

Phosphate Fertilizer Enhances Arsenic Uptake by Apricot Liners Grown in Lead-arsenate-enriched Soil

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Abstract. Phosphate fertilizer additions to soils containing lead arsenate (LA) pesticide residues can increase As volatility. Apricot (*Prunus armeniaca* L.) rootstock liners were grown in nondraining pots containing Burch loam soil that received a factorial treatment combination: 1) LA enrichment [no added LA (-LA), and LA added at 138 mg Pb/kg and 50 mg As/kg (+LA)]; 2) fertilizer type [monoammonium phosphate (MAP) and its sulfur analog ammonium hydrogen sulfate (AHS)]; and 3) fertilizer anion rate (0-26.1 mol/m³ soil). Measured response variables were soil salinity and pH, plant biomass, and plant As and Pb concentrations. Both MAP and AHS increased soil electrical conductivity (EC) and decreased soil pH, with AHS usually being more salinizing and acidifying than MAP was at equivalent rates. Adding LA reduced shoot and root mass and increased As and Pb concentration in shoots and roots. Shoot and root mass were inversely related to soil EC in the -LA soil but not in the +LA soil. Adding MAP increased shoot and root As concentration in the +LA soil, but adding AHS had no effect. Fertilizer type and rate did not influence shoot As concentration or root Pb concentration in the -LA soil or shoot Pb concentration in either the +LA or -LA soil. Adding AHS to the +LA soil increased root Pb concentration. These results are consistent with a P-enhanced solid-phase As release mechanism, which consequently increases plant uptake of soil As. Phosphate amendment had no effect on soil Pb phytoavailability.

Acid lead arsenate (LA, PbHAsO₄) was the preferred pesticide for insect control in deciduous fruit trees before dichlorodiphenyltrichloroethane's (DDT) introduction in 1947. Frequent high-rate applications led to substantial Pb and As accumulation in orchard topsoils, particularly in apple orchards (Peryea and Creger, 1994). In these soils, plant growth and performance are reduced compared to growth and performance in uncontaminated soil, principally because of As toxicity rather than to excessive Pb (Vandecaveye et al., 1938). Plant As content is positively related to soil As in LA-enriched soils (Creger and Peryea, 1992; Jones and Hatch, 1937).

Using starter fertilizers containing P, particularly monoammonium phosphate (MAP, NH₄H₂PO₄), has become increasingly popular in newly planted deciduous tree fruit orchards (Neilsen et al., 1990). Phosphorus fertilizers added to LA-enriched soils can increase soil As volatility (Peryea, 1991) and mobility (Dav-

enport and Peryea, 1991; Peryea, 1990b). Soil As released by P amendments tends to remain in a readily soluble form (Peryea, 1991) and, therefore, may be more phytoavailable. Woolson (1972) found that dilute HCl+NH₄F-extractable soil As, which he considered a measure of the phytoavailable portion of total soil As, to be substantially increased after adding MAP.

Plant responses to As and P interactions in the root zone are system-dependent. In nutrient solution, increasing dissolved P at a fixed concentration of dissolved As reduced plant uptake of As and sometimes reduced As phytotoxicity (Hurd-Karrer, 1939; Rumberg et al., 1960). Most P and inorganic-As interaction studies in soil-plant systems used a procedure in which P and As were added simultaneously to the soil at the time of planting (Carrow et al., 1975; Hurd-Karrer, 1939; Jacobs and Keeney, 1970; Woolson, 1972; Woolson et al., 1973). These studies' treatment effects were highly variable; the added P had deleterious, negligible, or beneficial effects on plant growth and similarly variable effects on As content. We are aware of only four reports in which plant responses were examined following P applications to soils that were already enriched with As. Calcium phosphate fertilizers decreased cowpea (*Vigna unguiculata* L.) yield in calcium arsenate-enriched soil (Cooper et al., 1931). Although it usually had no effect, in some cases it increased vegetative yields of barley (*Hordeum vulgare* L.), sudangrass (*Sorghum vulgare sudanense* L.), or alfalfa (*Medicago sativa* L.) grown in LA-enriched soils (Benson, 1948, 1953; Overley,

1950). None of these studies presented data concerning P effects on plant As content.

Adding P to Pb-enriched soils has reduced plant Pb concentrations (Bassuk, 1986; Rolfe, 1973; Zimdahl and Foster, 1976), presumably because of decreased Pb volatility resulting from *de novo* precipitation of sparingly soluble Pb-phosphate minerals.

The objective of our study was to determine if adding MAP starter fertilizer to LA-enriched soil could influence growth of and As and Pb concentrations in newly planted apricot trees.

Materials and Methods

A bulk sample of a Burch loam soil (coarse-loamy, mixed, mesic Aridic Haploxeroll) was collected in Apr. 1989 from a former apple orchard block that had been vacant since 1985. The orchard had been planted on virgin sagebrush steppe habitat between 1948 and 1950 and had never received LA pesticide sprays. Plant nutrient status of the soil was adequate: pH (1:1, distilled water) 6.2; 1.0% organic matter; 1 M NH₄OAc (pH 7)-exchangeable cations (in cmol·kg⁻¹): Ca 12.3, Mg 1.3, K 1.0, Na 0.0; hot water-extractable B, 0.4 mg·kg⁻¹; O, 15% CaCl₂-extractable SO₄-S, 6.5 mg·kg⁻¹; and DTPA-TEA-CaCl₂-extractable Zn and Cu, 1.6 and 1.3 mg·kg⁻¹, respectively (standard methods are described in Page et al., 1982). The soil contained 21 mg 0.5 M NaHCO₃-extractable P/kg, an amount considered high enough to preclude P fertilizer use on nontree crops (Dow et al., 1976). Soil-P test standards for fruit trees in Washington have not been established because tree responses are poorly related to soil-P tests (Tukey et al., 1984). Arsenic and Pb concentration in the untreated soil were low; concentrated HCl-extractable soil Pb was <5 mg·kg⁻¹ and soil As was 2.2 mg·kg⁻¹.

The treatments were arranged in factorial combination: two LA rates × two fertilizer sources × four fertilizer rates. Ten-kilogram soil subsamples were placed in 15-liter plastic pots (26 cm diameter, 30 cm tall) lined with nondraining polyethylene bags. We believed that we could minimize the potential risk of flooding closed pots by carefully managing water better than we could by imposing identical leaching regimes in open pots. Furthermore, the effects of flooding, if it were to occur, would be randomly distributed across treatments. Agricultural-grade acid LA [56.7% Pb and 20.5% As (w/w)] was uniformly mixed with the soil in half of the containers at a rate equivalent to 138 mg total Pb/kg and 50 mg total As/kg, resulting in post-equilibration concentrated HCl-extraction values of 110 mg·kg⁻¹ for Pb and 36 mg·kg⁻¹ for As. Reduced growth of newly planted apricots is expected when concentrated HCl-extractable soil As exceeds ≈ 25 mg·kg⁻¹, with As phytotoxicity symptoms appearing if the weather is hot during the growing season (Dow et al., 1983). The lead arsenate-free (-LA) and lead arsenate-amended (+LA) soils were irrigated to saturation with distilled water and allowed to dry and equilibrate open to the atmosphere

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for 3 weeks. After equilibration, soil bulk density averaged 1.1 Mg·m⁻³.

The specificity of the P interaction was evaluated by comparing the effects of MAP with its S analog ammonium hydrogen sulfate (AHS, NH₄HSO₄). On 12 May 1989, reagent-grade MAP or AHS was uniformly mixed with the soils at rates of 0, 8.7, 17.4, and 26.1 mol P or S/m³ soil. Fertilizer treatments were made on an amount-per-volume basis to reflect commercial practice and are expressed on a molal basis to permit direct comparison of anion rates. The experimental fertilizer rates bracketed the current commercial in-hole MAP application rate, which varies from 13 to 22 mol/m³ soil (1.5 to 2.5 g MAP/liter). Apricot was chosen as the test plant because it is sensitive to soil As and shows distinctive visual symptoms of As excess (formation of small circular reddish lesions on the leaf blades—particularly along the vascular network—followed by leaf chlorosis, necrosis of the lesions, and defoliation) (Benson et al., 1972). Apricot seedling rootstock liners were pruned to five vegetative buds per stem and 15 cm of rooted stem below the soil surface. Average dry mass at planting was 5.4 g for shoots and 12.5 g for roots. Immediately after the fertilizer amendment was added to the pot, one liner was planted.

The pots were arranged in a completely randomized design in a greenhouse. All treatments were replicated four times. All liners showed budbreak ≤ 10 days after planting. Then the pots were moved outdoors and re-randomized under a frame structure covered with shade cloth providing 47% attenuation of natural light. Soil moisture was monitored using a portable commercial moisture meter (Aquater Instruments, Fremont, Calif.). Distilled water was added periodically to maintain a soil moisture potential of -1.5 MPa in the lower third of the pots.

The plants had set terminal buds by early July. On 11 July 1989, 59 days after planting, the shoots were cut off at the soil line. The soil was removed from the containers and gently broken apart by hand. Roots—the below-ground portion of the stem+ true root—were manually separated from the soil, washed for 1 min in distilled water to remove adhering soil particles, rinsed in 3 M HNO₃ for 1 min to remove adsorbed Pb and As, and again rinsed in distilled water for 1 min (Zimdahl and Foster, 1976). The shoots and roots were oven-dried at 40C for 72 h, weighed, and ground in a Wiley mill (Arthur H. Thomas Co., Philadelphia). Subsamples of each tissue were wet-digested using sulphomonoperacid [1.6 concentrated H₂SO₄; 1 30% H₂O₂(v/v)]. *While this reagent is an effective oxidant, it is extremely corrosive and potentially explosive. Its use requires special safety precautions.* Total As in the digest was measured by the arsenic-silver diethyldithiocarbamate procedure (Page et al., 1982), replacing the gas dispersion tube with a Pasteur pipette. We found this modification to enhance As recovery and analytical precision. Subsamples of shoot and root tissue were dry-ashed at 525C, and the residue was dissolved in 5 M HCl. Total Pb in the digest was

measured by flame atomic absorption spectrophotometry. Plant Pb and As concentrations are reported on a dry-weight basis.

After harvest, the soil in each pot was mixed and subsamples were collected for analysis of extractable Pb and As, pH, and salinity. Soil Pb and As were measured in the filtered extract of a 1 soil : 10 concentrated HCl suspension shaken for 60 min; this procedure was a modified version of those described in Benson (1976) and Veneman et al. (1982). This procedure is used in Washington to estimate potential for soil As phytotoxicity in fruit trees (Dow et al., 1983). Total dissolved Pb and As in the extracts were analyzed using the same method described for the plant tissues. Soil pH was measured in a 1 soil : 1 distilled water suspension (Page et al., 1982). Soil salinity was estimated by the electrical conductivity (EC) of the supernatant of a 1 soil : 3 distilled water suspension shaken for 4 h, measured at 1 kHz, and corrected to 298 K (U.S. Salinity Laboratory, 1954).

Treatment effects were analyzed using three-way analysis of variance (ANOVA) for a completely randomized design (Table 1). Significant interaction terms (P ≤ 0.05) were examined for simple treatment effects using two-way ANOVA (Steel and Torrie, 1980). Regression analysis was used to relate selected variables.

Results and Discussion

Although shoot As concentrations were high in some treatments, visual symptoms of As toxicity, other than reduced shoot growth, did not appear on any test plants. Symptom expression in apricot seems to vary with cultivar (Creger and Peryea, 1992); the parentage of the test plants was unknown and may have included nonexpressive cultivars. Symptoms might have appeared eventually if the trees had been grown longer.

All treatment main effects and interactions, except LA × type interaction, influenced soil EC (Table 1). Soil EC increased with increasing fertilizer rates; AHS was more salinizing than MAP (Fig. 1a), reflecting the differential solubility of sulfate and phosphate salts (Lindsay, 1979). It is expected that vegetative growth would be reduced when apricots are grown in soils with saturation extract EC values exceeding 1.6 dS·m⁻¹ (Maas, 1986). EC values in Fig. 1a would have been higher if they had been measured using the saturation extraction procedure, and they also probably

underestimate the time-averaged root-zone salinity because of precipitation of the added fertilizer salts (Peryea, 1990a).

Soil pH was influenced by all main treatments, with a significant type × rate interaction (Table 1). Both MAP and AHS reduced soil pH, with AHS being more acidifying at equivalent anion rates (Fig. 1b). This effect could be caused by several factors: higher solution concentrations and greater dissociation of bisulfate ions in the AHS-amended soil (Adams, 1984) or inhibition of vitrification by P-released As in the MAP-treated soil (Bollen et al., 1977). Lead arsenate amendment in the absence of added fertilizer had no effect on soil EC or pH (Fig. 1 a and b). The +LA soil was more saline and more acidic than the -LA soil at the intermediate fertilizer rates, but it had no effect or an opposite effect at 26.1 mol·m⁻³.

Shoot mass was influenced by all treatment main effects, with significant LA × type and LA × rate interactions (Table 1). Adding LA reduced shoot mass at 0, 8.7, and 17.4 mol·m⁻³ (P ≤ 0.001) fertilizer rates, but not at 26.1 mol·m⁻³ (Fig. 2a). We attribute the failure of MAP to stimulate growth to the time period used for the experiment, not to sufficiency of available P in the unamended soil. In a separate study, MAP starter fertilizer initially retarded apple (*Malus domestica* Borkh) tree growth relative to unfertilized trees; however, as the growing season progressed, the growth of the MAP-amended trees caught up with and exceeded that of the unfertilized trees (F.J.P., unpublished data). This response was probably caused by reduced osmotic stress as dissolved phosphate salts precipitated and ensuing enhanced availability of added N and P. Under the hot summer conditions of central Washington, it is common for fruit trees to set terminal buds in midsummer, and for these buds to break and produce new growth after the hottest part of the summer. We speculate that a positive growth response to MAP would have been observed in the current experiment if it had been continued for a full growing season. In the -LA soil, shoot mass was greater in MAP-amended soil than in the AHS-amended soil (P ≤ 0.001). Shoot mass was inversely related to soil EC in the -LA soil (y = 22.218 - 10.875x, r² = 0.58***), suggesting that soil salinity may have been an important growth-limiting factor at the higher MAP and AHS rates. This relationship would likely have been improved if soil salinity had been integrated over time, rather than assessed at the

Table 1. Analysis of variance significance levels for treatment effects on soil electric conductivity (EC) and pH, on shoot and root mass, and on As and Pb concentrations.

Source	Soil		Shoot			Root		
	EC	pH	Mass	As	Pb	Mass	As	Pb
Lead arsenate (LA)	***	*	***	***	**	***	***	***
Fertilizer type (type)	***	***	**	***	NS	NS	***	**
LA × type	NS	NS	**	***	NS	NS	***	***
Fertilizer rate (rate)	***	***	***	NS	NS	***	NS	NS
LA × rate	***	NS	***	NS	NS	*	*	NS
Type × rate	***	***	NS	NS	NS	NS	*	NS
LA × type × rate	*	NS	NS	NS	NS	NS	*	*

NS, *, **, *** Nonsignificant or significant at P ≤ 0.05, 0.01, or 0.001, respectively.

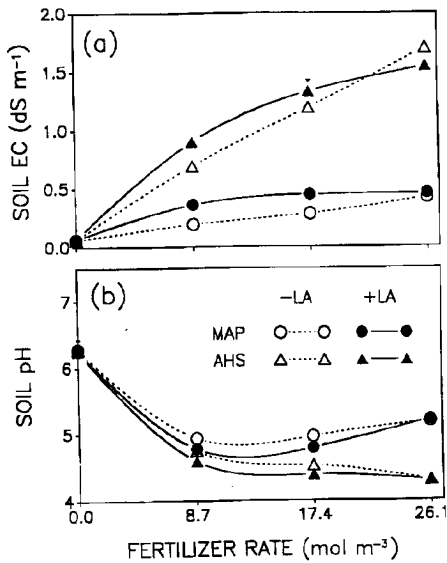


Fig. 1. Influence of lead arsenate amendment (-LA, +LA), fertilizer type [monoammonium phosphate (MAP); ammonium hydrogen sulfate (AHS)], and fertilizer rate on (a) soil electrical conductivity (EC) and (b) soil pH. Standard errors for individual mean values ($n = 4$) are indicated by error bars or are hidden by the symbols.

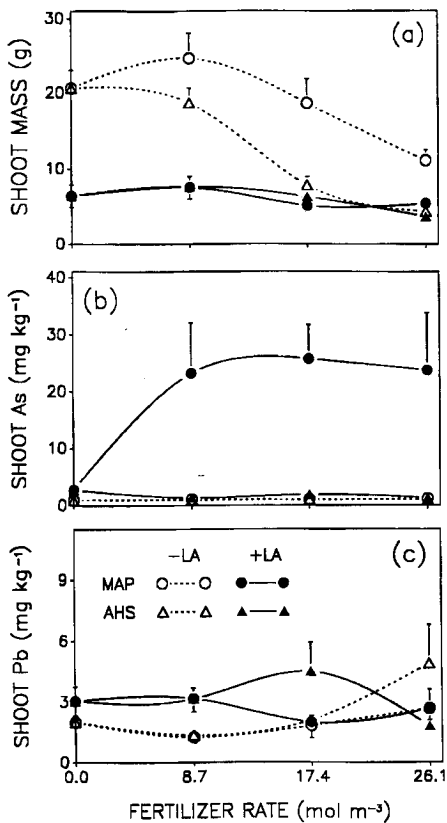


Fig. 2. Influence of lead arsenate amendment (-LA, +LA), fertilizer type [monoammonium phosphate (MAP); ammonium hydrogen sulfate (AHS)], and fertilizer rate on (a) apricot shoot mass, (b) As concentration, and (c) Pb concentration. Standard errors for individual mean values ($n = 4$) are indicated by error bars or are hidden by the symbols.

end of the experiment (Peryea, 1990a). Shoot mass in the +LA soil was not influenced by fertilizer type and rate and was independent of soil EC ($r^2 = 0.05^{NS}$), suggesting that toxic element stress had greater influence than osmotic stress.

Shoot As concentration was influenced by LA and type, with a significant LA \times type interaction (Table 1). In the +LA soil, shoot As concentration was higher in MAP-amended soil than in the AHS-amended soil ($P \leq 0.001$), and it was unaffected by fertilizer type in the -LA soil (Fig. 2b). It did not differ between all nonzero MAP rates in the +LA soil. Shoot As concentration was not biased by biomass dilution in the +LA soil because shoot mass did not differ among the MAP and AHS treatments. Shoot As concentration in the MAP-amended +LA treatments was high compared to the highest values ($<3 \text{ mg}\cdot\text{kg}^{-1}$) reported for leaves of mature apples, peaches (*Prunus persica* Batsch), and apricots grown in LA-enriched soils (Creger and Peryea, 1992; Woolson, 1983) and for first-season leaves ($7\text{--}17 \text{ mg}\cdot\text{kg}^{-1}$) of apple trees planted in LA-enriched soil amended with MAP starter fertilizer (F. J. P., unpublished data). The As concentration in the apricot shoots, which included woody and leafy tissues, may underestimate the concentration in the apricot leaves because leaf As concentration typically exceeds that of woody tissue (Woolson, 1983). Phosphate-released soil As seems to be accumulated quickly by newly planted trees, but it is eventually diluted as tree biomass increases.

Only the LA main effect influenced shoot Pb concentration (Table 1); they averaged $2.1 \text{ mg}\cdot\text{kg}^{-1}$ in the -LA soil and $3.1 \text{ mg}\cdot\text{kg}^{-1}$ in the +LA soil. These concentrations were lower than the averages reported for apple tree leaves ($6.9 \text{ mg}\cdot\text{kg}^{-1}$) grown in LA-enriched soil (Aten et al., 1980) and for leaves and shoots of forest tree species ($>5 \text{ mg}\cdot\text{kg}^{-1}$) (Rolfe, 1973). The $1\text{-mg}\cdot\text{kg}^{-1}$ increase in shoot Pb concentration in the +LA soil likely was insufficient to cause the reduced shoot growth. Shoot Pb concentration was independent of fertilizer rate in the MAP-amended +LA soil ($r^2 = 0.04^{NS}$), suggesting that soil Pb volatility was not reduced (Fig. 2c).

Root mass was influenced by LA and rate, with a significant LA \times rate interaction (Table 1). The added LA reduced root mass at fertilizer rates of 0 ($P \leq 0.001$) and $17.4 \text{ mol}\cdot\text{m}^{-3}$ ($P \leq 0.05$) but not at rates of 8.7 and $26.1 \text{ mol}\cdot\text{m}^{-3}$ (Fig. 3a). Like shoot mass, root mass was inversely related to soil EC in the -LA soil ($y = 16.271 - 3.104x$; $r^2 = 0.25^{**}$) but not in the +LA soil ($r^2 = 0.01^{NS}$). These results indicate that root growth was influenced by salinity and LA stress, with each factor's relative contribution depending on treatment. Root mass seemed to be a poor indicator of root physiological activity because substantially more shoot growth occurred in the -LA soil without commensurately increased root growth.

If soil or LA particles adhered to the root samples, the root As : Pb concentration ratio would be similar to that of the soil or LA source and, for a given LA treatment, independent of fertilizer source and rate. The root As

: Pb concentration ratio in all treatments of the -LA soil (ranging from 0.33 to 1) could not be unambiguously compared to that in the soil because soil Pb was below our detection limit of $5 \text{ mg}\cdot\text{kg}^{-1}$. The root As : Pb concentration ratio in all the +LA soil treatments ranged from 0.7 to 18 and was consistently higher than in the LA source (0.36) or in the +LA soil (0.34). Furthermore, instead of being constant across treatments, root As : Pb concentration ratio was proportional to fertilizer rate. It decreased with increasing AHS rate ($y = 2.818 - 0.083x$; $r^2 = 0.42^{**}$), indicating Pb enrichment relative to As; and it also increased with increasing MAP rate ($y = 4.260 + 0.529x$, $r^2 = 0.60^{***}$), indicating As enrichment relative to Pb. These results suggest that the root cleaning procedure effectively removed soil and LA particles that could bias the As and Pb analyses; hence, the root As and Pb concentrations accurately reflect As and Pb absorbed by the roots.

Root As concentration was influenced by LA and type, with significant two- and three-way interactions (Table 1). Root As concentration was higher in the +LA soil than in the -LA soil, consistent with a simple mass action effect (Fig. 3b). Fertilizer type and rate had no effect on root As concentration in the -LA soil. In the +LA soil, root As concentration was higher in the MAP than in the AHS

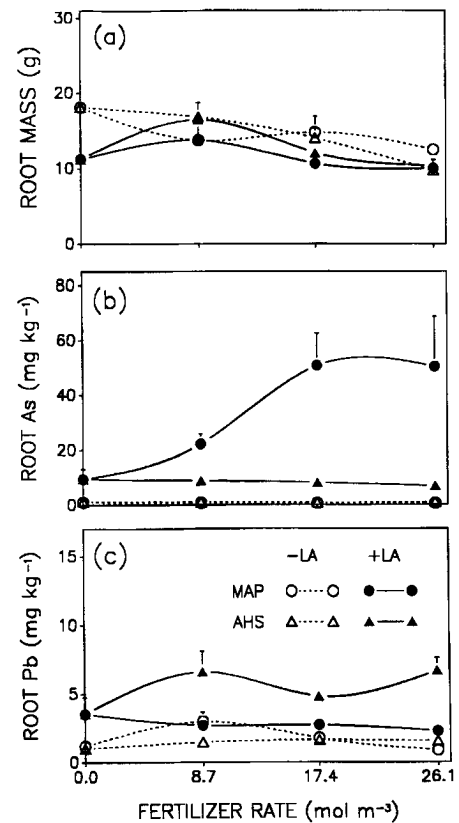


Fig. 3. Influence of lead arsenate amendment (-LA, +LA), fertilizer type [monoammonium phosphate (MAP); ammonium hydrogen sulfate (AHS)], and fertilizer rate on (a) apricot root mass, (b) As concentration, and (c) Pb concentration. Standard errors for individual mean values ($n = 4$) are indicated by error bars or are hidden by the symbols.

treatments at fertilizer rates of 8.7 mol·m⁻³ (P ≤ 0.05) and at 17.4 and 26.1 mol·m⁻³ (P ≤ 0.001). The highest values exceeded those reported for below-ground organs of other plant species grown in LA-enriched soils (Woolson, 1983). Root As concentration was positively related to fertilizer rate in the MAP-amended +LA soil ($y = 10.488 + 1.743x$; $r^2 = 0.41^{**}$) and was independent of rate in the AHS-amended +LA soil ($r^2 = 0.06^{**}$). Using higher-order regressions did not improve the relationships. For a given treatment, root As was higher than shoot As concentration; this finding is consistent with Woolson's (1983) conclusions.

Root Pb concentration was influenced by LA and type, with significant LA × type and LA × type × rate interactions (Table 1). Adding LA to the unfertilized soil increased root Pb concentration from 1.2 to 3.5 mg·kg⁻¹, consistent with a mass action effect (Fig. 3c). These concentrations were lower than the values reported for forest-tree species roots (50 to 250 mg·kg⁻¹) grown in soil containing ≈ 100 mg Pb/kg (Rolfe, 1973). Fertilizer type had no effect on root Pb concentration in the -LA soil. In the +LA soil, root Pb concentration was higher in the AHS treatments than in the MAP treatments at all fertilizer rates. This effect may be related to higher Pb volatility in the more acidic AHS-amended soil (Lindsay, 1979). Like shoot Pb concentration, root Pb concentration was independent of fertilizer rate in the MAP-amended +LA soil ($r^2 = 0.14^{NS}$).

Peryea (1991) reported that addition of P salts to aqueous suspensions of LA-contaminated Burch soils increased solution concentrations of As. Adding ammonium sulfate did not promote As release. Our experiment results, in which MAP but not AHS starter fertilizer increased the As concentration of apricot liners grown in a LA-contaminated soil, are consistent with a mechanism of P-enhanced As volatility and consequent increased soil As phytoavailability. This effect was not biased by biomass dilution because shoot and root biomasses were constant regardless of fertilizer type and rate. Although our experiment design does not permit definitive confirmation of this mechanism because soil pH, EC, and likely other soil chemical properties differ in the MAP and AHS systems, the empirical relationship between MAP amendment and increased plant As concentration is unambiguous.

Phosphorus addition had no effect on apricot shoot and root Pb concentrations. This result conflicts with several reports of decreased plant Pb concentrations following P additions to Pb-enriched soils (Bassuk, 1986; Rolfe, 1973; Zimdahl and Foster, 1976). In contrast to our study, these studies used PbCl₂ or Pb(NO₃)₂ as their Pb source (both of which are more soluble than LA), and they used monocalcium phosphate as their P source, which is initially less soluble and ultimately less acidifying than MAP. Reaction of these Pb salts with phosphate would result in formation of sparingly soluble Pb-phosphate compounds, probably a pyromorphite mineral (Ma

et al., 1991), with consequent reduction in soil Pb phytoavailability. Preliminary evidence suggests that Pb volatility in LA-enriched soil already may be controlled by pyromorphite (M. Kalbasi, F.J. P., and W.L. Lindsay, unpublished data); hence, adding more phosphate should have little effect on soil Pb phytoavailability, consistent with the effect observed in this experiment.

Use of nondraining pots increased the salinity stress and likely enhanced the As availability to the test plants. The experimental conditions may simulate trees that are planted in spring in MAP-amended holes augered into fine-textured LA-enriched soil. After planting, salts and As leaching may be restricted because of poor water movement through the compacted bottoms and sides of the planting holes and because of inadequate water application rates. These factors particularly are problems in arid fruit-growing areas where the newly planted trees are individually irrigated with water from portable tanks until irrigation canals are filled.

First-season results of a field experiment with apples are consistent with those reported in this study (F. J. P., unpublished data). The significance of MAP starter fertilizer-enhanced soil As phytoavailability over a longer time is unknown and requires investigation.

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