per unit fresh weight (FW) of the food if the food is consumed fresh. Thus, for spinach and other leafy greens, metals are expressed as the mass of the heavy metal per unit FW. However, tissue analysis requires that the fresh plant part be dried to remove water, and after analysis is reported as mass of metal per unit dry weight (DW) of the plant part. Thus, these data need to be converted to fresh weight by accounting for the water lost in the drying process for use in exposure assessments. Some studies do not indicate whether the reported values are

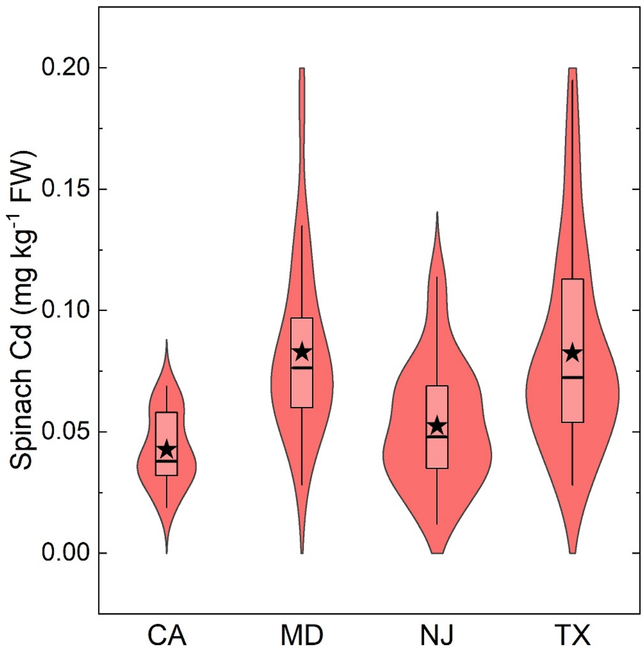

Figure 1. Violin plots of Cd concentrations in spinach leaves found in samples collected from CA (*n* = 19), MD (*n* = 20), NJ (*n* = 35), and TX (*n* = 31) between 1980 and 1982. Data are from Wolnik et al. (1985). The shaded area shows a kernel density plot, stars denote the mean, boxes denote the 25th and 75th percentiles, and the horizontal line denotes the median.

expressed in DW or FW, making comparison between studies challenging. Wolnik et al. (1985) reported that spinach harvested at the farm and processed rapidly contained 7.7% dry matter, or 92% water. Thus, when mg Cd or Pb kg^{-1} dry spinach is reported, that should be multiplied by 0.077 to reach mg Cd or Pb kg^{-1} fresh spinach.

2.1. Cd in U.S. Spinach

While leafy greens generally contain higher Cd levels than other plant foods, the concentrations in spinach are among the highest due to efficient transfer from roots to shoots (Mahaffey et al., 1975; Wiersma et al., 1986), and spinach is a known source of Cd to young children's diets (Pokharel & Wu, 2023). In a study where Cd, Hg, Pb, and As were added to soil, Cd accumulated more in the spinach tissues than the other metals (Chunilall et al., 2004). The concentration of Cd in spinach in the United States ranges 2 orders of magnitude from ~0.01 to 1.1 mg kg^{-1} FW (Brierley & Sanchez, 2017; P. J. Gray, 2023; Wolnik et al., 1985). These ranges are based on two surveys of paired soil and plant samples and on the Total Diet Study (Figures 1 and 2). In a joint USDA-FDA-EPA survey of metals in paired soil and crop samples collected in the early 1980s, spinach leaves from spinach-growing regions the United States ranged from 0.012 to 0.195 mg kg^{-1} FW with a geometric mean of 0.065 and median of 0.0605 mg kg^{-1} FW (Wolnik et al., 1985). Using that data and separating by the four states from where spinach samples were obtained, the sample with the lowest Cd was obtained in New Jersey, and the highest was from Texas (Figure 1). California and New Jersey had the lowest mean and Maryland and Texas had the highest; however, no sample was higher than 0.2 mg kg^{-1} FW in that study (Figure 1). The 1980 values were generally lower than those reflected in FDA's Total Diet Study (TDS), where samples of "spinach, boiled fresh frozen" collected 4 times per year from markets in 4 regions in the United States in 1991–2017 ranged from 0.03 to 1.09 mg kg^{-1} FW with a mean of 0.16 and median of 0.11 mg kg^{-1} FW (Figure 2).

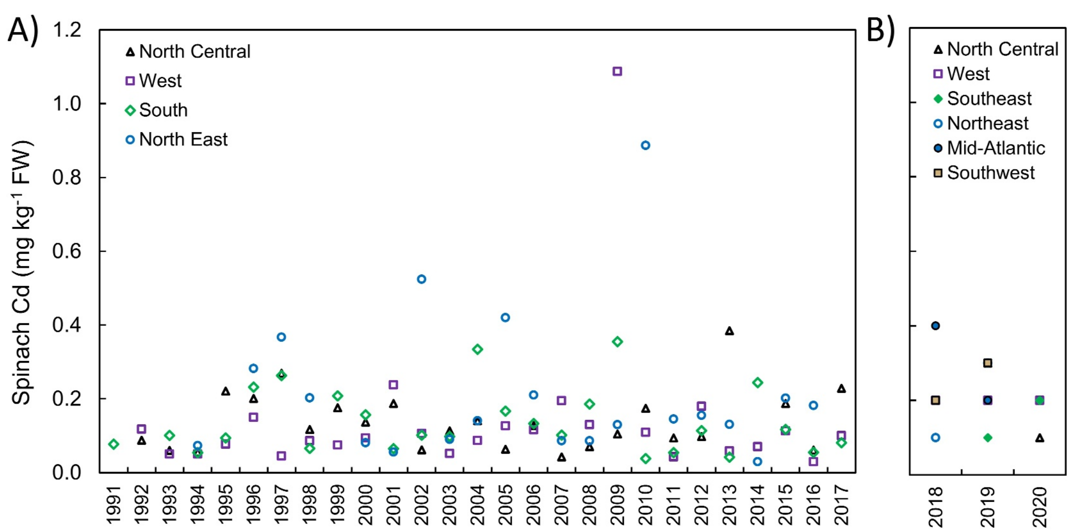

Figure 2. (a) Cd concentrations in spinach from the U.S. FDA Total Diet Study from 1991 to 2017. Spinach was collected from stores or markets in each of 4 regions, composited for the region, washed and cooked as consumers would for consumption, and then digested and analyzed for total metal content. (b) Cd concentrations in raw spinach food from the FTA Total Diet Study from 2018 to 2020. Raw spinach was collected from stores or markets in each of 6 regions, composited for the region, washed as consumers would for raw consumption, and then digested and analyzed for total metal content. Samples from each region of purchase do not necessarily represent the location in which they were grown.

Comparison of these data sets could suggest that the concentrations of Cd in spinach are increasing; however, this is likely not the case. Rather, a comparison of the soil collection maps (Holmgren et al., 1993) and the plant collection maps (Wolnik et al., 1983, 1985) from the 1980s reveal that while lettuce (*Lactuca sativa* L.) was sampled from the Salinas Valley where soil Cd was higher (∼1.2 mg kg^{-1}), spinach was only sampled farther north and just east of Monterrey Bay where soil Cd levels were lower (<0.16 mg kg^{-1}). Due to the current importance of the Salinas Valley region for spinach production in the United States, samples from this region more than likely represented a large proportion of the samples obtained in the TDS. The TDS is meant for consumer exposure estimates and does not include paired soil samples, so unfortunately, direct comparisons of plant concentrations and soil levels are not possible with the TDS. In fact, very limited data exists on paired soil and plant Cd levels for leafy greens in the United States. A recent data set from a non-peer-reviewed report (Brierley & Sanchez, 2017) paired samples of soils and spinach collected from the desert soils of southeastern California and southwestern Arizona, and values from 2016 ranged from 0.063 to 0.453 with a mean of 0.226 and median of 0.221 mg kg^{-1} FW despite the soil Cd levels being relatively low (∼0.1–0.3 mg kg^{-1} Cd). Clearly, new peer-reviewed data is needed that pairs soil and plant levels of Cd in leafy greens in the United States to fully understand the magnitude of potentially unsafe levels in leafy greens like spinach.

It is critical to obtain data on Cd concentrations in spinach to aid in the development of food safety regulations. If a 0.2 mg kg^{-1} FW Cd limit were established, as the European Union has done, then 0% of the samples collected in the 1980s would be out of compliance. In contrast, 40%–50% of the California and Arizona samples collected more recently would be out of compliance. The Total Diet Study recently underwent changes to sampling for spinach and now separates the United States into 6 regions and does not cook the food prior to analysis, as consumption has trended toward raw spinach. The newer data show that Cd in raw spinach ranges from 0.1 to 0.4 mg kg^{-1} with a mean of 0.222 and median of 0.2 mg kg^{-1} FW (Figure 2b) and would result in 37% of samples out of compliance if the 0.2 mg kg^{-1} FW European limit were established in the United States.

2.2. Pb in U.S. Spinach

Unlike Cd, Pb concentrations are generally low in spinach leaves and appear to be decreasing over time. The concentration of Pb in spinach in the 1980s studies ranged from 0.016 to 0.166 mg kg^{-1} FW with a mean of 0.045 and median of 0.038 mg kg^{-1} FW (Figure 3) (Wolnik et al., 1985). These concentrations were higher than those reflected in FDA's Total Diet Study, where samples of "spinach, boiled fresh frozen" collected 4 times per year from markets in 4 regions in the United States in 1991–2017 ranged from below detection to 0.062 mg kg^{-1} FW with a mean and median of 0.008 mg kg^{-1} FW. This decrease may reflect less atmospheric deposition on soils and leaf surfaces due to the phase out of leaded gasoline and other environmentally driven changes in industrial processes (Galal-Gorchev, 1993). Despite this decrease in Pb concentrations and the limited transfer of Pb from roots to shoots, the TDS data reveal that approximately 30% of samples exceed the 0.01 mg kg^{-1} limit in vegetables suggested in the FDA draft guidance on Pb in foods intended for babies and young children. The newer FDA TDS of Pb in raw spinach collected from 2018 to 2020 were lower (Figure 4b), ranging from below detection to 0.018 mg kg^{-1} FW with mean and median of 0.006 mg kg^{-1} FW and 15% of samples exceeding the draft limit. Since the FDA considers food policy intended to keep consumers safe, it is also paramount to understand both the magnitude of decrease in metal content needed and how to practically meet that goal.

3. Root Cause of Cd and Pb Accumulation in Spinach

It is paramount to understand the soil and plant factors that dictate contaminant accumulation in edible tissues to identify mitigation strategies for metals in foods. Whether a metal is phytoavailable in soil depends first on the ability for the soil to retain the metal (via adsorption and/or co-precipitation), second, whether the plant can affect the metal's mobility via chelation, reduction, or precipitation thereby affecting root uptake, and third, how readily the metal is transported from roots to the edible portion of the plant. For spinach, the edible portion is the leaves, so metals like Pb that are highly retained in soil and have limited mobility from roots to shoots are less likely to be found in high levels in spinach leaves. In contrast, Cd can be mobilized in soil and is readily transferred from roots to shoots; see McLaughlin et al. (2021) for a general review of these processes in agricultural systems. A conceptual diagram highlights the contrasting behaviors of Cd and Pb in the soil-to-spinach environment (Figure 5), discussed in detail below.

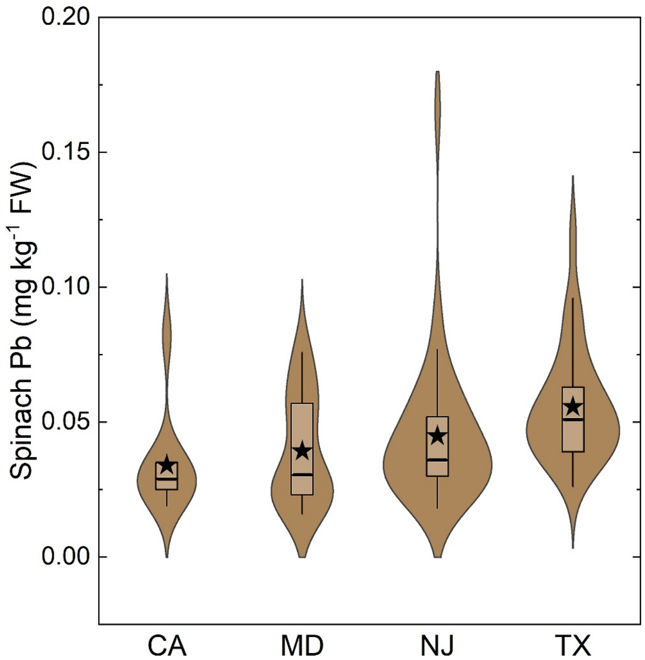

Figure 3. Violin plots of Pb concentrations in spinach leaves found in samples collected from CA (n = 19), MD (n = 20), NJ (n = 35), and TX (n = 31) between 1980 and 1982. Data are from Wolnik et al. (1985). The shaded area shows a kernel density plot, stars denote the mean, boxes denote the 25th and 75th percentiles, and the horizontal line denotes the median.

3.1. Soil Factors Affecting Cd in Spinach

Cd is taken up by plant roots primarily as the divalent cation, Cd^{2+}, or complexed with chloride, but may be present in soil solution as a natural organic matter chelate, which affects its mobility. In soil solution, soluble Cd is mainly chelated by low-molecular weight organic molecules, which keeps free metal ion activity low but increases soluble metal concentration many-fold. Most surface-bound metals in soils remain in equilibrium with the chelated soluble metals in the soil solution. Soils with a higher capacity to adsorb metals have a lower potential for plant uptake. Soils rich in organic matter, clays, and hydrous oxides of Fe and Mn have high metal sorption capacity and result in lower equilibrium values of free metal activity in soil solution (Mench et al., 2020; Sappin-Didier et al., 1997). For these reasons, Cd uptake is affected by soil texture and acidity, with sandy soils leading to more mobility and higher uptake than finer textured soils (Dheri et al., 2007).

Unlike most other metals, the concentration of Cd in higher plants is directly proportional to the concentration of Cd in the soil. Linear relationships have been found with soil total Cd (<60 mg kg^{-1}) and plant Cd in a wide variety of plants including lettuce, rice, broccoli (*Brassica oleracea* var. *italica*), spinach, barley (*Hordeum vulgare* L.), carrot, beet (*Beta vulgaris* subsp. *vulgaris*), and potato (*Solanum tuberosum* L.) (Dudka et al., 1996; Kukier & Chaney, 2002; Kukier et al., 2010; Liang et al., 2013; McGrath et al., 2000; Smith & Hartz, 2016). Soil pH is more important than other soil properties, and soil Cd:Zn ratio strongly affects plant Cd levels because most Cd is absorbed through the Zn uptake transporter (M. Wang et al., 2023). There is also a linear relationship that exists between total soil Cd and soil Cd that is extractable by diethylenetriamine pentaacetate (DTPA-extractable Cd), which approximates total soil Cd by a factor of 2 or 3, depending on the soil (Brierley & Sanchez, 2017; Smith & Hartz, 2016; Xiao et al., 2018). From a laboratory perspective, soil DTPA extraction is less time-consuming and

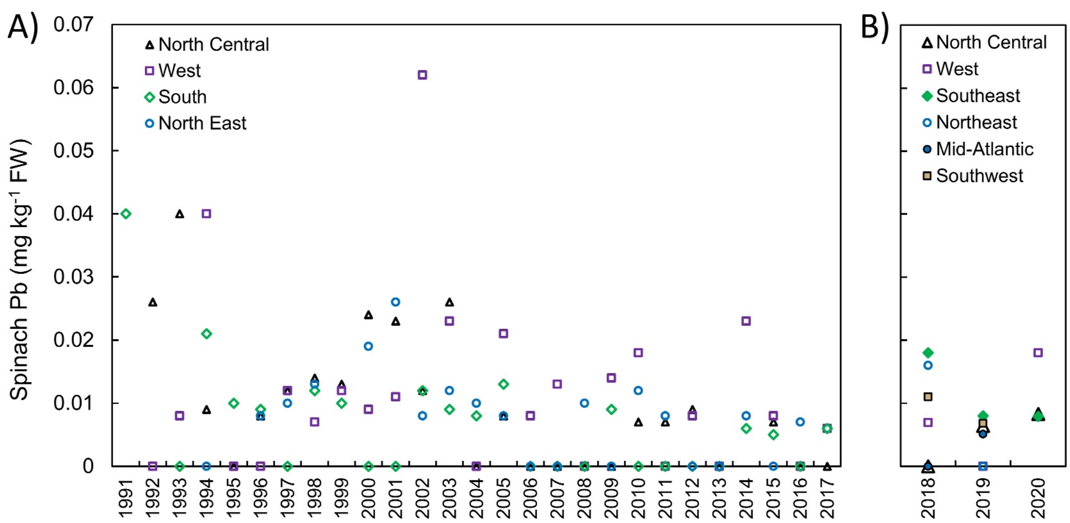

Figure 4. (a) Pb concentrations in spinach food from the FDA Total Diet Study from 1991 to 2017. Spinach was collected from stores or markets in each of 4 regions, composited for the region, washed as consumers would for consumption, and then digested and analyzed for total metal content. (b) Pb concentrations in raw spinach food from the FTA Total Diet Study from 2018 to 2020. Raw spinach was collected from stores or markets in each of 6 regions, composited for the region, washed as consumers would for raw consumption, and then digested and analyzed for total metal content. Samples from the four regions of purchase do not necessarily represent the location in which they were grown. Dashed line indicates the draft guidance (Center for Food Safety and Applied Nutrition, 2023) for Pb of 10 μg kg^{-1} FW in foods intended for babies and young children with a single vegetable ingredient.

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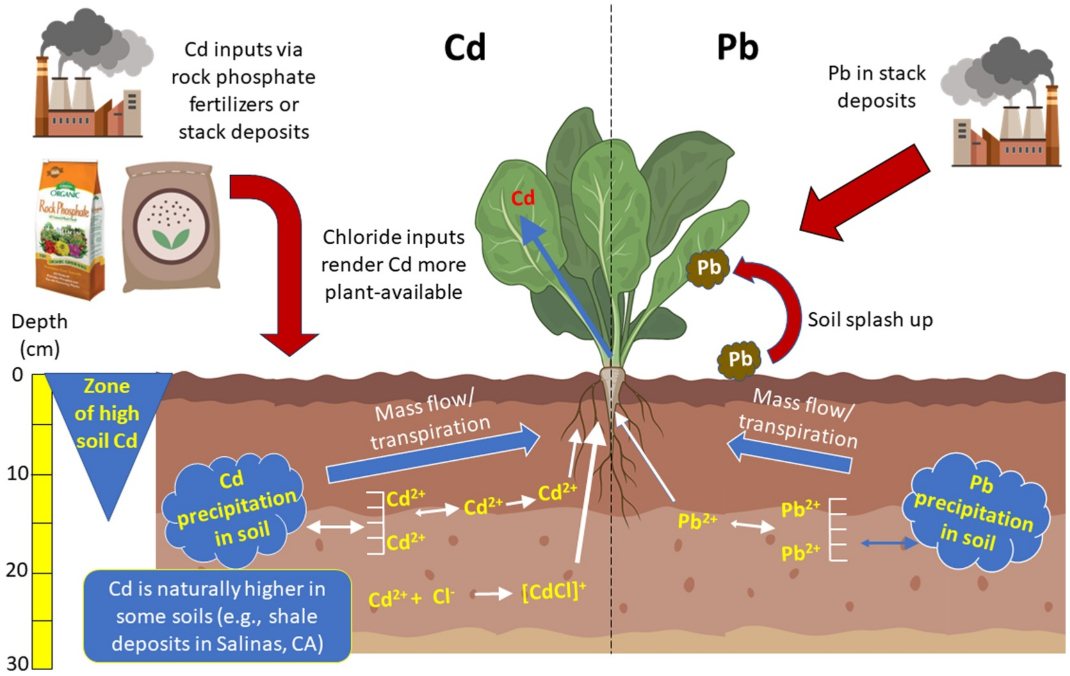

Figure 5. Conceptual diagram contrasting factors that influence Cd and Pb levels and mobility in soils created with elements from biorender.com. Cd and Pb are naturally present in all soils at trace levels, but Cd can be naturally higher in soils developed from shale parent material including soils in the spinach-growing region near Salinas, CA. Also, Cd can be added to soil via aerial deposition or from rock phosphate-derived fertilizers that are naturally enriched in Cd. Pb is added to soil mainly through areal deposition and direct soil contamination with Pb-rich sources (e.g., Pb-based paint products). Soil factors affecting Cd in spinach include precipitation and adsorption reactions with soil organic matter and Fe-Mn oxides as well as complexation with anions such as chloride, which renders Cd more mobile in soil and phytoavailable, where it can readily be absorbed by roots and be translocated to leaves. In contrast, Pb is strongly retained in soil via precipitation reactions and is sparingly soluble, where it can be absorbed by roots but not readily translocate to leaves; Pb associated with leafy greens likely is due to dust or fine soil particle accumulation on the leaf surfaces and improper washing (McBride et al., 2014).

is, therefore, less expensive than total soil Cd analysis and less subject to errors in analysis, so it is likely a better technique for higher throughput of soil screening for estimating Cd levels in plants. Because of the direct relationship between plant and soil Cd concentrations (either total soil Cd or DTPA-extractable Cd), spinach plants grown in soils with higher soil Cd have higher shoot Cd concentrations, which is mainly due to plant uptake by roots and translocation to the leaves. It should be noted that soil splash of Cd-rich soil onto the outside of leaves could also contribute to exposure if the soil is not properly washed from the leaves prior to consumption. In the United States, soil Cd measured in the early 1980s showed that the median concentration of Cd in agricultural soil is 0.2 mg kg^{-1} with a maximum of 2 mg kg^{-1} in soils derived from the Monterey Shale parent material (Holmgren et al., 1993) including Salinas Valley, CA, where spinach is currently a major crop. But studies of the shale-derived soils with high Cd and Cd:Zn ratio contained 5.2 mg Cd and only 54 mg Zn kg^{-1} (Chaney et al., 2017; Paul & Chaney, 2017).

Research on mechanistic understanding has revealed the important role of soil Zn concentration in Cd accumulation in leafy greens, including spinach. In particular, the high Cd soils derived from marine shales such as those in the Salinas Valley have low total Zn, and this relatively high Cd:Zn mass ratio (∼0.03–0.10 rather than <0.01) renders plants grown in these soils prone to Cd accumulation compared to most other soils with a low Cd:Zn ratio (US median about 0.003) (Holmgren et al., 1993). Moreover, these unique soils have even lower phytoavailable Zn relative to Cd based on phytoavailable soil extractions (Paul & Chaney, 2017), which makes plants grown on them prone to Cd uptake (Chaney, 2010). Research has shown that Cd enters the plant root through Zn uptake transporters in all crops except rice, so added Zn can inhibit Cd uptake by roots (M. Wang et al., 2023).

Soil pH is a master variable that affects the phytoavailability of metals, especially Cd (and Zn), by affecting several soil properties. In general, Cd is much more phytoavailable under acidic soil conditions. Theoretically, the activity of divalent cations like Cd in soil should increase 100-fold for each unit decrease in pH. As pH increases, adsorption of Cd in soil increases. Thus, for soils with pH above 7, Cd phytoavailability is limited (Yi et al., 2020). pH also has effects on how the ionic strength affects Cd adsorption. In general, Cd adsorption decreases with increasing ionic strength (Naidu et al., 1994), except when the increase in ionic strength is due to chloride, which further increases phytoavailability of Cd in a different way.

Chloride is known to increase solution Cd concentrations and plant uptake of Cd in diverse crops. Several studies have demonstrated relationships between soil or solution Cl^- and Cd accumulation in field-collected plants, including wheat (*Triticum aestivum* L.) (Norvell et al., 2000), sunflower (*Helianthus annuus* L.) (Li et al., 1994), swiss chard (*Beta vulgaris* subsp. *vulgaris*) (Smolders et al., 1998), potato (McLaughlin et al., 1994), and spinach (Brierley & Sanchez, 2017; Smith & Hartz, 2017). In general, increasing ionic strength decreases adsorption of cations like Cd^{2+}, so in theory an increase of Cl^- could increase the ionic strength and limit retention of Cd^{2+} on exchange sites thereby increasing Cd phytoavailability. However, studies have shown that the increase of plant Cd concentrations with increasing Cl^- is specific to the addition of Cl^- (rather than an ionic strength effect) due to the complexation of Cd^{2+} with Cl^-, forming chloride complexes with Cd (e.g., $CdCl^+$) in soil solution that are more mobile in soil than Cd^{2+} thereby increasing Cd phytoavailability. This mechanism has been demonstrated experimentally where the addition of Cl^- salts (e.g., NaCl) increases total Cd in soil solution whereas the addition of NO_3^- salts (e.g., $NaNO_3$) does not (Khoshgoftar et al., 2004; Smolders et al., 1998). These studies and others that show increased plant uptake of Cd with increasing Cl^- but not NO_3^- at the same ionic strength (Cabrera et al., 1988) illustrate that the enhanced phytoavailability of Cd due to chloride is not simply an ionic strength phenomenon. Instead, the $CdCl^+$ is either taken up directly by plant roots (Smolders & McLaughlin, 1996) possibly through ion channels for monovalent cations (Welch & Norvell, 1999) or the enhanced diffusion of Cd^{2+} through the plant apoplast by Cl^- complexation (Smolders et al., 1998). Either way, higher soil solution Cl^- increases the phytoavailability of Cd to plants. It should also be noted that nutrient solution studies where Cd is being investigated should be interpreted with caution if the salts used for the nutrient media or Cd delivery also contain Cl^- (and thus $CdCl^+$ complexes) (e.g., Michalska & Asp, 2001; Salaskar et al., 2011; Valderrama et al., 2013; K.-S. Wang et al., 2008).

In addition to increasing Cd phytoavailability, increasing soil solution Cl^- also decreases the phytoavailability of Zn. In one study, increasing Cl^- increased solution phase and plant Cd concentrations while decreasing solution phase and plant Zn in wheat shoots grown in Cd-contaminated soil (Khoshgoftar et al., 2004). The decrease in phytoavailability of Zn and increase in phytoavailability of Cd with increasing Cl^- illustrates why it is imperative to add more Zn than suggested from traditional soil test recommendations. Fertilizer recommendations are based on extraction of a nutrient from soil and do not factor in additional effects of Cl^- from irrigation water on Zn phytoavailablity. Khoshgoftar et al. (2004) found that the increase in Cd and decrease in Zn caused by added Cl^- was nearly alleviated by the incorporation of $ZnSO_4$. These findings have important implications for Cd mitigation strategies, which are detailed in Section 4.

The soil reduction-oxidation (redox) potential can also influence Cd mobility in soil and plant-uptake. Under low redox (i.e., reducing) conditions and in acid soils, sulfate can be reduced to sulfide, which can precipitate with Cd, forming sparingly soluble CdS (Arao et al., 2009; Reddy & Patrick, 1977). Flooding raises soil pH to neutral levels, but after drainage soil pH can rapidly decline and sharply increase Cd uptake. Because most crops are grown in oxic soils where redox conditions are high, Cd is not commonly immobilized by sulfide, with the exception of rice (Arao et al., 2009; Fang et al., 2021; Kukier & Chaney, 2002; Reddy & Patrick, 1977). Rice is typically grown under flooded paddy conditions where Cd mobility and uptake are low, but Cd mobility increases when alternative water management of periodic or prolonged drainage occurs, causing increased plant uptake under more oxic conditions relative to paddy rice. In paddy rice, most grain Cd is absorbed after the fields are drained at the beginning of flowering, allowing most grain Cd to be absorbed by roots during that flowering period (Simmons et al., 2008).

3.2. Plant Factors Affecting Cd in Spinach

Once delivered to the rhizosphere, Cd enters plants due to absorption through the root apoplast and transport through the symplast. Chelated or complexed Cd^{2+} can dissociate in the rhizosphere, and this buffering capacity of the soil solution can drive the equilibrium toward more free metal as the plant roots absorb Cd^{2+} (Degryse et al., 2009), creating a diffusion gradient from the bulk soil to the root apoplast. Similar to other toxic elements with no known biological function (e.g., Hg, As, Pb), transport from the symplast to the apoplast occurs via transporters intended for plant nutrients. These toxic ions can be transported into roots because they have similar valence, hydrated ionic radius, and chelation properties to plant nutrients. In spinach and in most other crops, the main mechanism of Cd^{2+} uptake is mainly via the Zn^{2+} uptake pathway (M. Wang et al., 2023). One contrasting crop is rice, where the Mn uptake transporter (OsNramp5) is responsible for Cd^{2+} uptake (Ishikawa et al., 2012; Sasaki et al., 2012; M. Wang et al., 2022). For a detailed review of the mechanisms of Cd accumulation in higher plants, see (Sterckeman & Thomine, 2020).

Once inside the root symplast, Cd is transported to the stele and pumped into the xylem, where it flows to aboveground tissues and accumulates in spinach leaves. Most plants, including spinach, have lower levels of Cd in leaves than in roots (Jarvis et al., 1976; McKenna et al., 1993), but spinach accumulates 2–3 times more Cd in the leaves than other crops (Cavanagh et al., 2019; Salim et al., 1995; Verma et al., 2007; Wolnik et al., 1983, 1985; Yi et al., 2020). Cd transport through the xylem is driven by transpiration and can occur as the free metal and as a complex with organic acids (Sterckeman & Thomine, 2020). For example, citrate is thought to aid in transport of Cd through tomato (*Solanum lycopersicum* L.) xylem (Senden et al., 1995). In hyperaccumulators, citrate, malate, and oxalate form complexes with Cd and these are thought to aid in tolerance of high Cd tissue concentrations upon vacuolar storage (Ueno et al., 2005; Villafort Carvalho et al., 2015). In the *Amaranthaceae* family, which includes spinach, oxalate accumulates in high amounts in the leaves, and can form insoluble crystals with Ca that are deposited in leaves (Siener et al., 2006). The co-localization of calcium oxalate crystals and Cd in some plants (e.g., Blommaert et al., 2024; Pongrac et al., 2018; van Balen et al., 1980; Villafort Carvalho et al., 2015) suggests that Ca-Cd oxalate crystals may also form in spinach and may be associated with higher levels of Cd in spinach leaves, as oxalate appears to increase with exposure to Cd (Sembratowicz & Rusinek-Prystupa, 2012; Villafort Carvalho et al., 2015). Joshi et al. (2021) argue that the formation of calcium oxalate crystals in spinach is detrimental to human health because the calcium within the insoluble crystals is less bioavailable and the crystals can lead to kidney stones. However, from a Cd standpoint, such crystals could render Cd less bioavailable to humans upon ingestion. It has been demonstrated that less Cd is bioavailable upon consumption of spinach compared to lettuce (Buhler, 1985; McKenna et al., 1992), the latter of which is not known to produce calcium/Cd oxalate crystals. To our knowledge, an analysis of Cd oxalate crystal formation across spinach cultivars has not been conducted, but perhaps low oxalate cultivars may lead to less Cd sequestration as oxalate crystals in spinach leaves or perhaps higher Cd sequestration in oxalate crystals would be less bioaccessible to humans upon ingestion. During spinach processing in animals, the pH of the stomach is quite acidic and Ca oxalate is soluble; but when the digesting food is moved to the small intestine, pH is raised which causes precipitation of Ca oxalate with co-precipitation of Cd in the crystals, reducing Cd bioavailability by about 50%.

Whether leaves are young or old when harvested has substantial impacts on concentrations of Cd and other nutrients in leaves. McKenna et al. (1993) grew spinach and lettuce in hydroponic media with variable Cd and Zn exposure and showed that the harvested young leaves consistently had lower Cd concentrations than harvested old leaves by approximately one order of magnitude. This finding is consistent with transpiration as the main driver of Cd accumulation in both spinach and lettuce leaves, as has been demonstrated for perchlorate in lettuce (Seyfferth et al., 2008). In the same experiment, McKenna et al. (1993) showed that Zn concentrations were less affected by the age of the leaf, which implies that plants grown for shorter durations (i.e., baby spinach) may have lower Cd: Zn ratio than those grown for longer periods.

The relative proportions of Cd and other nutrients such as Zn, Ca, and Fe in edible leaves have important implications for Cd bioavailability to humans. McKenna et al. (1992) illustrated that less Cd was absorbed and retained in the kidney and liver of quail that were fed lettuce and spinach with high-Zn than those with low-Zn at a similar leaf Cd concentration. This effect has also been demonstrated in rice, where low grain levels of Fe, Zn, and Ca, along with mild deficiency of these elements in subsistence diets results in 10-fold higher absorption of Cd in animals, including humans (Chaney, 2015; Reeves & Chaney, 2008). These findings suggest that increasing Zn, Ca, and Fe in spinach leaves or in the diets of spinach consumers will lessen the impact of a given Cd leaf concentration on the human body. Furthermore, it is important to consider not just the concentration of the element in the edible food tissue, but also the bioavailability of that element to the human, especially Fe, Zn and Ca (Buhler, 1985; Fox, 1979, 1988). Most Cd is absorbed on the ferrous uptake transporter of the intestine, so Fe deficiency strongly promotes Cd absorption (Reeves & Chaney, 2008).

3.3. Soil Factors Affecting Pb in Spinach

Pb is strongly retained in soil, and therefore it is much less phytoavailable to spinach than Cd and rarely reaches high levels in shoots. Similar to Cd, Pb sorbs onto organic matter, metal oxides, and clays, is more available as pH decreases, and is more strongly retained in finer textured soils with higher cation exchange capacity (CEC) (Brümmer & Herms, 1983; Farrah & Pickering, 1977; Forbes et al., 1976). In addition to sorption processes, Pb is also strongly retained in soil by precipitation as insoluble or sparingly soluble crystalline minerals such as pyromorphite (Cotter-Howells & Thornton, 1991; Mench et al., 2020). These sorption and precipitation reactions render Pb far less soluble in soil solution (i.e., sub nM), and therefore is less phytoavailable, than Cd (McLaughlin et al., 1999).

However, research on Pb in leafy vegetables grown in Pb-rich urban gardens found elevated Pb levels. Further, Pb contamination derives from soil particles adhering to the leaves, not from uptake within the plant (McBride et al., 2014). Crop types with little chance of soil or dust contamination are thus seldom enriched in Pb.

3.4. Plant Factors Affecting Pb in Spinach

Pb that is solubilized in soil can move into the root apoplast, but its uptake into the symplast for transport to plant shoots is limited (see Pourrut et al., 2011 for a detailed review). While ion channels such as those for Ca^{2+} may also transport Pb^{2+} (Huang & Cunningham, 1996), approximately 95% of the Pb that is absorbed by roots stays in the roots (Pourrut et al., 2011) and is precipitated with insoluble P minerals in the rhizosphere or root cells (Chaney & Ryan, 1994). For this reason, consumption of root vegetables such as carrots or sweet potatoes have a higher risk to human health than leafy vegetables such as spinach or stem tubers such as potato (Codling & Onyeador, 2017; Codling et al., 2015, 2016). In peeled carrots, the Pb is mostly within the xylem in the center or core of the carrots (Codling et al., 2007). Thus, garden crops that are phloem-fed, such as potatoes and fruits, remain low in Pb because Pb has low solubility in phloem fluid, but xylem-fed crops, such as the expanded hypocotyl root vegetables accumulate Pb as pyromorphite (Pb-phosphate compound) which reduces Pb transport to shoots. Thus, soil splash contamination is important for low-growing leafy crops, but uptake and trapping within the food is important in xylem-fed root crops.

It is often difficult to determine if the Pb found associated with spinach leaves is primarily located on the outside of the leaf or if it was transported to the leaf via symplastic uptake and translocation. Because Pb is strongly adhered to soil particles, any such particles that are dusted onto the plant leaves and are not thoroughly washed off can be mistaken for uptake into the leaf (Amir et al., 2018; Peterson, 1978). Spinach has a high surface roughness, which contributes to its ability to harbor food pathogens such as *Escherichia coli* even after washing (Truschi et al., 2023), so it is reasonable to assume that Pb-bearing soil particles could also be entrapped within the rough surface of spinach leaves and remain after washing. There is also some emerging evidence that heavy metals such as Pb could be directly absorbed through the leaf from the atmosphere (Shahid et al., 2017), a mechanism that has been demonstrated for lettuce (Schreck et al., 2012).

4. Mitigation Strategies for Cd

Before any remediation technique is considered, it is first recommended to obtain a soil test for Cd and Zn. The Western Growers Association (WGA) in conjunction with University of California Cooperative Extension has developed guidance for such testing and for remediation steps in California (Smith, 2018). These guidelines recommend that if a soil has lower than 0.2 mg kg^{-1} total Cd, there is no need for mitigation and leafy greens can be safely grown. To our knowledge, other states do not have such guidelines, but it is probably relevant for most soils. If instead of total soil Cd, a DTPA test is used, then a level less than 0.1 mg kg^{-1} DTPA-extractable Cd most likely is safe for growing leafy vegetables. If the value is higher, then either avoidance is needed (e.g., if soil Cd is ~5 mg kg^{-1} or higher) or mitigation steps are needed to produce low-Cd products.

To mitigate human health risks of Cd and Pb via spinach consumption, we will divide such strategies into pre-harvest techniques and post-harvest techniques (Table 1). Pre-harvest techniques are generally actionable by the grower and include soil preparations and amendments, irrigation practices, and cultivar selection. Post-harvest techniques are further downstream and include spinach handling in the field, washing either by a factory or by the consumer, and other consumer-driven choices regarding diets. We will also consider the feasibility of each mitigation strategy through the results of stakeholder surveys.

Table 1
Summary of Effective Pre- and Post-Harvest Techniques to Minimize Cd Exposure From Spinach

Method	Mechanism of Cd reduction in plant: Uptake competition	Mechanism of Cd reduction in plant: Improves growth	Mechanism of Cd reduction in plant: Limits soil on leaf	Mechanism of Cd reduction in soil: Limits Cd phytoavailability	Cd reduction	Notes
Cultivar Selection	✓	?	–	–	0%–50%	New breeding shows range
Organic Matter Amendment	–	?	–	✓	0%–75%	Depends on many factors
Lime Amendment	–	✓	–	✓	~60%	Better with Zn amendment
Zinc Amendment	✓	–	–	–	20%–80%	Differences based on Zn source and soil
Lime + Zn Amendment	✓	–	–	✓	~60%	Highly effective
Limit Chloride Inputs	✓	–	–	✓	40%	Highly effective
Leaf Washing	–	–	✓	–	8%–15%	Minimal effectiveness
Eat a balanced diet with replete Zn, Fe, Ca	–	–	–	–	≥50%	Minimizes bioavailability of Cd

Note. The % reduction column is estimated based on the literature cited within this review.

4.1. Pre-Harvest Techniques for Cd Mitigation

4.1.1. Soil Amendments

Because pH is a master variable that controls Cd dissolution in soil, it can be manipulated to control Cd phytoavailability. In some areas of the United States such as the Mid-Atlantic or Northeast, soils are slightly acidic (Holmgren et al., 1993) and thus liming could be effective for reducing Cd phytoavailability. If soils are acidic, liming the soils to above pH 7.2 is effective at decreasing the phytoavailability of Cd in spinach and other leafy greens (Cavanagh et al., 2019; Chaney et al., 2009; Kumarpandit et al., 2017; Pandit et al., 2012; Smith & Hartz, 2016; Yi et al., 2020). However, in the Salinas Valley of California, soils are already limed to above pH 7.2 to decrease the incidence of club root in cruciferous crops (brassicas) (Smith & Hartz, 2016), so additional liming is probably not advisable unless adding large amounts of Zn sulfate, which causes soil acidification. In addition, liming these soils can even increase Cd accumulated by spinach because liming reduces soil Zn phytoavailability which induces more expression of Zn transporters that causes increased uptake of Cd (Chaney et al., 2017).

The addition of Zn as a soil amendment can be effective to decrease Cd concentrations in plant leaves such as spinach because Cd^{2+} is taken up by Zn^{2+} transporters in all plants except rice, but several other factors must also be considered. Because the anion affects how metals interact with the soil surface, different Zn salts behave differently as soil amendments. Using the sulfate salt results in more adsorption of Zn in soil compared to nitrate or chloride salts (Shuman, 1986). Of several sources tested, Zn sulfate and Zn chelate most decrease Cd in crops and Zn sulfate is cheaper than Zn chelate, so Zn sulfate amendment is the preferred choice (Smith & Hartz, 2017). However, Zn sulfate can also acidify the soil, so it is important to add this material with lime to keep Cd phytoavailability low (Paul & Chaney, 2017). Californian agronomists have established best practices for reducing Cd in spinach, and they suggest that powdered forms are more effective than granular forms, foliar forms are not effective, and incorporating the material into the rooting zone (0–30 cm) is best (Smith & Hartz, 2016). However, practically speaking it may be challenging to incorporate Zn that deep, but disking (10–15 cm incorporation) can be done, which is much better than top-dressing.

Most importantly, the amount of Zn that should be applied is higher than what would be recommended to avoid Zn deficiency based on a soil fertility test. In one field trial, the application of 100 or 200 lbs $acre^{-1}$ Zn (or 277 and 555 lbs $acre^{-1}$ $ZnSO_4$, equivalent to 310 and 622 kg $ZnSO_4$ ha^{-1}) to soil with 2.4 mg Cd kg^{-1} and ~3 mg Zn kg^{-1} reduced Cd in lettuce by 40% and 55%, respectively, resulting in spinach Cd concentrations of 0.22 and 0.17 mg kg^{-1} fresh weight, respectively (Smith & Hartz, 2017). Similar reductions of ~50% in spinach Cd concentrations were observed in a different soil with 1–1.2 mg Cd kg^{-1} and 1.7 mg Zn kg^{-1} where the application of just 100 lbs $acre^{-1}$ Zn (or 277 lbs $ZnSO_4$ $acre^{-1}$, equivalent to 310 kg $ZnSO_4$ ha^{-1}) decreased spinach Cd concentrations from 0.29 to 0.15 mg kg^{-1} fresh weight (Smith & Hartz, 2017). The high rate of Zn application can sustain multiple years of effectiveness, but additional application will be necessary approximately every 3 years.

The Zn application has been most successful in Cd-rich soils with high Cd:Zn ratio of the Salinas Valley and adjacent western soils in California where Zn phytoavailability is also low (Paul & Chaney, 2017). Increasing phytoavailable Zn relative to Cd can suppress uptake of Cd in spinach, but it is important to keep Zn lower than the level that could cause phytotoxicity. Other work in New Zealand soils showed that Zn addition was effective at decreasing Cd in spinach when Zn was adequate for growth and soil Cd:Zn ratio was low (C. W. Gray & Wise, 2020).

Organic-rich soil amendments can also be effective in decreasing Cd uptake into spinach. Incorporating organic amendments such as compost, biosolids, biochar, and humic acids increases the soil cation exchange capacity (CEC), providing a reactive surface for Cd sorption and retention (Degryse et al., 2009; Kumarpandit et al., 2017; Paul & Chaney, 2017; Prokop et al., 2003; Sato et al., 2010). In addition, Zn sulfate addition with compost and lime worked better than Zn sulfate alone in decreasing Cd uptake but did so at a significant yield reduction (Paul & Chaney, 2017). Amendment with biochar provides inconsistent results (Hu et al., 2020; Younis et al., 2016) probably because of the variable properties of biochar from batch to batch and across different feedstocks (Shaheen et al., 2019).

4.1.2. Minimize Cl^- and Cd Inputs

Because Cl^- can form a $CdCl^+$ complex that increases Cd phytoavailability (Smolders et al., 1998), limiting Cl^- in irrigation water and from inputs can reduce Cd uptake. Typically, irrigation is from surface water, well-pumped groundwater, or recycled water. The amount of Cl^- in water sources can vary with the use of road salts, fertilizers, treatment of irrigation water with Cl-bearing disinfectants, and the use of recycled water for agricultural irrigation. Leafy greens like spinach are particularly vulnerable to human pathogens from irrigation water, necessitating treatment of irrigation water to minimize outbreaks; Cl-bearing chemicals are the disinfectant of choice (for detailed review, see Dandie et al., 2020). Recycled water use and treatment of irrigation water with Cl-bearing chemicals as disinfectants increases Cl^- in the soil. While this increase is not deleterious to plants (Platts et al., 2014), it could enhance Cd phytoavailability, so the impact of Cl-bearing chemicals as disinfectants and recycled water use on Cd phytoavailability in spinach growing areas should be explicitly tested. In one study, the concentration of Cd in spinach increased by 0.25 mg kg^{-1} (dry weight) for each 1 meq L^{-1} increase in Cl^- concentration in irrigation water (Smith & Hartz, 2017). In addition, Cl^- can enter the soil as the counter ion from other fertilizer salts (e.g., KCl), so fertilizers with Cl^- should be avoided in fields where Cd is prone to uptake into spinach. Finally, because Cd can be a contaminant of other fertilizers (e.g., rock phosphate) (Mulla et al., 1980), these potential sources should be monitored and even minimized by lowering Cd limits in P-fertilizer for spinach soils to avoid adding more Cd to soils.

4.1.3. Cultivar Selection

Growers also could select spinach cultivars that have low Cd accumulation potential or low oxalate, the latter of which may decrease leaf levels of Cd (but increase the bioavailability of the Cd still absorbed). Recent breeding efforts have identified cultivars that are genetically limited in their ability to accumulate Cd resulting in about a 50% difference in low versus high Cd concentrations, but the specific genes responsible are still being investigated (Greenhut, 2018; Hilborn, 2020; Osorio et al., 2018; Smith & Hartz, 2017). Thus, growers could soon begin to select low Cd cultivars. Cultivars that are low in oxalate may also limit Cd accumulation in leaves, so cultivars that have low oxalate can be healthier for humans. But lower oxalate cultivars may have higher Cd bioavailability due to the effect of oxalate on Cd absorption in the intestine. In addition to minimizing Cd accumulation, the low-oxalate cultivars can also minimize the potential for kidney stones from spinach consumption (Kawazu et al., 2003).

4.2. Post-Harvest Techniques for Cd Mitigation

4.2.1. Leaf Washing

Washing leaves can help to lessen the amount of Cd on spinach that arises from splash of soil particles. In factories that make processed spinach products, washing is the first step that occurs. Washing is typically done to remove bacteria as well as soil, and this step is effective for both. Flushing of the soil particles during this step is critical to ensure that downstream processes are not contaminated with soil that might contain metals. However, the equipment used for spinach processing may contain Cd in galvanized steel, and it is unknown if Cd leaches into the downstream product from the use of this equipment. Several authors have stated that the use of such equipment as well as Cd-plated utensils and Cd in pottery enamel is a source of Cd in the diet (Faroon et al., 2013; Galal-Gorchev, 1993; Schaefer et al., 2020), but these statements cited a JEFCA report (World Health Organization, 1989) in which the committee suggested that those products might release Cd into foods, but no references to the peer-reviewed literature were given. Other work showed that galvanized steel led to Zn contamination in soil and plants, but not Cd (Jinadasa et al., 1997; Jones, 1983); the Zn metal used in galvanizing must be low in Cd or the coating fails rapidly in outdoor weather, so Cd is about 100-fold lower in galvanizing Zn metal. It was previously determined that neither the processing of cereals to make flour and bread nor canning affected Cd concentrations (Galal-Gorchev, 1993). More research is needed to determine if processing spinach with galvanized equipment could contribute to higher Cd in spinach products, but to our knowledge, no data suggests that this occurs. At home, most consumers wash fresh spinach to remove soil particles. Adding small quantities of lime or lemon juice to the water wash could be more effective at removing soil particles, as was demonstrated previously by Amir et al. (2018), which showed that citric acid was more effective than water alone at removing surface Cd contamination.

4.2.2. Dietary and Consumption Changes

Several dietary and consumption considerations can affect the exposure of consumers to Cd from spinach. When spinach is harvested, it has approximately 93% water content. When consuming spinach raw, the amount of Cd per wet weight of leaf is diluted by the water within. When spinach is cooked on the stovetop, much of the water is driven off, and the Cd becomes more concentrated. This concept may be important for baby food manufacturers who cook spinach before canning. As noted earlier, because Cd transport and accumulation in leaves is driven by plant transpiration, older leaves with larger surface area have higher concentrations of Cd than newly formed "baby" leaves. A field study in New Zealand showed that "baby" spinach had lower Cd concentrations than the subsequent bunching spinach crop (Cavanagh et al., 2019). Additionally, plenty of evidence shows that bioavailability of metals to humans through ingestion is strongly affected by the nutritional status of the human. Diets low in Zn, Fe, and Ca can render any given Cd concentration more bioavailable (Chaney, Stoewsand, Bache & Lisk, 1978; Chaney, Stoewsand, Furr, et al., 1978; Chaney et al., 2004; Reeves et al., 2005). Likewise, high dietary Cd can decrease the absorption of those essential elements. Therefore, eating a balanced diet that is rich in Zn, Fe, and Ca can minimize the impact of Cd on human health (Fox, 1979; Reeves & Chaney, 2002, 2008). It has also been suggested that these micronutrients can be added to diets via nutritional supplements or fortification to lessen the impact of Cd on the human body (Chaney et al., 2004).

5. Mitigation Strategies for Pb

Pb is not readily accumulated in spinach leaves, and the concentrations of Pb in spinach are low and have decreased over time. Therefore, it would be a safe assumption that for most non-contaminated soils, Pb mitigation is not necessary to reduce Pb levels in spinach. However, it is noteworthy that the draft U.S. guidance for Pb of 10 μg kg^{-1} FW in foods intended for babies and young children with a single vegetable ingredient (Center for Food Safety and Applied Nutrition, 2023) might be challenging to achieve. With this draft guideline, 30% of the TDS samples would be non-compliant and exceed 10 μg kg^{-1} (Figure 4) and should not be used for the production of baby foods according to the "closer to zero" action plan. Therefore, mitigation steps might be necessary even for non-contaminated soils if the final product is intended for babies and young children.

First, a soil Pb test should be performed to identify any high Pb soils. Brown et al. (2016) suggest that leafy greens grown in soil with >900 mg kg^{-1} Pb should not be fed to children, while mitigation steps may be needed for soils with <900 mg kg^{-1} Pb. For Pb, pre-harvest techniques are focused on avoidance (e.g., using mulch to limit splash contamination; or replacing Pb contaminated urban soils with clean soil in raised beds) and soil stabilization, and post-harvest techniques are focused on washing, peeling, and consumer actions.

5.1. Pre-Harvest Techniques for Pb Mitigation

5.1.1. Soil Amendments

If needed, both organic matter addition (Degryse et al., 2009) and P fertilization (Codling, 2007) are helpful to decrease Pb phytoavailability. Because Pb is not readily translocated to leaves, there are limited actions that promote competition with Pb for uptake into spinach. Addition of organic matter and P help to stabilize Pb in soil and promote the formation of Pb phosphate and pyromorphite, which both have low Pb solubility (Cotter-Howells & Thornton, 1991). Using mulch to limit soil splash onto leaves of low growing crops, and using raised beds with low Pb soil are effective.

5.2. Post-Harvest Techniques for Pb Mitigation

5.2.1. Leaf Washing

Removing Pb-bearing soil particles from the leaf surface is critical to decreasing Pb ingestion through consumption of leafy greens. Studies have shown that better washing of leaves was effective at removing Pb from leaves of Swiss chard and from tomato fruit, but kitchen washing with just tap water was not as effective as laboratory washing with water and the surfactant sodium laurel sulfate (Attanayake et al., 2014). In another study, different washing techniques were tested, and washing with tap water removed 8% of Pb on spinach leaves, but this increased to 15%–26% with 5%–10% lemon juice extract (Amir et al., 2018), which among the washes tested might be the most practical for spinach consumers to do at home.

While this paper is focused on spinach, it is also important to consider post-harvest techniques that could be used for root and tuber vegetables, which have higher concentrations of Pb than spinach leaves. Peeling potatoes is effective at removing Pb-bearing soil particles that are adhered to the skin (Codling et al., 2016); however, peeling does not work well for sweet potatoes and carrots (Attanayake et al., 2014; Codling & Onyeador, 2017). Potatoes and yams (*Dioscorea* spp.) are phloem-fed tubers, which means that the part consumed is a modified stem, rather than a root. For these crops, Pb must be absorbed by the roots, translocated to the shoots, and then pumped into the phloem system for transport to the storage tissues. Phloem fluid is rich in phosphate which causes Pb to precipitate quickly so little can move to the potato, etc. Therefore, potatoes are more similar to spinach in that minimal Pb is translocated to the stem/leaves compared to the roots, and thus potatoes and yams have minimal Pb in their edible tissues if peels are not consumed. In contrast, sweet potatoes and carrots are root tubers and roots, respectively, and Pb is absorbed by the root xylem and concentrates there, as it is not readily transported to shoots. Therefore, peeling and washing sweet potatoes and carrots cannot remove the Pb contained within the hypocotyl xylem tissue (Attanayake et al., 2014; Codling et al., 2015). Therefore, if sweet potatoes and carrots were to be cultivated in Pb-rich soil, minimization of Pb in the edible tissue may require mitigation measures. Avoiding such soils for carrot and low-growing leafy vegetables is advised while tree fruit may be grown on high Pb soils without increasing Pb in the harvested fruit (Creger & Peryea, 1992).

6. Feasibility Considerations and Future Directions

We conducted interviews (Table 2) with stakeholders in the spinach sector to obtain their perspectives on mitigation strategies and the feasibility of implementation of such strategies. We attempted to obtain survey respondents from the four major spinach-producing states and from other states that produce minor amounts to gain a fuller reflection of spinach sector stakeholders. Despite our best efforts, we could not obtain respondents from Salinas Valley, California—where soil is naturally elevated in Cd. Nevertheless, we were able to obtain a total of 7 respondents who agreed to be interviewed for this study, allowing us to obtain viewpoints from stakeholders in the spinach sector from five states that included spinach producers, packers, processors, and marketers. Interviews occurred either via Zoom or in person where each respondent was asked the questions in Table 2 according to approved IRB protocols.

The general conclusion from these stakeholder conversations is that the spinach sector is willing to do what it can to reduce metals in foods, but some level of incentives, testing, and education is needed. Every grower we spoke with stated that soil amendments for spinach mitigation would be relatively easy to implement for growers/producers because spinach is already carefully monitored, and soil amendments are already being used. However, clear guidance and testing on where mitigation is necessary is needed, because soil testing for metals like Cd and Pb is expensive and is therefore not routinely performed in states other than in California. In addition, if Zn application were needed, it is important to educate producers that they would need to add more than what a soil fertility test estimate would suggest due to Zn deficiency and that it must be incorporated deeply into the rooting zone.

It is also important to recognize that growers often work on thin economic margins, so changes that incur additional costs for soil amendments and labor could affect the spinach market, and growers could choose to grow

Table 2
Interview Questions for Members of the Spinach Sector Used to Shape the Stakeholder View of Mitigation of Metals in Spinach

1.	Who are you, and what is your professional relationship to the food and agriculture sector?
2.	How would you rate your concern regarding metals (Cd or Pb) in the spinach you produce?
3.	What steps are you currently taking to mitigate metals in your product?
4.	What within your production system would you be willing to modify in order to reduce metals (Cd, Pb) in spinach?
5.	What would it take for you to implement practices that are known to reduce metals in spinach?
6.	If implemented, what type of incentive would you require to implement these practices?
7.	If implemented, how would it impact your operation-i.e., time, people, cost?
8.	Do you have two to five suggested contacts of other people we could interview?

a different crop than spinach if those costs were not offset. It is also clear that more availability of inexpensive and high throughput analysis of soil and water is needed to first evaluate if mitigation for metals is necessary. We suggest that regional governmental testing labs be established throughout production regions and operate at low or cost to growers for this purpose.

Minimization of chloride in soil used for spinach production is needed to reduce the phytoavailability of Cd, but this is currently challenging to accomplish for several reasons. First, irrigation water applied to leafy greens must have a disinfectant to reduce the risk of pathogens, and Cl-containing disinfectants are often the disinfectant of choice, which exacerbates the Cd issue. Thus, a grower could be forced to choose between reducing the risk of pathogens or reducing the risk of Cd in their product. Second, scarcity of water is a complex issue in CA, and recycling irrigation water is practiced but this increases salt concentrations, including Cl^-. Use of tertiary treated urban wastewater for irrigation has been shown to increase soil Cl^- in northern Salinas valley compared to use of well water (Platts et al., 2014). Third, seawater intrusion in coastal soils increases Cl^- concentrations, and Cl^- accumulates in poorly drained soils. Finally, the counter ion of some fertilizers is Cl^-. Research efforts should focus on minimizing Cl^- in the soil solution possibly by adsorption or absorption below ground or using an affordable and alternative disinfectant than Cl-based disinfectants.

Finally, consumers should also be more educated on the risks of metal intake via foods through educational programs. Such programs could be developed by science communication experts in conjunction with scientists and perhaps delivered via social media. Such a campaign could emphasize ways to lower risk of metal intake, such as washing of soil from vegetables, and metal absorption, such as eating a balanced diet with sufficient micronutrients to minimize the absorption of heavy metals in the body.

7. Conclusions and Future Directions

Guidance exists on promising pre- and post-harvest techniques to mitigate Cd and Pb in vegetables, but more consideration must be given on (a) if intervention is needed, (b) if the strategies are practical, and (c) how much the risk is lessened by the field intervention. Our interviews illustrated that most stakeholders do not know what the levels of Cd or Pb are in their soils or in products they produce or handle, and there was thus a clear need for faster and cheaper testing of soils and plants where spinach is grown. While the U.S FDA Total Diet Study and similar worldwide market surveys are excellent for assessing potential human exposure to metals, such surveys cannot elucidate the edaphic factors that lead to higher metal levels in plant foods. In the United States, the USDA-Agricultural Research Service (ARS) and land grant institutions, or their equivalents in other production regions, could house regional core facilities for this purpose to enable widespread screening of paired soils and plants for metal content at a fraction of the cost that private labs charge. Such an effort would help to identify areas where mitigation practices might be needed and would enable researchers to develop a database of paired soil and plant samples to better predict metal risk in edible foods. Unfortunately, scarce data exists on this topic in the United States and many of the agronomists working on these topics have or will soon retire, so a pipeline of new researchers to address these questions is needed. There was also a general lack of understanding of links between soils, metals, and plants, so USDA-ARS and land grant institutions could increase extension efforts perhaps through integrated projects on this topic.

To achieve "closer to zero" Cd in plant foods, it is imperative that Cl^{-} inputs are at a minimum and that Zn applications are higher than what a soil test would recommend for optimal Zn nutrition, but not so high that Zn phytotoxicity would occur. Practically speaking, to lessen uptake of Cd, it is probably easier to choose low Cl^{-} inputs from fertilizer or other soil amendments compared to lessening Cl^{-} in irrigation water, especially for growers in California and Arizona where water is scarce and where Cl^{-} is used as a disinfectant for leafy greens. Future work could focus on developing non-chloride techniques that are safe and affordable to keep leafy greens both pathogen and metal "free." All surveyed growers indicated that adding a soil amendment such as Zn sulfate, organic matter, or limestone would have minimal impact on the operation, but it would require some cost and labor that is outside of the normal operation. Moreover, field-scale trials of such interventions are found in reports and are not peer-reviewed. Thus, more applied research on mitigation strategies at the field scale is warranted. Because growers require cost-effective or -neutral solutions, policy efforts should focus on incentivizing this change for growers to keep them competitive in global markets.

Conflict of Interest

The authors declare no conflicts of interest relevant to this study.

Data Availability Statement

This is a review article and all data presented in the figures is openly available in Wolnik et al. (1985) and in the U. S. FDA Total Diet Study at https://www.fda.gov/food/reference-databases-and-monitoring-programs-food/fda-total-diet-study-tds. All data from the interviews is presented within the manuscript without identifying information according to the University of Delaware Internal Review Board protocol (2020651-1).

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