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## ACCUMULATION OF LEAD AND ARSENIC BY CARROTS GROWN ON LEAD-ARSENATE CONTAMINATED ORCHARD SOILS

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□ Concerns have been raised of possible human food chain transfer of lead and arsenic from crops grown on orchard soils with histories of lead arsenate use. The objective of this study was to determine arsenic and lead uptake by three cultivars of carrots grown on four orchard soils with histories of lead arsenate use. Total concentrations of arsenic and lead in these soils ranged from 93 to 291 and from 350 to 961 mg kg<sup>-1</sup> for arsenic and lead, respectively. Arsenic in peeled carrot ranged from 0.38 to 1.64 mg kg<sup>-1</sup>, while lead ranged from 2.67 to 7.3 mg kg<sup>-1</sup> dry weight. This study demonstrated that carrots will accumulate arsenic and lead in the root, which may become a human health risk when consumed. However, further studies are needed to determine what fraction of arsenic and lead in these carrots are bioavailable to humans when consumed.

**Keywords:** orchard, lead-arsenate, carrots, lead, arsenic

### INTRODUCTION

Lead arsenate was used as a foliar spray from the 1900s to the 1960s to control codling moth (*Cydia pomonella*) in apple (*Malus sylvestris* Mill) orchards (Merry et al., 1983; Peryea, 1998a). Lead (Pb) and arsenic (As) are generally immobile in soil and remain in the surface soil due to adsorption by fine silt and clay particles, amorphous oxides, and organic matter (Merwin et al., 1994; Peryea, 1991; Renshaw et al., 2006). Accumulated Pb and As from lead arsenate pesticides are persistent in the environment (Sharma et al., 2007). Concentrations of Pb and As in some orchards exceeded 900 and 200 mg kg<sup>-1</sup> respectively, (Codling and Ritchie, 2005; Peryea, 1998b).

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The present study was prompted by the discovery of elevated Pb levels in commercial carrots sampled by the US Food and Drug Administration in 1998 (Davila, 1999). In their normal market basket food sampling and analysis program, two carrot samples had anomalous levels of Pb. Both samples, one from Washington State and the other from Michigan, had been grown on soils which historically had been used for orchards. Because of food safety concerns about Pb in carrots, the United State Food and Drug Administration (US FDA) asked the United States Department of Agriculture (USDA)'s Agricultural Research Service to investigate the nature of Pb accumulation in carrots grown on former orchard soils with a history of lead arsenate use.

## Lead

Lead is a toxic element that occurs naturally in rocks, soil, plants, water, and the atmosphere. Lead has been used in many products such as an anti-knock compound in gasoline, in pesticides, in paints, and in storage batteries (Peryea, 1998a). It can be harmful to humans when inhaled, ingested directly through the consumption of contaminated water, or ingested indirectly by consuming crops grown on contaminated soils or irrigated with contaminated water (Andra *et al.*, 2006; Fewtrell *et al.*, 2004).

Diseases such as neurobehavioral impairment, hypertension, and cardiovascular disease in humans are attributed to excessive lead exposure, especially in developing children (Fewtrell *et al.*, 2004). However, plants generally do not accumulate substantial amounts of Pb in tops or edible tissues (Chaney and Ryan, 1994). Lead is most available for plant uptake in soils with low organic matter, low pH, and low phosphate (Zandstra and De Kryger, 2007). When phosphate is present in the rhizosphere, an insoluble Pb compound (chloropyromorphite) can be formed near or in the roots (Cotter-Howells *et al.*, 1999). Brown *et al.* (2004) reported a reduction in soluble Pb with increased concentration of phosphate. Nriagu (1978) suggested that Pb in soils with high pH and phosphate will precipitate as lead phosphate, lead carbonate, or lead hydroxides. All of these processes will reduce Pb availability for crop uptake.

Studies have shown that some vegetable crops remove Pb from Pb contaminated soils. Chisholm (1972) reported that Pb concentration in carrots grown on two lead arsenate contaminated soils exceeded the Canadian residue tolerance levels for human safety of 2 mg kg<sup>-1</sup>. Boon and Soltanpour (1992) found that the Pb concentration of leafy vegetables such as spinach ranged from < 5 to 45 mg kg<sup>-1</sup> when grown on soils with a total Pb concentration above 2000 mg kg<sup>-1</sup>. Creger and Peryea (1994) concluded that the increased Pb concentration of leafy vegetable crops grown on a Pb contaminated soil may have resulted from soil particles containing Pb adhering to the plant surface. Carrot root Pb concentrations reported in the literature vary widely. Davies and White (1981), for example, observed that

carrot root Pb concentration ranged from 11–65 mg kg<sup>-1</sup> when plants were grown on lead contaminated soil having a hot nitric acid extractable Pb level of 11,620 mg kg<sup>-1</sup>. Jorhem et al. (2000) reported carrot root Pb concentrations ranging from <0.004 to 1.6 mg kg<sup>-1</sup> (fresh weight) in plants grown on soils with 2 M HNO<sub>3</sub> extractable Pb ranging from 19 to 265 mg kg<sup>-1</sup> and with pH ranging from 4.7–5.0, with the highest tissue Pb concentration occurring at the highest soil Pb concentration and lowest pH.

### Arsenic

Arsenic (As) is found naturally in the environment. High exposure to arsenic has been shown to cause skin, lung and bladder cancer in humans (Jones, 2007; Ng et al., 2003). Humans can be exposed to As directly through the consumption of water and indirectly by consuming crops grown on As contaminated soils or irrigated with contaminated water (Chaturvedi, 2006; Chen et al., 1992; Diaz et al., 2004; Merwin et al., 1994; Smith et al., 2000; Zhao et al., 2004). It has been estimated that 35 to 77 million people in Bangladesh are at risk of As poisoning from drinking As contaminated well water (Smith et al., 2000). Ahmad et al. (2005) reported that water is the major pathway for human As contamination. The recommended guideline set by United States Environmental Protection Agency for As in drinking water is 10 µg L<sup>-1</sup> (United States Environmental Protection Agency, 1998), but it currently has no recommended guidelines for As in food crops.

Inorganic arsenite (As III) and arsenate (As V) are the dominant toxic arsenic species found in food and drinking water (Chaturvedi, 2006; Diaz et al., 2004). Arsenic is not essential for plant growth and inorganic As species are generally regarded as potentially phytotoxic when they accumulate in soils (Chaturvedi, 2006; Sheppard, 1992). Uptake, accumulation, and translocation of As by plants is influenced by factors such as 1) soil properties, 2) soil As concentration, 3) presence of other ions which compete with As for sorption on soil surfaces, and 4) plant species and age (Chaturvedi, 2006; Jiang and Singh, 1994; Matschullat, 2000). Geng et al. (2006) stated that soil-plant transfer of As is one pathway for human exposure to As. It is believed that most food crops are injured by soil As before the crops can accumulate enough As to become a health risk to humans and animals (Elfving et al., 1978). For example, Paivoke (1983) observed yield and chlorophyll reduction in garden pea (*Pisum sativum* L.) grown hydroponically when As concentration was 0.7 mg L<sup>-1</sup>, while Jiang and Singh (1994) observed a significant yield reduction in barley (*Hordeum vulgare* L.) when As was applied to soil at 50 mg kg<sup>-1</sup> either as sodium arsenite or disodium hydrogen arsenate. Arsenic concentration in plants also varies within crop species. Zandstra and De Kryger (2007) reported As concentrations of 0.135 mg kg<sup>-1</sup> in peeled carrot roots grown on orchard soil when total soil As level was 110 mg kg<sup>-1</sup>, while Elfving et al. (1978) reported As concentration in peeled carrot roots

as high as  $0.9 \text{ mg kg}^{-1}$  when plants were grown on a silt loam orchard soil with  $31 \text{ mg kg}^{-1}$  total As.

## Carrots

Carrot is an important fresh market and processing crop that provides yearly income of over \$300 million to US growers. With the conversion of orchard land to vegetable crop production, there is the potential for planting carrots on orchard land with a history of lead arsenate use (Preer et al., 1980; Zandstra and De Kryger, 2007). Therefore, carrot is an economically important crop to study in relation to lead and arsenic accumulation in crops.

There are growing concerns that Pb and As will enter the human food chain when orchards with histories of lead-arsenate use are converted to vegetable crop production (Merwin et al., 1994). Therefore, the objective of our study was to evaluate the effect of residual Pb and As in four lead-arsenate pesticide contaminated orchard soils on yield of and Pb and As uptake by three cultivars of carrots.

## MATERIALS AND METHODS

### Collection and Preparation of Lead Arsenate Contaminated Orchard Soils

Soils were collected from four orchards with histories of lead arsenate use. The soil series were Bagstown loam (*Oxyaquic Hapludults*), Spike sandy loam (*Psammentic Haplaquolls*), Hudson silty clay loam (*Glossaquic Hapludalf*), and Cashmont silt loam (*Aridic Haploxerolls*). A non-orchard soil, Christiana fine sandy loam (*Typic Paleudult*), was collected for use as a control soil. All soils were adjusted to near pH 6.5 with calcium and magnesium carbonate and then fertilized with  $300 \text{ kg ha}^{-1}$  of phosphorus (P) as calcium phosphate [ $\text{Ca}(\text{H}_2\text{PO}_4)_2$ ] and potassium phosphate ( $\text{KH}_2\text{PO}_4$ ),  $100 \text{ kg ha}^{-1}$  nitrogen (N) as ammonium nitrate ( $\text{NH}_4\text{NO}_3$ ),  $60 \text{ kg ha}^{-1}$  magnesium (Mg) as magnesium sulfate ( $\text{MgSO}_4$ ) and magnesium carbonate ( $\text{MgCO}_3$ ) and  $232 \text{ kg ha}^{-1}$  potassium (K) as  $\text{KH}_2\text{PO}_4$ . Soil and fertilizers were mixed and incubated moist (near field capacity) for four weeks.

### Experiment

Four kilograms of each soil (based on air dried weight) were placed in  $20 \times 18 \text{ cm}$  plastic pots and planted with three cultivars of carrots: 'Gold King', 'Monique', and 'Danvers 126'. The cultivars included varieties used for fresh produce or processing. Soil surfaces of each pot were covered with 2 cm of plastic beads to reduce splashing during watering. Each soil x carrot cultivar

was replicated four times. Each pot was placed into a plastic saucer, and pots were placed in a growth chamber in a randomized complete block design with 16 hr light at 25°C and 8 hr dark at 19°C. Plants were watered every two days or as needed to maintain field capacity. Any leachate collected was returned to the pots. Eight days after germination, plants were thinned to 5 plants per pot and grown for 90 days.

Carrot tops and roots were harvested. Tops were washed with sodium lauryl sulfate and rinsed with deionized water. Roots were scrubbed using a vegetable brush, washed in sodium lauryl sulfate, rinsed multiple times with de-ionized water to minimize the presence of soil particles in the carrot peel layer before peeling. Peeled carrot roots, peel, and tops were frozen at -4°C overnight and freeze-dried for 5 days. After yield was determined, plant tissue was ground using an acid washed mortar and pestle, mixed-well, and stored until digestion.

### **Soil and Plant Analysis**

Soil pH was measured in a 1:1 soil to water slurry using a combined electrode; organic carbon was determined with a LECO-CN analyzer (LECO Corp., St Joseph, MI, USA). Soil texture was determined by the pipette method (Gee and Bauder, 1986). Total soil As and Pb were determined by aqua regia digestion (McGrath and Cunliffe, 1985). The Mehlich-3 method (Mehlich, 1984) was used to determine extractable soil metals and nutrients. The extracts were analyzed for calcium (Ca), Mg, K, P, manganese (Mn), copper (Cu), Pb, and zinc (Zn) using a Perkin Elmer inductively coupled plasma-optical emission spectrometer (ICP-OES) (Perkin Elmer, Waltham, MA, USA) with scandium as an internal standard. Soil As was determined by an ICP-OES hydride method outlined by Anderson and Isaacs (1995). The procedure was as follows: 4 ml of soil extract solution was placed into 15 mL test tubes and 1.5 mL concentrated hydrochloric acid (HCl; trace element grade), 2 mL potassium iodide solution, and 2.5 mL 1.73 M sulfamic acid were mixed and allowed to stand for 30 minutes for reaction to occur. A 0.5% sodium borohydride and 0.05% sodium hydroxide solution was used to generate arsine gas as it entered the nebulizer of the ICP-OES.

A microwave digestion method as outlined by Codling and Ritchie (2005) was used for plant sample digestion. Lead and P in the digested solution were determined using ICP-OES with scandium as an internal standard. Plant As concentration was determined using the hydride procedure previously outlined. To ensure accuracy and precision, all samples were analyzed in duplicate with one blank and one lab standard (peeled carrot root) included for every ten samples. Peach leaf Standard Reference Material from National Institute of Standards (NIST-1547, Gaithersburg, MD, USA), was included after every 30 samples.

**TABLE 1** Characteristics of the soils before and after pH adjustment. Total metal concentrations for unadjusted soils are given for Pb and As. After pH adjustment and fertilization, extractable metals and nutrients of the five soils used in the experiment were determined by Mehlich-3 extraction

Parameter	Christiana sandy loam	Bagstown sandy loam	Hudson silt loam	Spike sandy loam	Cashmont silt loam
	Before pH adjustment				
pH	5.32	5.16	4.49	7.08	5.26
EC ms cm <sup>-1</sup>	0.48	0.80	1.05	0.35	0.20
OC mg kg <sup>-1</sup>	18.0	100	43.9	14.4	12.4
As mg kg <sup>-1</sup> †	4.00	133	153	93	291
Pb mg kg <sup>-1</sup> †	20.0	676	435	350	961
	Mehlich-3 extraction after pH adjustment				
pH	6.29	6.69	6.12	6.07	6.32
As mg kg <sup>-1</sup>	0.45	20.29	13.15	37.75	110
Pb mg kg <sup>-1</sup>	<0.01	263	164	252	524
Ca mg kg <sup>-1</sup>	581	2662	2115	1012	1768
Cu mg kg <sup>-1</sup>	0.63	8.63	5.61	4.79	2.50
Fe mg kg <sup>-1</sup>	260	272	359	316	291
K mg kg <sup>-1</sup>	241	628	247	291	522
Mg mg kg <sup>-1</sup>	77	287	288	328	333
Mn mg kg <sup>-1</sup>	9.9	72	35	102	49
P mg kg <sup>-1</sup>	127	414	201	446	383
Zn mg kg <sup>-1</sup>	1.70	10.26	0.85	2.60	5.89

†Aqua Regia digestion, McGrath and Cunliffe (1985).

In this study, soil and cultivar effects were examined using a factorial analysis of variance (ANOVA) conducted in a randomized complete block design. Statistical Analysis System PROC MIXED (SAS Institute, Cary, NC, USA) was used to model the treatment effects. Means comparisons were done with Sidak adjusted *P*-values so that the experiment-wise error was 0.05.

## RESULTS AND DISCUSSION

Selected characteristics of soils used in the experiment are presented in Table 1. The pH of all soils was adjusted to obtain a target pH of about 6.5, which is considered optimum for carrot growth (Sanders, 1998). In the contaminated soils, total As concentration was 93, 133, 153, and 291 mg kg<sup>-1</sup> for the Spike, Bagstown, Hudson, and Cashmont soils, respectively, while total Pb concentration was 350, 435, 676, and 961 mg kg<sup>-1</sup> for the Spike, Hudson, Bagstown, and Cashmont soils, respectively. As expected, Mehlich-3 extractable As concentrations in the contaminated orchard soils were much lower than the total concentrations. Total As concentration in these orchard soils exceeded the 40 mg kg<sup>-1</sup> limit United State Environmental Protection Agency (USEPA) has set for As contaminated soil (USEPA, 1998). Total Pb



**TABLE 2** Analysis of variance for carrot root

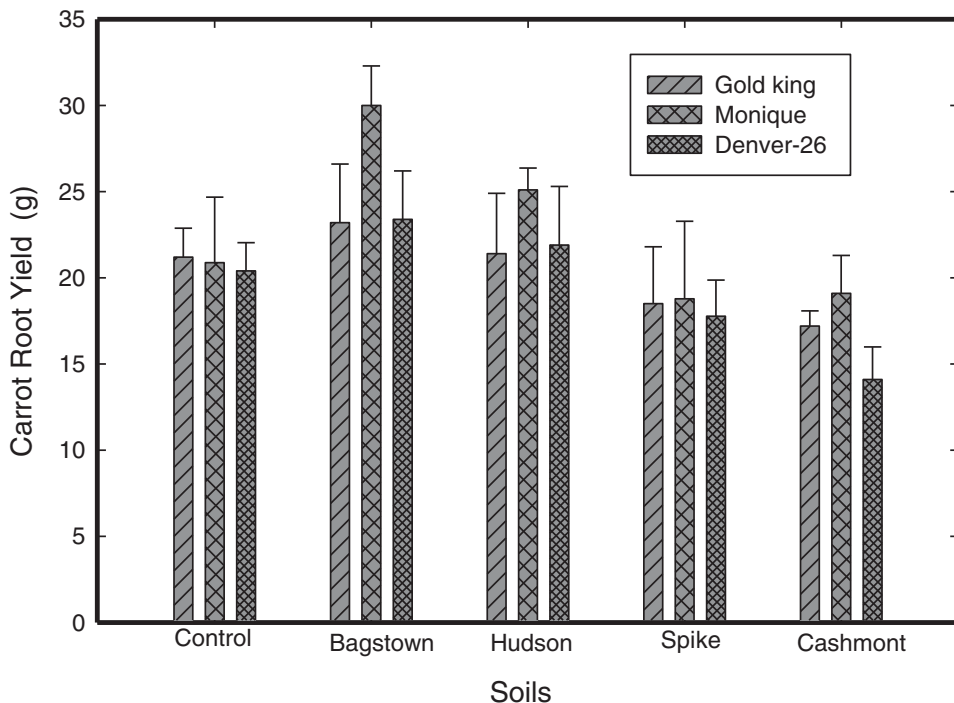
Source	DF	Yield		Pb		As		P	
		F-value	p-value	F-value	p-value	F-value	p-value	F-value	p-value
Cultivar	2	8.83	0.0023	1.16	0.3462	5.95	0.0259	3.82	0.0514
Soil	4	28.6	0.0001	234	0.0001	97.1	0.0001	2.78	0.0698
Cv × soil	8	2.05	0.0999	3.16	0.0247	9.05	0.0002	1.39	0.2751

Cv = cultivar.

concentration in three of the four orchard soils exceeded the 400 mg kg<sup>-1</sup> level considered hazardous to exposed organisms (Dudka and Miller, 1999).

### Carrot Yield

Carrot yield was significantly affected at the  $P < 0.05$  level by the main factors soil and cultivar but not by their interaction (Table 2). Carrots grown on the Bagstown and Hudson soils had the highest dry matter yield, while carrots grown on the Spike and Cashmont soils yielded less than the control soil (Figure 1). The higher dry matter yield observed on the Bagstown and

**FIGURE 1** Carrot root yield as affected by lead-arsenate contaminated orchard soils. Means plus standard deviation  $n = 4$ .

Hudson soils compared to the Spike and Cashmont soils may have resulted from 1) the lower levels of Mehlich-3 extractable soil As, and 2) the higher levels of organic matter, which sequesters Pb (Brown *et al.*, 2004), in the Bagstown and Hudson soils (Table 1). Zandstra and De Kryger (2007) reported that lead is most soluble in soils with low organic matter. High levels of extractable Pb and As have been shown to reduce crop yields (Codling and Ritchie, 2005; Paivoke, 1983). Comparing cultivars, yield was highest for 'Monique' on all orchard soils but the effect was significant only on the Bagstown soil (Figure 1). The 'Monique' cultivar had larger roots than the other two cultivars, which contributed to its higher yield.

### Elemental Composition of Peeled Carrot Root

Peeled carrot root lead (Pb) concentration was highly significantly affected by soil, slightly affected by the soil  $\times$  cultivar interaction, and not affected by cultivar (Table 2). Peeled carrot root Pb concentration within carrot cultivar was significantly higher in carrots grown on the lead arsenate contaminated orchard soils than in those grown on the control soil (Table 3). Carrots grown on the Cashmont soil had the highest Pb concentration. The lower Pb concentrations in carrots grown on the Bagstown and Spike soils may have been the result of higher phosphorus in these soils (Table 1). Phosphorus is known to precipitate Pb when applied to lead-arsenate contaminated orchard soils, making it unavailable for plant uptake (Peryea, 1991). Brown *et al.* (2004) and Kalbasi *et al.* (1995) also reported a reduction in soil soluble lead with addition of phosphate. Although Pb concentration in peeled carrot roots grown on orchard soils is elevated, it is not clear that this Pb comprises a risk to consumers. Many human feeding studies with Pb isotopes have shown that Pb in food is absorbed to a much lower extent (1–5%) compared to Pb in water (50–80%) (James *et al.*, 1985). The presence of phytate, fiber, and calcium in foods tends to inhibit Pb absorption, and the presence of food in the stomach raises the pH, which reduces the ability of stomach fluids to dissolve Pb from foods or ingested soil. Only one study has been conducted using Pb incorporated into a food (spinach) fed to human subjects, and the bioavailability of that Pb was very low (Heard *et al.*, 1983).

The concentration of arsenic (As) in peeled carrot roots was highly significantly affected by soil and the soil  $\times$  cultivar interaction but only slightly by cultivar (Table 2). Peeled carrots roots grown on the lead arsenate contaminated soils had a significantly higher As concentration than did those grown on the control soil (Table 3). Arsenic concentration in peeled carrot roots was significantly higher on the Spike and Cashmont soils compared to the Bagstown and Hudson soils. Mean As concentration was 0.30, 0.40, 1.72, and 1.61 mg kg<sup>-1</sup> for the Bagstown, Hudson, Spike, and Cashmont soils, respectively. The higher As concentration in carrots grown on the Spike

**TABLE 3** Lead, arsenic and phosphorus concentrations in carrot root grown on four lead arsenate contaminated orchard soils

	Pb				As				P			
	Gold King	Monique	Denver 126	Soil mean	Gold King	Monique	Denver 126	Soil mean	Gold King	Monique	Denver 126	Soil mean
Control	0.19d†x‡	0.28cx	0.12cx	0.20d§	0.05cx	0.04dx	0.06cx	0.05c	2798	2308	2319	2475
Bagstown	2.53cx	2.55bx	2.67bx	2.58c	0.47bx	0.39cx	0.05cy	0.30b	2319	2253	2272	2307
Hudson	3.74x	2.72by	3.55xy	3.34b	0.44bx	0.34cx	0.41bx	0.40b	2350	2225	2075	2217
Spike	2.47cx	2.40bx	2.84bx	2.57c	1.17ay	2.48ax	1.15ay	1.72a	2312	2253	2272	2279
Cashmont	7.23ax	7.11ax	7.12ax	7.15a	1.68abx	1.32bx	1.87ax	1.61a	2313	2182	2202	2233
Cv mean	3.23	3.01	3.26		0.76ab	0.99a	0.69b		2418	2244	2244	

†Soil means within Cv with different letters (a, b, c, d) are different at the 0.05 significant level.

‡Cv means within soil with different letters (x, y, z) are different at the 0.05 significant levels.

§Soil means with different letters are different at the 0.05 significant levels.

**TABLE 4** Analysis of variance for carrot peel

Source	DF	Pb		As		P	
		F-value	p-value	F-value	p-value	F-value	p-value
Cultivar	2	3.23	0.066	0.15	0.867	41.1	0.0001
Soil	4	91.3	0.0001	34.6	0.0001	17.2	0.0001
Cv × soil	8	2.38	0.069	0.12	0.997	4.5	0.0078

Cv = cultivar.

and Cashmont soils may have been due to higher Mehlich-3 extractable As and P in these soils (Table 1). It has been shown that P application to lead arsenate contaminated orchard soil increases arsenate solubility making it more available for crop uptake (Peryea, 1991; Codling, 2007). Carrot As concentrations in our study were similar to those found by Elfving *et al.* (1978) and Zandstra and De Kryger (2007) but higher than those found by Diaz *et al.* (2004).

Phosphorus (P) concentration in peeled carrot roots was not significantly affected by soil, cultivar, or the soil × cultivar interaction (Table 2). In all cases, P concentration in peeled carrot roots was within levels considered sufficient for carrots (Jones *et al.*, 1991; Reuter and Robinson, 1986). There was no relationship between tissue P concentration and tissue Pb or As concentration.

### Carrot Peel

Carrot peel Pb concentration was highly significantly affected by soil but not by cultivar or the soil × cultivar interaction (Table 4). Pb concentration in carrot peel was significantly higher in carrots grown on the lead arsenate contaminated orchard soils than in those grown on the control soil (Table 5). Carrots grown on the Cashmont soil had the highest Pb concentration in the peel followed by those grown on the Spike soil. In all cases, tissue Pb was higher for peeled carrot roots than for peel (Tables 3 and 5). This result was unexpected because we initially assumed that soil contamination of the peel layer was responsible for Pb in root vegetables such as carrots. The high levels of Pb within peeled carrot roots might be explained by the presence of xylem vessels that adsorb Pb on their surfaces. Additionally, the formation of lead phosphates within the xylem may trap Pb in storage roots. Further studies are needed to verify the form and location of Pb in carrot tissue.

Carrot peel As concentration was highly significantly affected by soil but not by cultivar or the soil × cultivar interaction (Table 4). Averaged over cultivars, As concentration in carrot peel was significantly higher in carrots grown on the orchard soils compared to the result for the control soil (Table 5). The highest As levels were found in carrot peel grown on

TABLE 5 Lead, arsenic and phosphorus concentrations in carrot peel grown on four lead arsenate contaminated orchard soils

	Pb				As				P											
	Denver 126		Soil mean		Gold King		Monique		Denver 126		Soil mean		Gold King		Monique		Denver 126		Soil mean	
	Gold King	Monique	Denver 126	Soil mean	Gold King	Monique	Denver 126	Soil mean	Gold King	Monique	Denver 126	Soil mean	Gold King	Monique	Denver 126	Soil mean	Gold King	Monique	Denver 126	Soil mean
Control	0.19	0.13	0.14	0.15c§	0.07	0.07	0.07	0.07c	5981atx†	4929axy	4087ay	4999a	5981atx†	4929axy	4087ay	4999a	5981atx†	4929axy	4087ay	4999a
Bagstown	0.74	0.88	0.70	0.77b	0.93	0.92	0.80	0.88b	3990bx	3957axy	3067by	3671b	3990bx	3957axy	3067by	3671b	3990bx	3957axy	3067by	3671b
Hudson	1.27	0.95	0.73	0.98b	0.85	0.74	0.58	0.72b	3787bcx	3474axy	2882by	3382b	3787bcx	3474axy	2882by	3382b	3787bcx	3474axy	2882by	3382b
Spike	0.91	1.32	1.05	1.09ab	2.09	3.62	2.40	2.71abc	3418cy	3756ax	3273aby	3482b	3418cy	3756ax	3273aby	3482b	3418cy	3756ax	3273aby	3482b
Cashmont	1.87	1.44	1.29	1.53a	5.48	5.14	5.34	5.32a	3548bcx	3814ax	3155aby	3506b	3548bcx	3814ax	3155aby	3506b	3548bcx	3814ax	3155aby	3506b
Cv mean	0.99	0.94	0.78		1.89	2.10	1.84		4145a	3986a	3293b		4145a	3986a	3293b		4145a	3986a	3293b	

†Soil means within Cv with different letters (a, b, c, d) are different at the 0.05 significant level.

‡Cv means within soil with different letters (x, y, z) are different at the 0.05 significant levels.

§Soil means with different letters are different at the 0.05 significant levels.

**TABLE 6** Analysis of variance for carrot tops

Source	DF	Pb		As		P	
		F-value	p-value	F-value	p-value	F-value	p-value
Cultivar	2	5.95	0.0259	8.83	0.0023	1.16	0.3462
Soil	4	97.1	0.0001	28.6	0.0001	234	0.0001
Cv × soil	8	9.05	0.0002	2.05	0.0999	3.16	0.0247

Cv = cultivar.

the Cashmont and Spike soils. The lower As concentration observed in peel of carrots grown on the Hudson soil reflects the lower level of Mehlich-3 extractable As in this soil (Table 1). Unlike in the case of Pb, As concentration was higher in carrot peel than in peeled root, with values for carrot peel of 0.88, 0.72, 2.71, and 5.32 mg kg<sup>-1</sup> for the Bagstown, Hudson, Spike, and Cashmont soils, respectively, compared to values of 0.30, 0.40, 1.72, and 1.61 mg kg<sup>-1</sup> for peeled carrot roots grown on the same soils, respectively (Tables 3 and 5). The higher levels of As in the peel compared to the peeled root may have resulted from direct contact between the root and the lead arsenate contaminated soils, even though the carrots were washed thoroughly before peeling. Alternatively, the cells of the peel layer may accumulate As to higher levels than do other carrot cells. Our results are consistent with those of Helgesen and Larsen (1998), who found that the As concentration of carrot peel was two to seven times higher than that of peeled carrot roots. Nevertheless, peeling carrot roots before eating should not have a substantial effect on the intake of total As due to the low weight of the carrot peel with respect to the total weight of the carrot (Munoz et al., 2002).

Carrot peel P concentration was significantly affected by soil, cultivar, and the soil × cultivar interaction (Table 4). Carrot peel P concentration was higher on the control soil compared to the lead arsenate contaminated soils (Table 5). Averaged over soils, carrot peel P was significantly lower in Danvers-126 compared to the 'Gold King' and 'Monique' cultivars.

### Carrot Tops

The concentration of Pb in carrot tops was highly significantly affected by soil, cultivar, and the soil × cultivar interaction (Table 6). Pb concentration was significantly higher in carrot tops grown on the orchard soils compared to the control soil (Table 7). As was the case for peeled roots and peel, Pb concentration in carrot tops was highest on the Cashmont soil, which had the highest levels of total and extractable soil Pb (Table 1). The extremely high Pb concentration in tops of the 'Monique' cultivar grown on the Cashmont soil, compared to the other cultivars, resulted in very high variation within

**TABLE 7** Lead, arsenic and phosphorus concentrations in carrot tops grown on four lead arsenate contaminated orchard soils

	Pb				As				P			
	Gold King	Monique	Denver 126	Soil mean	Gold King	Monique	Denver 126	Soil mean	Gold King	Monique	Denver 126	Soil mean
Control	0.18c x ‡	0.17cx	0.15cx	0.17b§	0.09cx	0.07cx	0.11bx	0.09c	4826ax	4387ax	3129ay	4114a
Bagstown	2.23by	3.59ax	2.54abxy	2.79a	1.42bx	1.35bx	2.71bx	1.83b	3582bx	3698ax	2761abx	3347bc
Hudson	2.77abx	2.69ax	3.13ax	2.86a	0.75bx	0.50bcx	1.29bx	0.84b	2688bx	2191bxy	2069by	2316d
Spike	2.35bx	2.80ax	1.8bx	2.32a	5.46ay	5.19abexy	8.75ax	6.47abc	3429by	4812ax	4032axy	4091ab
Cashmont	4.74ax	9.15abx	2.88abx	5.59ab	5.99ax	4.61ax	8.18ax	6.26a	2838bx	3686ax	2641abx	3055c
Cv Mean	2.45	3.68	2.01		2.74b	2.34ab	4.21a		3473a	3755a	2926b	

‡Soil means within Cv with different letters (a, b, c, d) are different at the 0.05 significant level.

‡Cv means within soil with different letters (x, y, z) are different at the 0.05 significant levels.

§Soil means with different letters are different at the 0.05 significant levels.

the Cashmont soil treatment, and consequently there was no significant difference between the Cashmont and control soil means (Table 7). Lead concentration of carrot tops was similar for the three cultivars when averaged over soils including the control. In the present study, Pb concentration in the parts of the carrot plant was in the order peeled carrot root > tops > peel. These findings disagree with those of Spittler and Feder (1979), who reported higher Pb concentrations in vegetable tops than in roots.

Arsenic concentration in carrot tops was highly significantly affected by soil and cultivar but not by the soil x cultivar interaction (Table 6). As concentration was higher in carrot tops of plants grown on the orchard soils, compared to results for the control soil (Table 7). Comparing results for the four orchard soils, As concentration of carrot tops was significantly higher on the Spike and Cashmont soils, which had higher extractable soil As than the other two orchard soils. Averaged over soils, As concentration in the parts of the carrot plant was in the order tops > peel > peeled carrot root. Similar findings were observed by Elfving *et al.* (1978) for carrots grown on a silt loam orchard soil with 31 mg kg<sup>-1</sup> total As.

Carrot top P concentration was significantly affected by soil and the soil x cultivar interaction (Table 6). Carrot top P was highest in plants grown on the control and Spike soils (Table 7). Averaged over soils, P concentration in carrot tops was significantly higher for the 'Gold King' and 'Monique' cultivars than for Danvers-126. In all cases, P concentration in carrot tops was at a level that is generally considered sufficient for crop tissue (Jones *et al.*, 1991).

## CONCLUSIONS

This study demonstrated that carrots grown on lead arsenate contaminated soils accumulated Pb and As within the edible root, which potentially could contribute to dietary Pb and As intake by consumers. The concentration of Pb in the peeled carrot root was higher than in the peel, confirming that Pb uptake and not just surface contamination contribute to the Pb that has been found in commercial carrot samples. The Pb concentration in carrot tops was also higher than in the peel, though not as high as in the peeled root, demonstrating that some Pb is translocated from the root to the tops. Because of the low bioavailability of Pb in food, it is not clear that the Pb levels found in this study comprise a risk to consumers. In contrast to the results for Pb, As accumulated more in the peel and tops than in the peeled root. Further studies are needed to determine what fraction of Pb and As in these carrots would be bioavailable if consumed by humans.

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