

The History of Lead Arsenate Use in Apple Production: Comparison of its Impact in Virginia with Other States

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Abstract

Lead arsenate (PbHAsO₄) was first used in apple orchards in the 1890s to combat the codling moth, *Cydia pomonella* (L.), a destructive insect pest. This pesticide was very popular among farmers because of its effectiveness, low cost, ease of use, and persistence. Over the next 60 years the frequency and amount of lead arsenate applications increased. Increased use eventually led to development of pesticide resistance, which started the downward spiral of decreased efficacy requiring growers to increase rates and application frequency. Growers eventually switched to more viable alternates such as DDT. The basic nature of the elements in lead arsenate and its widespread use contributed to the contamination of thousands of acres across the United States. As more landowners become aware of the lead arsenate issue, questions arise about the potential risks to human and environmental health. The story of lead arsenate provides rich insight into pesticide application practices of the past and a benchmark by which to judge current practices in pesticide safety education.

Keywords: lead, arsenate, arsenic, history, soil, contamination, human, health, Virginia, apple, fruit, pest, management, pesticide, safety, environment

Introduction

In the late 1800s inorganic pesticides were used extensively to control pests in agriculture. These natural chemicals, including arsenic (As), copper (Cu), lead (Pb), and sulfur (S), were mixed in varying formulations and were quite effective in controlling pests. Newly emerging pests and growing labor costs led to increased pesticide use by growers.

Lead arsenate (PbHAsO₄) was first used as an insecticidal spray in 1892 against the gypsy moth, *Lymantria dispar* (Linnaeus), in Massachusetts. A few years later, growers began using it to combat the codling moth, *Cydia pomonella* (Linnaeus), a destructive insect pest of apples. This inorganic pesticide was very popular among farmers because of its immediate effectiveness. It was also inexpensive, easy to mix, and very persistent. Over the next six decades, multiple

applications of lead arsenate were sprayed each season to control apple pests. This increase in use eventually led to development of pesticide resistance, which in turn decreased its efficacy thus requiring growers to increase rates and application frequency (Frear and Worthley, 1935) to compensate. This issue eventually forced growers to switch to other methods of treatment, such as DDT.

The characteristics of the compounds that created this persistence – the basic nature of the elements in the pesticide – and the increase in application frequency and rates were key factors contributing to the contamination of thousands of acres across the United States.

Apple production in the late 19th and early 20th centuries was very widespread. High production reflected localized agricultural markets and low fruit yield per acre. This contrasts



Figure 1. Orchard workers harvesting apples with horses and wagons (1918). Norfolk Southern Collection, Virginia Tech Library.

sharply with today's world market and higher yields per acre. In 1925, for example, apples were grown on over 300,000 acres in Virginia (Taylor, 1926). Today Virginia farmers grow less than 12,000 acres of apples (Virginia Agricultural Statistics Service, 2006). In 1914, 15 million bushels of apples were harvested in Virginia (Fig. 1), the high for the years 1906 and 1925 (Mattice, 1927). There were 5,238,095 bushels (based on 42 pounds/bushel) harvested in Virginia in 2006. This national trend toward reduced acreage is evidenced by the fact that Virginia's present-day ranking among the 32 apple-producing states has slipped only three positions from its peak of third in the early 20th century (Taylor, 1926) to sixth today (Virginia Agricultural Statistics Service, 2006).

As a growing post-World War II population began migrating from urban areas into the American rural landscape, farmland was developed into subdivisions. These and other socioeconomic trends caused dramatic changes in agricultural practices. Foreign trade globalized farming which in turn caused many small farms to fail. Farm wages could not compete with those of modern technical and manufacturing jobs. Farmland values increased in parallel with the decreasing



Figure 2. A subdivision (Waynesboro, VA) built in a former apple orchard (2001).

ability of small farms to turn a profit, which resulted in land sales. Today, housing developments occupy the land where productive apple orchards once stood 75 years ago (Fig. 2).

As more homeowners become aware of lead arsenate's historical use pattern and its persistence in the environment, they raise questions about the potential risks to human and environmental health from soil residues (Hood, 2006). Can lead and arsenic move off site? Do they pose a hazard elsewhere? How do people who live on lands containing residues deal with the issue?

Many states throughout the country have dealt with this problem by setting state-specific standards for arsenic and lead residues in soil and drinking water. The Environmental Protection Agency (EPA) has also set national standards for arsenic and lead in drinking water and for lead in bare soil.

How did we arrive at this point in history? The story of lead arsenate provides a rich insight into the technology and pesticide application practices of the past. It also gives us a benchmark by which to judge today's pesticide safety practices and pest control methods. The following is an overview of lead arsenate's history and where its use has taken us.

History

Inorganic pesticides commonly used in agriculture from the mid-1800s to the mid-1900s are stable in the environment and soluble in water. Sulfur, one of the oldest pesticides, was used as a fungicide against powdery mildew. It was also quite effective in controlling mites of all species. Accidental discovery that Bordeaux mixture, a combination of copper and lime, was fungicidal came in 1885: a French farmer applied it to his grapes to keep children from eating them. Copper arsenate (copper acetoarsenite), famously known as Paris green, was the first commonly used arsenical (Ware and Whitacre, 2004). Copper arsenate was successfully used against the Colorado potato beetle, *Leptinotarsa decemlineata* (Say) in the 1860s and the codling moth, *C. pomonella*, in the 1870s. London purple, a mixture of arsenic, lime, and acids, was also used to control the Colorado potato beetle in the late 1800s. Calcium arsenate was a heavily used arsenical in the early to mid-1900s, mainly in the southern United States to control grasshoppers and the cotton boll weevil, *Anthonomus grandis grandis* (Boheman). It was also used as a general insecticide on a variety of vegetable crops (Murphy and Aucott, 1998). The most heavily used arsenical throughout the United States and worldwide was lead arsenate.

Arsenicals are stomach poisons. They only exert their toxic effects when ingested and have a very complex mode of action. First, they uncouple oxidative phosphorylation, an important energy-producing step of cells, by substituting the arsenate ion for phosphorus. Subsequently, the arsenate ion inhibits certain enzymes that contain sulfhydryl groups. Finally, both the arsenite and

arsenate ions cause the shape or configuration of proteins to change via coagulation (Ware and Whitacre, 2004).

During the First Industrial Revolution (late 18th to early 19th centuries), families migrated from rural settings to cities in search of steady jobs. This population shift caused a greater demand for many goods. The standards for produce increased along with a demand for variety. Many small sustenance farms suddenly grew into large commercial producers. Apple production, for example, grew from small-acre plots to orchards often hundreds of acres in size.

Accompanying this increase in acreage came an exponential jump in pest populations. Growers were suddenly faced with dual challenges of newly emerging pests and rising labor costs for pest control.

During the Second Industrial Revolution (late 19th to early 20th centuries), countries such as the United States, the United Kingdom, and France began investing large amounts of resources in agricultural research. Poor health of fruit crops in Europe and the United States during the second half of the 19th century prompted much of this effort. Phylloxera infestations of grape and numerous disease and insect problems with apples, peaches, and other fruit crops plagued agriculture on both continents. As a result, new chemical products, application technology, and methods introduced in Europe found their way to the United States. Nowhere was this more evident than in the 1889 Paris Exposition (Riley, 1891). The works of American and European pioneers in pest management were exhibited at the event. New spray machines, nozzles, and application methods were demonstrated and

eventually adopted by farmers anxious to combat the pest and disease problems of the day. The U.S. Department of Agriculture (USDA) expanded its efforts in this area, creating new departments and hiring some of the first pest management specialists. This activity gave rise to much of the early land grant university pest management programs, and pioneers such as C.V. Riley and W. B. Alwood. The land grant university system tested new technologies and transferred these and other innovations to farmers via early Extension programs.

Demands for viable pest controls involved adopting both chemical and nonchemical remedies. The beginnings of integrated pest management (IPM) were documented during this era. USDA published a method for growers to trap codling moths by using bands of cloth on the trunks of apple trees integrating this with chemical controls (Marlatt and Orton, 1906). Alwood (1935) who documented the use of this method also used scouting and sampling methods to monitor the population of codling moths in his commercial orchard to determine effectiveness of his spray applications. Despite increased awareness of IPM, however, chemical controls far outgrew any other method. The reason was simple: economics. Even when the need for caution in pesticide use was well documented, farmers continued to use chemicals because they were economically effective.

As early as 1907, lead was recognized for its cumulative properties. Agricultural workers preferred using arsenic because lead was an accumulative poison. It was believed that arsenicals, except lead arsenate, should be permitted in agriculture. In 1907, a controversy erupted in Europe when the

German Imperial Health Commission opposed the use of lead arsenate on grapes because arsenic and lead were found in the wines (Moore, 1935). Nonetheless, lead arsenate became the most popular and widely used insecticide of the period.

Lead arsenate

As mentioned earlier, lead arsenate (PbHAsO_4) was first used in 1892 as an insecticidal spray against the gypsy moth, *Lymantria dispar* (Linnaeus). It replaced copper arsenate because of the latter's phytotoxic effects when used at high rates. When lead arsenate was used as a foliar spray (Fig. 3), it adhered well to the plant surfaces, allowing the pesticidal effects to last a long time (Peryea, 1998a).

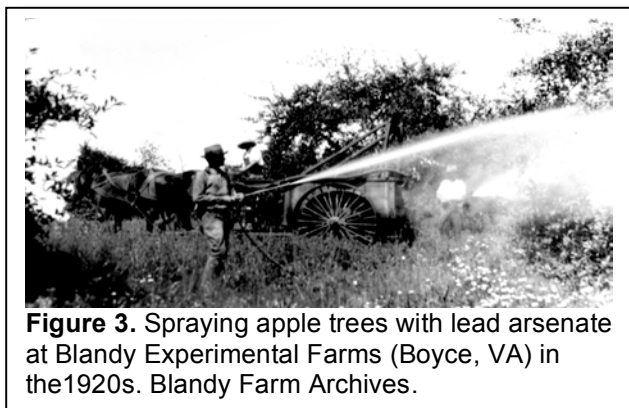


Figure 3. Spraying apple trees with lead arsenate at Blandy Experimental Farms (Boyce, VA) in the 1920s. Blandy Farm Archives.

Because of its persistence, lead arsenate quickly became a commonly used insecticide in fruit tree orchards and one of the earliest insecticides used against major pests such as the codling moth. Fruit damage occurs when the larvae tunnel into the sides and calyx end of the apple and proceed all the way to the core. This damage greatly lowers the storage quality and market value of the fruit, leading to a loss in yield (Hull et al., 1995). According to the American Pomological Society (1976), without treatment, the codling moth



Figure 4. A former pesticide-mixing site in Mint Spring Recreational Park (Crozet, VA) (2002).

would regularly damage 20% to 95% of the apples in every orchard.

Initially, farmers mixed lead arsenate on site (Fig. 4) by reacting soluble lead salts with sodium arsenate. However, lead arsenate was also sold as powders and pastes.

Lead arsenate formulations became more refined over time. Eventually, two principal forms emerged: basic lead arsenate, $Pb_5OH(AsO_4)$ (Fig. 5), was

used in certain areas in California; acid lead arsenate, $PbHAsO_4$, was used everywhere else (Peryea, 1998a). Lead and arsenic were mined locally. For example, in the mid-Atlantic region of the United States, the Briton Mine, located in Southwest Virginia (Floyd County), was an important source of arsenic. That mine, which opened in 1903 and closed in 1919 (Chaffin, 2003), also converted arsenopyrite ($FeAsS$) to white arsenic (As_2O_3) on site and shipped it to various pesticide manufacturers. White arsenic is a base component of lead arsenate. Southwest Virginia was also an important source of lead. A mine in the Saltville area (Smyth County), which supplied lead as far back as the Revolutionary War, closed in the early 1980s.

Lead Arsenate as Part of a Commercial Fruit Operation

Unique farming techniques were used in a typical commercial orchard in the early 1900s to culture apples and other fruit

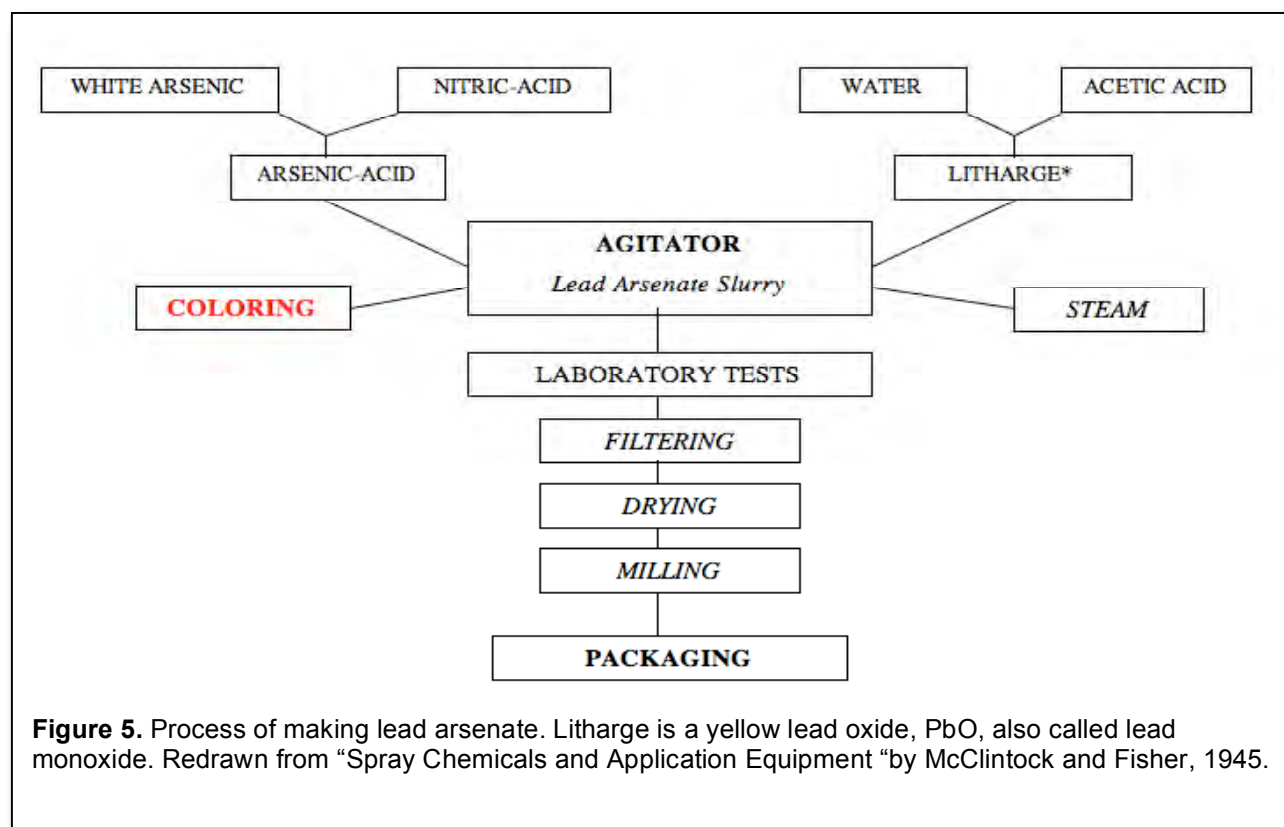


Figure 5. Process of making lead arsenate. Litharge is a yellow lead oxide, PbO , also called lead monoxide. Redrawn from "Spray Chemicals and Application Equipment" by McClintock and Fisher, 1945.

trees. To achieve maximum yield, an orchard had to be well pruned, well cultivated, and well fed (Dickens and Headlee, 1910). One of the most important aspects of commercial production was the fungicide and insecticide spray schedule. Not only was the schedule important – spray mixing, materials, and equipment also played vital roles in a successful operation.

Copper sulfate, lime, and Bordeaux mixture were the three main fungicides applied in commercial orchards. Copper sulfate was used alone during the dormant season, while lime was mainly used to neutralize acid solutions. Lime combined well with other materials to make a mixture that stuck to leaves and killed fungal spores and insects. Copper sulfate and lime were mixed on site and usually kept as stock solutions during the growing season. The two stock solutions were then combined to make Bordeaux mixture, called a 3-4-50 mixture (3 lbs. copper sulfate, 4 lbs. lime, 50 gals. water) (Kansas State Agricultural College, 1915).

Fungicides and insecticides were usually applied together (Table 1) to save growers time and money. Contrary to the belief at the time (ca. 1915) by some growers, Bordeaux mixture alone had no insecticidal qualities and had to be combined with an insecticide. Standard procedure was to add 2 pounds of lead arsenate to every 50 gallons of water or 10 pounds to a 250-gallon tank. Lime-sulfur was also mixed on site and stored for use during the growing season. It was combined with lead arsenate at 2 pounds to 50 gallons of solution for insecticidal purposes (Ibid).

The spray schedule was divided into three important sprays:

- The dormant spray was preferably applied in the spring before the buds opened as well as in the fall after leaf drop. A nozzle with a fine mist spray was used to coat the entire tree, including all the limbs and the trunk.
- The summer spray was applied with a fine mist nozzle to completely coat the leaves and fruit from every angle. Once the water from the spray evaporated, the fruit and foliage were examined for complete or partial coverage.
- The petal-fall spray was the most effective single application for the control of the codling moth (Ibid). After petal-fall, the calyx was left open a short time. Because this was where most of the insect pests entered the fruit, it was very important to fill the calyx cups with insecticide.

Lead arsenate came in two forms, powder and paste. It was either purchased or made by the grower (Fig. 6). The dry formulation was easily stored from season to season, and the paste formula was more easily mixed with water. However, the dry formulation was usually more than double the price of the paste. Whether using the dry or paste formulation, it was important to keep the resulting mixture agitated so the solution would not separate before it was sprayed on the trees. Copper arsenate (Paris green), arsenite of lime, and nicotine were other recommended insecticides.

Reliable machinery was very important to ensure efficient spraying. The kind of machine best suited for an orchard depended upon the size of the trees and the acreage. When choosing equipment the first consideration was power, and second was the pump, hose, extension

Table 1. General spray recommendations for apples in 1915.

Insects or Fungi	Spray to Use and Time of Application*
1. <i>San Jose scale</i>	Dormant strength commercial or home-boiled lime-sulfur (6-7-8). Just before the buds swell.
2. <i>Apple scab</i> <i>Plum curculio</i> <i>Spring cankerworm</i> <i>Apple rust</i>	Lime-sulfur, summer strength (6-7-8), plus 2 to 4 pounds arsenate of lead. When cluster buds are showing pink, but before blossoms open.
3. <i>Codling moth</i> <i>Apple scab</i> <i>Black rot</i> <i>Apple rust</i>	Lime-sulfur, summer strength (6-7-8), plus 2 to 4 pounds arsenate of lead. Start when petals are two-thirds off the blossoms.
4. <i>Apple blotch</i> <i>Plum curculio</i> <i>Codling moth</i> <i>Black rot</i>	Bordeaux mixture (3-4-50) plus 2 pounds arsenate of lead, if the weather is dry. Three weeks after No. 3. If wet, use lime-sulfur and follow with the Bordeaux as soon as dry.
5. <i>Apple blotch</i> <i>Black rot</i> <i>Plum curculio</i> <i>Codling moth</i>	Bordeaux mixture (3-4-50) plus 2 pounds arsenate of lead. Two to four weeks after No. 4.
6. <i>Codling moth</i> <i>Apple blotch</i> <i>Bitter rot</i> <i>Black rot</i>	Bordeaux mixture (3-4-50) plus 2 pounds arsenate of lead. Eight to ten weeks after No. 3.
7. <i>Bitter rot</i> <i>Blotch</i> <i>Codling moth</i>	Lime-sulfur or Bordeaux mixture (3-4-50) plus two pounds arsenate of lead. Shortly before fruit begins to ripen.
*This was a general guideline reproduced from the Kansas State Agricultural College: Agricultural Experiment Station Bulletin (1915) and not all applications were likely needed. Spray Nos. 2, 3, 4, and 6 are those ordinarily recommended.	

rods and nozzles (Dickens and Headlee, 1910). Although hand pumps were recommended at the time for fewer than 500 trees (Ibid), power sprayers (Fig. 7) were the choice for large commercial orchards of 500 to 2,000 trees (Ibid). Power to pump the liquid was obtained three ways: traction gearing, by hand, or by gas engine (Kansas State Agricultural College, 1915). Gas engines supplied high and continuous pressure and were found on the best sprayers.

Selecting the right nozzle was also crucial. The nozzle affected the spray application and worked differently depending on the pressure obtained by the pump (Fig. 7). For example, during the petal-fall spray, the spray had to be

delivered from above and applied in a downward fashion to fill the calyx cups. The "Bordeaux" nozzle was often recommended during petal-fall (ibid). However, it delivered a flat, undesirable spray. The "Friend" nozzle had a medium-sized aperture and was found to be especially effective during petal-fall. In "Spraying Apples" (Dickens and Headlee, 1910), two types of spray were used: the strong dashing type and the mist type. The Bordeaux nozzle produced the first type and the Friend nozzle or the Vermorel produced the second. Dickens and Headlee (1910) recommended the Friend nozzle because it never caught the branches of the tree.



Figure 6. Lead arsenate displayed in two different types of formulations, powder (left) and paste (right).

An important indicator of the type or quality of spray produced was measured by the effective spray drive. This term referred to the distance from the nozzle opening at which the spray turned the leaves sufficiently to wet them on both sides while the nozzles were in motion. A nozzle with six holes in the whirl disc (Fig. 8), usually preferred by farmers for its effective spray drive, produced a spray cone that carried three times farther than the fanlike cone produced by a nozzle with only two holes in the whirl disc (Hough, 1928).

Not only did nozzles and their arrangement affect spray efficiency – so did the operator. In addition, the spray equipment selected, the condition and the pump capacity all played an important role (Ibid).

In 1915 wooden tanks were the rule. Metal tanks were the exception because of the corroding effects of certain spray solutions. A round tank was preferred

because agitation was easier and all of the solution could be removed from the tank. Another important piece of equipment necessary to successfully spray large trees was a “tower” (Fig. 9). Made of wood or metal, it allowed the applicator to spray down onto the tree. This was especially important during the petal-fall spray when the insecticide had to reach inside the calyx cups. Tower height was a factor because the spray pattern depended on using a combination of pressure, nozzle type, extension wand length, and tower height to reach the tops of apple trees, which at that time could exceed 30 feet.

Some operations used elaborate underground piping systems (Fig. 10) to pump water from spring-fed wells through pipes to equipment in the orchard. Tanks were usually established throughout the orchard for storage and gravity sometimes played a role in distributing water to pumps or hydrants.

F. E. MYERS & BRO. ASHLAND, OHIO.

The Myers Junior Power Spray Outfit.

Fig. 1602. Fitted With 2 Inch Myers Duplex Pump.
Gear Driven. Complete except Engine. Guard Rail in Position.

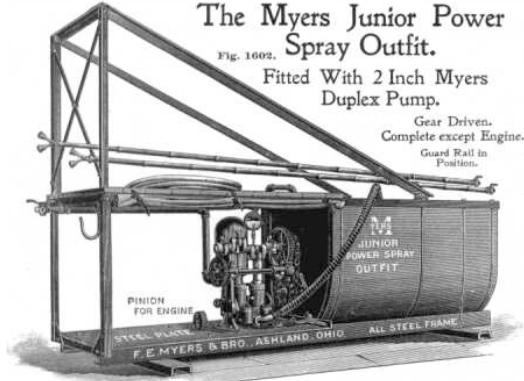



Fig. 1603. Guard Rail Folded and Curtain Closed.



Figs. 1602 and 1603 illustrate the Myers Junior Power Spray Outfit. It is furnished complete as below: in fact everything except the engine. The frame is made of channel steel, making it very stiff with the least possible weight. It is fitted with wrought steel bed plate on which the engine and pump are mounted, thus making a solid foundation that will not vibrate when under the most extreme pressure. The tank is made of selected cypress, holds 100 gallons, and is fitted with a rotary agitator with wooden blades. Agitator is driven by a link chain, which makes a flexible joint and relieves the tap of all strains. There are no pipe connections between the pump and tank. The pipes extend upward from the pump and enter tank from the top, thus preventing all twisting strains which cause leaks. The two cross channel bars underneath the frame fit the bolsters of wagon, preventing forward or backward movement. Side curtains for protection of engine and pump. Guard rails fold up for transportation or storage. See Fig. 1603. Length of outfit is 8 feet.

Be sure to state on Order the H. P. M. of Engine, Diameter of Crank Shaft and Size of Key.

PRICE LIST, Represented by Figs. 1602 and 1603.

Fitted for 1 or 1 1/2 H. P. Gasoline Engine.

No. 10, Myers Junior Power Spray Outfit, as illustrated above, complete with No. 436, (2 inch) Duplex pump, with plunger for crankshaft of Engine. Price.....(F.O.B.) \$150.00

Equipped as follows:

1 100 gallon cypress tank.	2 Fig. 1560 Y's.
1 foot 1 inch suction hose.	2 8 foot Bamboo Pipe Extensions.
3/4 inch discharge hose with fittings for 2 leads.	Rotary Agitator, Pressure Gauge,
1 Fernbro Nozzle—(2 straightaway and 2 45-degree.)	Ball Valve, Double Cut off,
Engine not included.	and Strainer.

No. 12, Myers Power Spray Outfit, with 150 gallon cypress tank and heavier sills, otherwise the same as the No. 10 Junior Outfit. Price.....(F.O.B.) \$170.00

F. E. MYERS & BRO. ASHLAND, OHIO.

Myers Spray Nozzles.

Fig. 1563. Regular Vermorel Nozzle. (French) \$0.80.

Fig. 1564. Graduating Vermorel Nozzle. (French) \$0.80.

Fig. 1019. Regular Bordeaux Nozzle. (Imp.) \$0.90.

Fig. 1378. Bordeaux Special (Para) \$1.00.

Fig. 550. Cyclone, Side Outlet. (Imp.) \$0.40.

Fig. 1291. Long Distance. (Paley) \$0.90.

Fig. 642. Imperial. (Glass) \$0.40.

Fig. 548. Sprinkling. (Imp.) \$0.40.

Fig. 1342. Myers Spray Nozzle (S&C).

All Myers Spray Nozzles are put up in individual boxes as shown in Fig. 1342, making a convenient package for placing on shelf.

Fig. 1563. All fruit growers rightly contend that the nozzle is the most important part of the spraying outfit. It must be arranged to throw a mist and not drops. It is also a fact that more harm is done by using Sprinkling Nozzles than if no spraying was done at all. By putting on too much material you damage the foliage. The best results are obtained by covering the entire surface and with just as little in one place as possible. We manufacture a complete line of nozzles for the smallest hand pump to the largest power outfit.

Fig. 1564. The Vermorel Nozzle fitted with two removable plates giving fine and coarse spray.

Fig. 1019. Shows Open Cap and Removable Spray Disc, also Packing Ring.

Fig. 1378. Sectional View of Fig. 1544.

Fig. 550. The Bordeaux Nozzle will throw a fine fan-shaped spray and will readily free itself should it become clogged, by turning the cock handle. It will throw a solid stream or it may be shut off altogether, as desired. Fig. 1378 has double face; gives greater wear.

Fig. 642. All nozzles fitted with standard 1/4 in. pipe thread.

Fig. 1291. Shows Open Cap and Removable Spray Disc, also Packing Ring.

Figure 7. Pages of 1922 Myers Sprayer catalog showing state-of-the-art equipment (Myers & Myers, 1922).

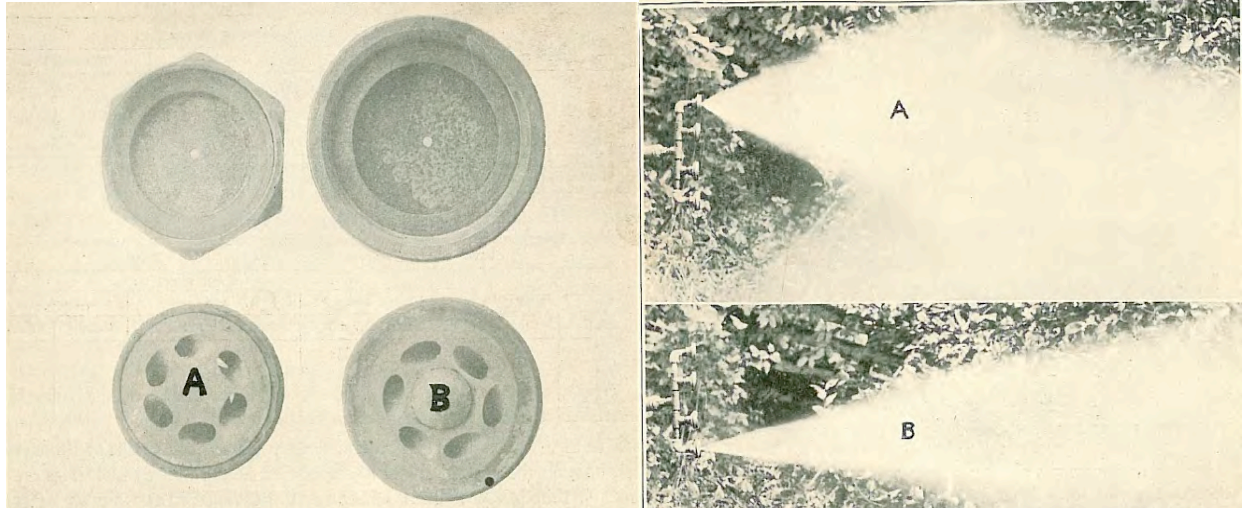


Figure 8 shows two types of whirl discs used in sprayers. On the left, disc A has two holes and disc B has six holes. On the right, two photographs show the spray patterns created by these discs. Pattern A, corresponding to the two-hole disc, shows a wide, shallow spray. Pattern B, corresponding to the six-hole disc, shows a narrower, more concentrated spray.

Figure 8. Two-hole (A) and six-hole (B) whirl discs (left) and corresponding spray patterns (right) (Hough, 1928).



Figure 9. Applicator spraying apple trees with a handheld boom while standing on a “tower.” Sprayer is followed by a horse-drawn wagon carrying water (1940). Norfolk Southern Collection, Virginia Tech Library.

The spray chemicals were transported in barrels and loaded onto wagons pulled by one or two horses. The wagons were pulled to the top of the orchard and the barrels were attached to a pump by hoses connected to a water tank. Water was continuously pumped to the barrel (by hand or by gas engine) to mix the chemicals properly. Trees were sprayed individually by hand to ensure good coverage. Sometimes, however, the pipes in the orchard were used to pump chemicals up to the storage tanks, making application easier for the workers. Instead of pulling the chemicals by wagon, workers could then hook their hoses up directly to the hydrants located throughout the orchard and spray. This process called “chemigation,” was very risky because of the possibility of leaking pipes. The long-term environmental effect of leaking pipes was demonstrated in the contamination of Barber Orchard in Waynesville, NC in 1999.

Fruit Residues

Because of its great success in controlling the codling moth, lead arsenate became the primary insecticide used in apple orchards. According to



Figure 10. Rusty pipes pulled from the orchard and piled together at Snead Farm (Shenandoah National Park, VA), 2001. The pipes were once part of an underground system used to deliver water and possibly pesticides throughout the orchard.

Marlatt (1905), essentially all commercial apple orchards were treated with arsenic.

Several sprays of lead arsenate, at a rate of 2 to 4 pounds per 100 gallons of water, were applied to the apples during a single season. Applications generally occurred one to three times a season and, over time, gradually increased to five to six times a season. According to Murphy and Aucott (1998), the USDA’s Agricultural Statistics yearbook states that in 1929 the U.S. consumption rate of lead arsenate was 29.1 million pounds. The rate peaked in 1944 with an estimated 86.4 million pounds and then dropped to 3.9 million pounds in 1973. From the peak usage in the 1940s until the 1970s, lead arsenate use declined because more effective pesticides became available. However, it was still used extensively, and the application rates and frequency continued to climb. An example of the high rates used is documented in a 1966 agricultural handbook (USDA, 1966). It suggests that to control codling moth in apples a grower should use 3 pounds of active ingredient per 100 gallons or 30 pounds of active ingredient per acre.

By the end of the 1930s other pests that were no longer susceptible to these high doses of lead arsenate became major problems. Alternatives to lead arsenate were either less effective or were more toxic to non-target plants and animals.

With the heavy use of lead arsenate well documented, researchers across the country began to look at residues on the fruit. In 1919 they discovered that common washing practices did not adequately remove arsenic residues from produce (Shepard, 1939). A study conducted by the Virginia Agricultural Experiment Station (Hough et al., 1931) concluded that three sprays of lead arsenate applied in May and June did not require removal of spray residue at harvest time. However, when a third or fourth spray of lead arsenate was applied in July, followed by dry weather, excessive residues remained on the apples at harvest. Wiping or brushing apples removed only about one-third of the total arsenical residues. In contrast washing fruit in a diluted hydrochloric acid solution consistently removed excessive residues (roughly 70%) (Fig. 11).

According to Frear and Worthley (1935), the smaller apples from the lower tree limbs usually carried the greatest amount of spray residue and were collected for their study. A hydrochloric acid bath (various strengths) in a flotation washer could remove only 70% of lead and 75% of arsenic residues from the apples. Adding wetting or foaming agents to the bath did not significantly enhance removal of residues. Raising the temperature of the bath however increased residue removal to approximately 80% of lead and 85% of arsenic. The variety of apple also played an important factor in the ease of the residue removal.

When lead arsenate was later used in an oil-spray program, it was necessary to remove the spray residue with a two-unit (tandem) type washer (Walker, 1949). The fruit was put through the first unit which contained 40 to 60 pounds of sodium silicate per 100 gallons of water at 90°F to 100°F. Then the fruit went through the second unit which contained 1.5% hydrochloric acid at 90°F to 100°F. The apples were then rinsed with a large amount of fresh water. Of course, residue removal in the field is affected by many factors, including temperature, humidity, wind speed, rainfall, and the chemical makeup of the spray mixture (Frear and Worthley, 1935).

On February 21, 1933, the federal government passed a law limiting lead residues on fruit to 0.025 grains of lead per pound of fruit. It was believed that arsenic residues were acceptable on fruit, but lead was hazardous. Although nothing could beat its efficacy, lead arsenate as a compound was unacceptable (Moore, 1935).

Finally, a revolutionary breakthrough in 1947 changed pest management. Soon after the introduction of DDT and other

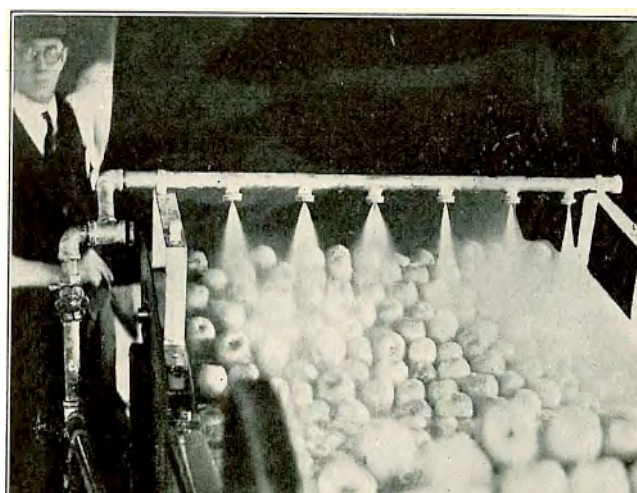


Figure 11. Residues were such a problem that apples were run through special rinses of water and acid baths to remove them. (Hough et al., 1931).

synthetic insecticides the use of inorganic insecticides began to decline. Moreover, the codling moth had already developed resistance to the arsenical compounds. DDT and other synthetic insecticides were more effective in controlling pests. EPA officially banned all insecticidal uses of lead arsenate in the United States and cancelled its registration on August 1, 1988 (USEPA, 1988).

Soil Residues

The heavy use of lead arsenate in apple orchards over many years may have contributed to major environmental problems, specifically the buildup of arsenic and lead in the soil. Because naturally occurring arsenic and lead are quite stable and do not break down in the environment, they accumulate with each use in orchard soils. Depending on soil type, each element, particularly lead, tends to linger in the top 5 - 20 cm of the soil. Leaching of arsenic has occurred below the 20-cm level (Veneman et al., 1983). Given favorable soil conditions (pH, mineralogy, and precipitation), arsenic has been found to leach deeper (Peryea and Creger, 1994; and Warner, 1996a) and possibly contaminate groundwater.

Farmers and homeowners who use old orchard sites for crop production or gardening must be mindful of lead arsenate residues in the soil. Crops planted in contaminated soils take up heavy metals (Kenyon et al., 1979; Levander et al., 1977; Merry et al., 1986). The amount of lead taken up by plants is proportional to the amount applied to the soil (Kenyon et al., 1979). However, most plants do not hyperaccumulate either arsenic or lead. While certain metals can be phytotoxic to crops such as carrots, turnips, and

peanuts (Alloway, 1990), food crops that are more tolerant to these metals will not show signs of toxicity. The outcome of harvesting these crops could be harm to people or livestock eating these foods.

When lead arsenate-contaminated soil is amended with a fertilizer containing phosphate, the fertilizer enhances the release of adsorbed arsenic to the solution phase by competing for adsorption sites. This process increases solubility, mobility, and phytoavailability, (Peryea, 1998b) making arsenic more available for plant uptake, which has implications for plant growth and food safety. Newly released arsenic might also move off site by sediment runoff or through groundwater if soils are very sandy (Peryea and Kammereck, 1997).

The Association for the Environmental Health of Soils (1998) surveyed all 50 states to determine how soil arsenic is regulated nationally. Survey questions probed soil screening thresholds, establishment of remedial action levels, and use of risk assessments. Twenty-one of 34 states who responded to the survey indicated they had established specific cleanup levels for soil arsenic. Background levels, site-specific levels, and risk-based levels were the primary determinants for setting cleanup levels.

Health Risks

In November 2005, under authority of the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA), EPA released its biannual list of hazardous substances. The top two substances on the list were arsenic and lead; they've held that ranking for the preceding ten years. These two substances are most commonly found at sites on the National Priorities List (NPL).

Arsenic, listed first on EPA's Hazardous Substances List (ATSDR, 2007a, 2007b), is a natural element found in both the earth's crust and in water. It's also found in plants, which absorb lead and arsenic through their roots, and in animals that eat these plants and drink contaminated water. The Food and Drug Administration (FDA) found that humans consumed a small amount of arsenic daily in food (Levander et al., 1977). According to Peryea (1998a), arsenic is released into the environment through ore smelting, cement manufacturing, and the combustion of fossil fuels in electrical power plants. Arsenic can also leach from lumber treated with chromated copper arsenate (CCA). CCA can increase soil arsenic levels near the site where the wood is in contact with the soil (Stilwell, 2005) (USEPA, 2007a, 2007b).

Short-term, or acute, arsenic poisoning can appear very quickly, usually within an hour if ingested orally or if there is exposure to very high levels not normally found in the environment. First symptoms are difficulty in swallowing or burning lips followed by excruciating abdominal pain, forceful vomiting, diarrhea, thirst, and leg cramps. This

can lead to spasms, shock, paralysis, coma, and death. In a few cases of acute arsenic poisoning "Mees lines" may appear. (Mees lines are transverse white bands on the nails.) Arsenic is extremely toxic: a minimal lethal dose for humans is 50 - 300 milligrams (mg)/kg of body weight (ATSDR, 2008a).

Long-term, or chronic exposure, is classified as either carcinogenic or noncancerous. Noncancerous symptoms include numbness, tingling or burning sensations in the extremities (a sensation of "pins and needles"), hair loss, weakness, loss of appetite, nausea, or vomiting. Damage to the central nervous system, liver, and kidneys may occur. One of the most common characteristics of chronic arsenic exposure is the appearance of skin disorders, including small corns or warts on the palms, soles of the feet (Fig. 12), and torso. These corns or warts may ultimately develop into non-melanoma forms of skin cancer. Thickening of the skin and darkening pigmentation on the neck, eyelids, nipples, and underarms are also common.

Lifelong ingestion of 0.05 - 0.10



Figure 12. Cancerous lesion on hand (left) and nodular hyperkeratosis on feet (right) caused by chronic exposure to natural arsenic in groundwater in Bangladesh (Wilson, 2000 and Chowdhury et al, 2000).

mg/kg/day increases the risk of skin cancer (ATSDR, 2007a). Arsenic ingestion (oral, dermal, or inhalation) has also been reported to increase the risk of cancer throughout the body, especially in the liver, kidneys, bladder, and lungs. Humans with long-term exposure to high levels of airborne arsenic in or around smelters have a greater risk of developing lung cancer (Ibid). The federal government has established a maximum contaminant level (MCL) of 10 parts per billion (ppb) for arsenic in drinking water (Ibid). In addition, the Occupational Safety and Health Administration (OSHA) has established a maximum permissible airborne exposure limit of 10 $\mu\text{g}/\text{m}^3$ for inorganic arsenic (Ibid).

No federal safety levels have been established for arsenic in soils. Arsenic is found in both organic and inorganic forms. Inorganic arsenic may be present in either the pentavalent (AsO_4) or the trivalent (AsO_2) form. It is believed that the pentavalent form exerts a toxic action only after conversion to the trivalent form, which is the more toxic of the two (Rybolt, 1970).

Lead, listed second on EPA's Hazardous Substances List, is also naturally occurring and can be found in all parts of our environment (Hammond et al., 1972). Lead was not only used in insecticides, but also in gasoline until 1995 (Peryea, 1998b), and in paints until it was banned from home use in 1978.

Lead dust released by smelters and mines can contaminate nearby soil. It can also be found in everyday items like pottery glazes, lead shot, and fishing weights and can leach from improperly glazed ceramic ware and leaded crystal. However, because of health concerns,

its uses have been reduced or, in some cases, eliminated altogether (ATSDR, 2007c). There are four main sources of high lead contamination in soils: paint, gasoline, insecticides, and industrial fallout (Peryea, 1998b).

Lead can affect almost every organ and system in the human body. The effects are the same whether swallowed or inhaled. The central nervous system is the most lead sensitive. Toxic levels cause neurological problems, especially in children (Hammond et al., 1972). Exposure to high levels of lead can lead to premature births, decreased mental capacity, learning difficulties, and reduced growth in young children. Unborn babies can also be exposed to lead through their mothers. In adults, lead exposure mainly affects the peripheral nervous system. Symptoms include loss of appetite, fatigue, anemia, and abdominal pain (ATSDR, 2007c). Eventually, vision, hearing, and muscle coordination may be impaired. EPA limits lead in air to less than 1.5 $\mu\text{g}/\text{m}^3$ and in drinking water to 15 $\mu\text{g}/\text{L}$. EPA considers lead a hazard if it exceeds 400 parts per million (ppm) in bare soil in children's play areas (ATSDR, 2008b).

In old orchards treated with lead arsenate, rodents (pine voles, meadow voles, and white-footed mice) inhabiting these soils had lead concentrations in kidney, liver, and bone tissues markedly higher than in tissues from control animals (Haschek et al., 1979). Lead accumulation correlated with the degree of substrate feeding and movement of rodents. Predators, however, showed no increase in mortality by feeding on contaminated prey from lead-contaminated orchards (Stendell et al., 1989). Arsenic accumulation was not tested in rodents.

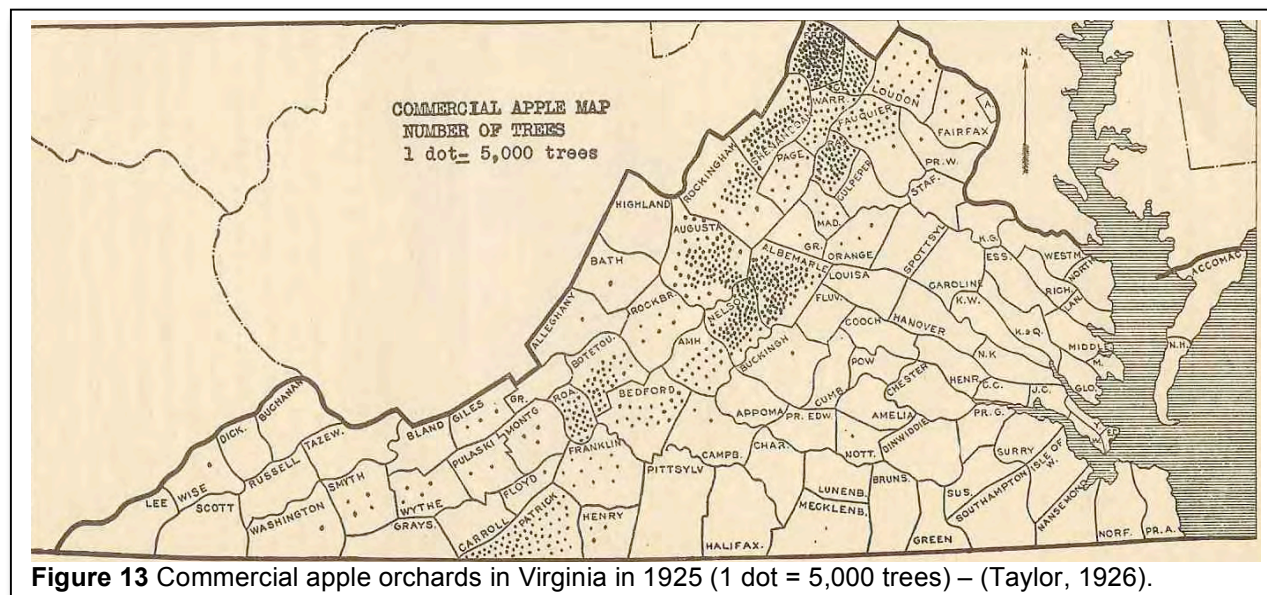


Figure 13 Commercial apple orchards in Virginia in 1925 (1 dot = 5,000 trees) – (Taylor, 1926).

Mortality of orchard workers in Washington State who were exposed to lead arsenate was assessed during a 1938 study and later reexamined in 1990 to determine whether mortality could be attributed to lead arsenate exposure. The 1990 study found that mortality due to cancer among men or women was not significantly higher than the original study. However, the 1938 study could have been flawed due to the low number of study subjects, the type of arsenical compound studied, the lower cumulative concentration of arsenic exposure, or the fact that no other diseases were studied among the subjects (Tollestrup et al., 1995).

The Impact of Years of Lead Arsenate Use across the United States

Virginia

At some point between 1890 and 1950, lead arsenate was likely used to treat pests on most agricultural lands in Virginia.

The size of the apple industry in Virginia in the 20th century was such that it is likely lead arsenate was heavily used. From surveys conducted by the Virginia

Agricultural Statistical Survey (VASS) and the Virginia Agricultural Experiment Station, we know there were more than 10.4 million apple trees (8.1 million fruit bearing and 2.3 million nonbearing) in the state according to the Census of Agriculture (Taylor, 1926). Apple trees covered about 300,000 acres, depending on row and tree spacing.

According to the 1925 census, 4.3 million apple trees (43% of the total number of apple trees reported) were listed as commercial trees (Fig. 13). Approximately 125,000 acres of orchards in Virginia were operated by commercial growers who probably sprayed lead arsenate extensively to combat pests. Given the size of commercial orchard operations in the state, an astounding 77,400 tons of lead arsenate was estimated to have been applied in Virginia over a 20-year period (see Table 2 for calculation).

There were 116,000 farms (62% of all farms in Virginia) that reported growing apples in the 1920 Census of Agriculture. Most of the noncommercial orchards sold apples locally but could

Table 2. Estimated lead arsenate (tons) applied over a 20-year period in Virginia's commercial apple orchards.

# Trees	Trees /Acre	Acres	Gal. /Tree	Lbs. a.i./100 gal.	Loading /Tree (lbs)	Loading /A (lbs)	Loading /Yr (lbs) (20 yrs)	Loading /A (lbs) (20 yrs)	Statewide Loading (lbs) (20 yrs)	Statewide Loading (tons)
4,300,000	35	122,857	10	3	0.3	10.5	63	1,260	154,800,000	77,400

not ship because of their poor quality. Likely, these apples were not sprayed at all or not sprayed intensely enough to control pests like the codling moth. So lead arsenate might have been used, but not at the number of applications or coverage of a commercial orchard.

The leading commercial apple orchard counties in Virginia at that time were Albemarle, Clarke, Frederick, Nelson, and Warren. Winchester had the largest cold storage facility for apples in the United States. It anchored the Virginia apple industry, which was the third highest in the country (Taylor, 1926). Staunton and Crozet were also major apple shipment centers.

Virginia's apple industry has been declining steadily for many decades. Much of the decline can be attributed to poor tree maintenance and failure to replace dead trees. It is likely that the crop will always be grown in Virginia, but commercial operations are slowly being pushed out as land values continue to rise.

Land use patterns also changed as more people moved from the city into the country. Commercial orchards were usually built close to centrally located shipping hubs. Over time, orchard sites increased in value because of their proximity to towns or cities and became convenient sites for new homes. Building houses and subdivisions on former agricultural lands where pesticides were once heavily used has caused many homeowners to ask questions about soil residues and

safety. Many homeowners are questioning the safety of the soil in their backyards as well as the purity of their drinking water. They want to know what can be done to reduce their risks of exposure to lead and arsenic.

This issue gained an even higher profile in 1999. Several clients called Virginia Tech in reference to a major soil residue problem in a North Carolina subdivision that was once an apple orchard. They were concerned that a similar situation could exist in Virginia. In 2001, Schooley (2006) began a study to address their concerns. Research was conducted to determine the amount of lead arsenate residues remaining in a typical orchard site that had been in production between 1890 and 1988. To avoid real estate disclosure laws (Virginia General Assembly, 2006), most sites tested were on public lands located in western Virginia from Winchester in the north to Wytheville in the south. Findings from commercial orchard sites dating to 1897 showed residues ranging from nominal amounts to levels far exceeding standards set as safe in other states. The highest amounts of lead and arsenic found on an orchard site in the study were from the Snead Farm, now part of Shenandoah National Park in Warren County. Two case studies are documented below.

Case Study 1: Snead Farm –
Shenandoah National Park – Warren
County, VA

This 250-acre farm located in Warren County was a thriving commercial

orchard from the early 20th century until 1962 (Fig. 14 -16). Previously, several families owned and operated the Dickey Hill orchard before the Snead's. These families planted the original trees in the orchard dating back to 1897. In 1962, it was sold to the federal government by the Snead family for water rights to the Dickey Ridge Visitor Center along the Skyline Drive located in the Shenandoah National Park.

Due to increasing labor and equipment costs, stricter regulations for grading and inspecting, and a declining overseas market in the 1930s only large orchards could successfully compete. When the park was created in 1936, the Dickey Hill orchard was still too profitable to be condemned. However, by the 1960s, the orchard was no longer viable and was eventually incorporated into the park.



Figure 14. A few apple trees at Snead Farm in the Shenandoah National Park were the only ones left of a (1930's) commercial orchard.

Only a few apple trees still grow on the property interspersed within a dense mature forest (Fig. 14). The barn is still standing and is well maintained by the park, but only the house foundation remains (Fig. 15). Piles of old, rusty pipes show signs of a once-extensive



Figure 15. Snead Farm stone house foundation and barn (Shenandoah National Park), 2001.

irrigation system that supplied water to the entire orchard (Fig. 10). Although steep and extremely rocky, the terrain still shows evidence of terraces where the trees once stood in rows (Fig. 16). Soil analysis results showed very high levels of contamination for both lead and arsenic. There were definite examples of "hot spots" in the orchard. These spots could have been contaminated from mixing or spilling pesticides. Alternatively, a large amount of lead arsenate might have been sprayed and accumulated in the top few inches of the soil. Soil samples were taken in visible tree rows and combined for each row (Schooley, 2006).

Total lead and arsenic residues (ppm) found in soils sampled at Snead Farm

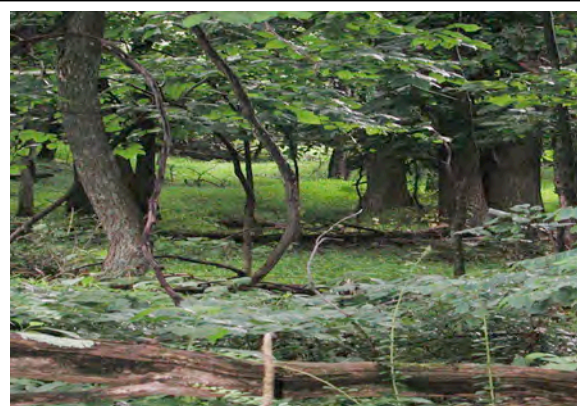


Figure 16. Snead Farm apple orchard in Shenandoah National Park. Former tree rows are evident here as terraces on the hillside.

varied depending upon location. Samples taken in the main orchard behind the farmhouse showed a lead concentration range of 104 – 701 ppm and an arsenic concentration range of 25.67 – 228.81 ppm. Samples taken near an apple-packing shed revealed lead concentrations of 474 – 548 ppm and an arsenic concentration of 116.49 – 154.50 ppm. A sample taken near an old well and a pile of what appeared to be irrigation or chemigation pipe showed a lead concentration of 436 ppm and an arsenic level of 41.60 ppm. Samples taken where apple trees were still growing (Fig. 15) contained lead concentrations of 167 – 774 ppm and arsenic concentrations of 51.50 – 286.12 ppm.

Case Study 2: Mint Springs
Recreational Park – Albemarle County,
VA

Mint Springs Park was developed from over 500 acres of land near Crozet, originally part of a commercial apple orchard in operation since 1880. Most of the hundreds of acres of apple trees in the park were overgrown with weeds and surrounded by new-growth forest (Fig. 17).

Portions of the original orchard located next to the park are still in commercial production today. According to a former orchard worker, lead arsenate used in the orchard or sold elsewhere was made on site at the farm. Other parts of the orchard were sold for development years ago and houses were built there. After learning of the orchard's history, homeowners in the development, concerned about their drinking water, turned to county officials for help.

A fishing lake and a swimming lake in the park were the main attractions for visitors. Albemarle County officials

concerned about possible exposure to the public, hired a Richmond firm, Froehling & Robertson, Inc. (F&R). In April 2002, F&R sampled surface water, groundwater, and sediment from both lakes to determine if there was any contamination. Water in both lakes showed minimal signs of lead and arsenic. These residues were too low to be considered a problem. However, the sediment in the fishing lake had slightly elevated levels of lead (74 ppm) and arsenic (18 ppm). It was suggested that further investigation was needed to determine if there was potential bioaccumulation of pesticides in the fish.

By looking at a United States Geological Survey (USGS) topographical map, it

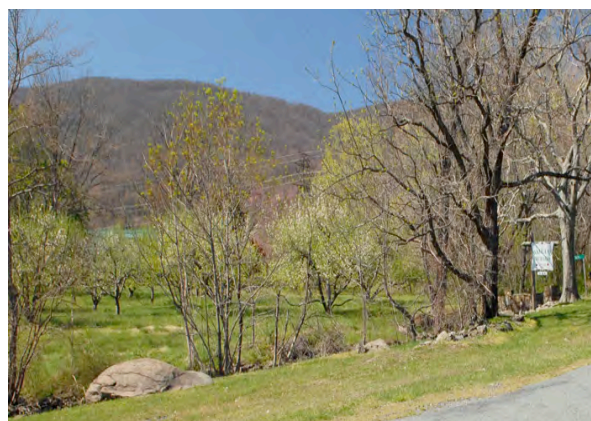


Figure 17. Apple trees growing in and around Mint Springs Park (Crozet, VA), 2002.

was evident that an intermittent channel and incidental runoff from the mountain ran through the former apple orchard and into the fishing lake. Years of well-documented use of pesticides in the orchard are believed to have caused the slightly elevated levels in the lake sediment. Soil analysis results showing high levels of lead and arsenic suggested heavy use of lead arsenate. For some of the sampling sites, trees were still standing and individual samples were taken around the drip line. However, for areas where trees were missing, samples were taken at random and mixed from each area. The samples indicate an overall contamination, not just hot spots located intermittently. Mint Springs Park showed soil contamination with lead levels as high as 494 ppm and arsenic levels as high as 130 ppm (Ibid).

North Carolina

At almost 500 acres, Barber Orchard was once a thriving commercial apple orchard located in mountainous Haywood County, 3 miles west of Waynesville. Production in the orchard began around 1908, but with a declining market, it was sold by the Barber family in 1977. Several purchasers tried and failed at the apple business because of foreign competition and a decline in demand in the 1980s. In 1988, the farm declared bankruptcy and was sold to a developer and divided into 9 to 40 acre parcels. Homes were built on the site between 1993 and 1994, but most of the property remained undeveloped. By 1999, there were 90 homes and about 200 undeveloped lots in the subdivision.

In January 1999, a local resident heard a rumor of birth defects in children born to women living in the former orchard. On the advice of a former orchard

worker, the resident contacted the North Carolina Department of Environment and Natural Resources Division of Water Quality (DWQ) to test the well water for pesticide contamination. DWQ detected high levels of benzene hexachloride (BHC) and notified the Haywood County Health Department and the state toxicologist (North Carolina Department of Health and Human Services, 1999a; 1999b). The state toxicologist recommended that the resident stop drinking well water and drink bottled water instead. This recommendation prompted a large-scale sampling effort by DWQ. Of 88 wells sampled, 34 contained BHC concentrations above the state standard of 0.019 ppb. The North Carolina Department of Agriculture and Consumer Services also conducted soil sampling on 16 properties. High concentrations of arsenic and lead were found that exceeded the North Carolina Inactive Hazardous Sites Program's soil remediation goals (400 ppm for lead, 4.4 ppm for arsenic). These results were forwarded to EPA. More sampling began in June 1999. Of the 55 properties sampled, EPA found arsenic levels in the soil above 40 ppm at 25 locations. Arsenic and lead were not detected in the groundwater samples. Due to health risks, EPA initiated an emergency removal action at Barber Orchard in 1999. (A removal is a short-term cleanup intended to stabilize or restore a site that poses an imminent and substantial threat to human health or the environment.) See Table 3 for the initial soil sampling results for arsenic and lead and the cleanup levels for the site.

Emergency cleanup of 28 residential lots began in September 1999. The cleanup involved soil excavation (Fig.

Table 3. Arsenic and lead soil concentrations and cleanup levels at Barber Orchard, NC, 2003.

Contaminant	Concentration Range (ppm)	Frequency of Detection/Total	EPA Reg. 4 Emergency Response Level (ppm)	EPA Remedial Level (ppm)	NC Soil Remediation Goal (ppm)
Arsenic	*ND – 1,340	210/273	40	20	4.4
Lead	*ND – 3,090	273/273	400	400	400

*ND = not detected.

18) where arsenic levels exceeded EPA's short-term exposure cleanup criteria. An estimated 31,500 tons of contaminated soil were removed from the lots and transported to EPA-approved landfills in Buford, GA, and Johnson City, TN. The lots were then replaced with clean fill. Remediation of the properties was completed in August 2000. The soil excavation, at an estimated cost \$4 million (USEPA, 2000) was completed under the authority and direction of EPA's Emergency Response and Removal Branch. The site was officially listed on the NPL on September 13, 2001 (McLeod, 2001)

According to EPA, the contamination may have occurred because of long-term pesticide application, spills, leaks, and improper disposal of pesticides and pesticide containers. A central mixing area, consisting of two 500-gallon concrete tanks, was identified in the former orchard. From the mixing area, pesticides were transported to other areas of the orchard with high-capacity pumps through an elaborate underground pipeline system buried about one foot below the ground. Trees were then sprayed with pesticides throughout the orchard by connecting a hose and nozzle to the pipeline. Pipes reportedly would freeze in winter, causing them to rupture and leak. During the initial sampling in 1999, pipes from the underground system were

observed on the lots and in many areas were protruding from the ground. Sediment samples were collected from one of the leaky pipes, and extremely high levels of arsenic (2,460 ppm) and lead (6,970 ppm) were discovered.

EPA released a public health assessment of Barber Orchard on July 2, 2002. According to the Agency for Toxic Substances and Disease Registry (ATSDR, 2002), Barber Orchard was considered a "past public health hazard." Current exposure to site contaminants is not likely to cause adverse health effects. However, the site could pose a problem in the future if undeveloped lots are not characterized properly and remediated, if necessary, before new residential construction begins. In September 2004, EPA issued a "Record of Decision" stating that they intended to clean up the site by removing contaminated soil and disposing of materials properly in offsite landfills. Once the cleanup was



Figure 18. A bulldozer removes soil from a yard at Barber Orchard in Haywood County, North Carolina in 1999 (Raleigh News & Observer)

completed, building could begin. The grant provided by EPA to build a new waterline stipulated that property owners could not build on the land until arsenic levels in the soil were below 40 ppm (USEPA, 2006).

Researchers at Western Carolina University (WCU) are conducting studies at Barber Orchard with private funds. Given the expensive and disruptive nature of bulldozing to remove contaminated soil, WCU is exploring other ways to remediate the soil in the undeveloped lots of the subdivision. In field and greenhouse experiments, WCU studied the use of corn, pea plants, and Indian mustard (*Brassica juncea*) for phytoremediation of lead. The brake fern, *Pteris vittata* (Fig. 19), was also studied as a way to draw out arsenic from the soil. (See the section on "Remediation" for more information on phytoremediation and the brake fern.) A nontoxic chemical agent, EDTA, was applied first to "loosen" the metals from the surrounding sand and clay, making them more available to the plants (Quillin, 2000). Fern shoots, grown in contaminated soil, had arsenic concentrations up to 20 times higher than the soil arsenic concentrations found in the soil (Salido et al., 2003).

Results indicated that increasing soil pH might also improve arsenic removal. However, increasing the pH of the soil would also reduce the removal of lead. It takes about eight years to reduce soil arsenic to levels below 40 ppm. EDTA was found to improve lead extraction. This process is cost-effective but slower and could be a suitable alternative, especially for soils that do not require immediate remediation (Ibid). However, it is also important to realize that adding EDTA to soil to "loosen" the lead increases its bioavailability, which could

Remediation

The complexity of soils and the presence of multiple contaminants make most remediation efforts arduous and costly (Sparks, 1995). Two types of methods are being used to decontaminate polluted soil sites, in situ and non-in situ.

In situ

In situ methods are used at the contamination site. This minimizes exposure to the contaminant, and the soil does not need to be excavated. These remediation techniques are biodegradation and phytoremediation.

Biodegradation involves using naturally occurring microorganisms that help degrade soil contaminants. A number of factors could affect biodegradation of soil pollutants, including pH, temperature, moisture, indigenous microbes, and the availability of nutrients (Ibid). Microbes may be effective in degrading one pollutant but not another.

Phytoremediation is the use of plants to clean up contaminated environments such as soil, water, or sediments (Arthur and Coats, 1998; Commis, 1995). Plants, known as hyperaccumulators, take up toxic metals through their roots and transport them to stems or leaves where they can be removed by harvesting. The process of using these plants to remediate soils is termed, "phytoextraction" (Arthur and Coats, 1998). One possibility for phytoremediation is the brake fern, *Pteris vittata* (Fig. 19).

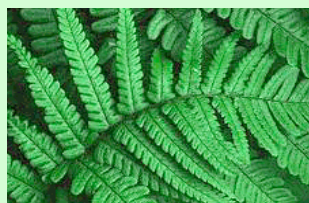


Figure 19. Brake fern, *Pteris vittata*, sold by Edenspace (2006) as the Edenfern™ is used to remove arsenic from contaminated soil.

Researchers discovered that the fern can accumulate up to 10 times the concentration of arsenic found in the soil (Ma et al, 2001). The brake fern was found flourishing at a site in Florida, which had been contaminated with chromated copper arsenate (CCA). The fern is one of very few plants that can accumulate arsenic and may be a promising answer to removing arsenic from contaminated soil.

It is very important to be familiar with the chemistry of the pollutant. Depending on soil type (ex. clay, sandy loam, silt loam), pH; soil moisture; and temperature, arsenic and lead may or may not be biologically available. To make these metals more available to the plants, chelating agents may be added, pH may be lowered, and calcium or other minerals may be added. Of course, increasing the availability of the metals also increases the likelihood that arsenic and lead to leach will leach into the groundwater or move off site. However, there are few chemicals that will allow both arsenic and lead to become biologically available. The cost of using plants to decontaminate these polluted soils could be less than one-tenth the price of either digging up and transporting the soil to a hazardous waste landfill or making it into concrete (Comis, 1995). Unfortunately, the process time can take many years.

Non-In Situ

Non-in situ methods of remediation involve removing the soil from the contamination site. The soil is then either treated on site or transported to another location, such as a landfill, and treated. There are obvious concerns about exposure to the contaminants during the moving and hauling process (Sparks, 1995). Non-in situ methods are much quicker but tend to be extremely expensive.

increase the likelihood of lead leaching through the soil or being moved off site. A second study at WCU is looking at the possible uptake of lead and arsenic by garden vegetables and the implications of consumer safety. (Washington State Area-Wide Soil Contamination Task Force. 2003b)

Washington

Washington State has prepared itself for the consequences of using lead arsenate in orchards that at one time covered thousands of acres. In 1989, the Model Toxics Control Act (MTCA) was established to raise funds to clean up hazardous waste sites and prevent future contamination. The Washington State Department of Ecology implements the regulations using money raised from a tax on toxic substances. MTCA instructs the state Department of Ecology to introduce cleanup standards for sites contaminated by toxic substances, including lead, arsenic, and DDT (Warner, 1996a). MTCA's cleanup action levels in soil are 22 ppm for arsenic, 250 ppm for lead, and 1ppm for DDT (Warner, 1996b). Topsoil removal was the main strategy for remediation.

In 2002, the Washington State Departments of Agriculture, Ecology, and Health and the Office of Community Trade and Economic Development chartered a group known as the Area-Wide Soil Contamination Task Force (2003a). This task force consisted of representatives from local government, agriculture, environmental organizations, business/development, financial institutions, and education/schools as well as elected officials. The group was asked to provide findings and to recommend steps to better address area-wide soil contamination problems. (Area-wide soil

contamination is low-level contamination that is dispersed over a large geographic area, ranging from several hundred acres to many square miles (Washington State Area-Wide Soil Contamination Task Force, 2002).) Due in part to pesticide use over the years, many areas of Washington State have low to moderate levels of arsenic and lead. In general, arsenic levels up to 100 ppm and lead levels between 500 and 700 ppm are considered "low to moderate" levels. The levels of arsenic and lead generally associated with area-wide soil contamination are not high enough to trigger an emergency response action under the state cleanup regulations.

With population growth and changes in land use, many such areas have been developed into schools, parks, and residential neighborhoods.

Approximately 188,000 acres of orchard land (apple and pear) may be affected by past lead arsenate use. The estimated total acreage in Washington affected by soil contamination, including smelters and orchards, is 677,000 acres (Washington State Area-Wide Soil Contamination Task Force, 2003a).

New Jersey

Burlington Heights, located in Burlington Township in Burlington County, NJ, is a housing development situated on a former orchard. Burlington County, in south-central New Jersey, is historically one of New Jersey's leading agricultural counties. According to Murphy and Aucott (1998) the total amount of arsenic applied in Burlington County from 1900 to 1980 was about 2 million pounds. Soil was sampled in 1995 by a developer who wanted to develop the rest of the orchard. Arsenic levels were found that exceeded the New Jersey

Department of Environmental Protection (NJDEP) soil cleanup criterion of 20 ppm. Based on the sampling results, NJDEP realized that some residences might be contaminated by pesticides. In 1996, NJDEP conducted emergency soil removal from residential yards at Burlington Heights. This process included sampling yards, removing contaminated soils, and replacing the soil with clean fill and sod. NJDEP paid \$500,000 for public outreach, sampling, soil removal, and soil and sod replacement at Burlington Heights.

After remediating Burlington Heights, NJDEP was required to recommend strategies to address historical pesticide contamination throughout the state. NJDEP formed the Historic Pesticide Contamination Task Force in 1997 to help the department “identify technically and economically viable alternative strategies that will be protective of human health and the environment for sites with contamination due to historical use of pesticides” (NJDEP, 1999). The task force made recommendations to help homeowners whose properties might be contaminated by past pesticide use.

Similar to the task force in Washington, the New Jersey task force consists of nine members representing several interest groups, including agriculture, the environment, real estate development, banking, local government, and research institutions. Primary pesticides of concern include arsenic, lead, DDT, and aldrin. These pesticides were selected based on several factors. These include their extensive agricultural use over time, their persistence in the environment, and their presence at elevated concentrations at various locations in New Jersey. Murphy and Aucott (1998)

calculated cumulative lead and arsenic applied in New Jersey from 1900 to 1950, based on national consumption and crop recommendations, as 48 to 200 million pounds in apples alone. For arsenic, NJDEP set the cleanup criterion at 20 ppm; for lead, 400 ppm.

With the publicity surrounding the Burlington Heights cleanup and the formation of the task force, other townships began to look at their history and wonder whether they, too, had pesticide-contaminated property. Mount Laurel Township, also located in Burlington County, is another township that discovered high arsenic levels in the soil due to naturally occurring arsenic and to contamination from historical pesticide use. Mount Laurel was one of the first townships to enact an ordinance requiring soil testing and property cleanup before new development -- both residential and non-residential -- in the township. Testing and remediation requirements, however, apply ONLY to properties that were formerly agricultural use or part of an orchard (Washington State Area-Wide Soil Contamination Task Force, 2003c).

The Historic Pesticide Contamination Task Force has also had an impact on lending institutions in New Jersey. Lenders have adopted requirements for environmental site assessments as a condition for granting loans to develop agricultural properties. In fact, according to the New Jersey Bankers Association, some banks in New Jersey no longer lend to farms (Ibid). Banks often ask developers or property owners to provide a “No Further Action Letter” from NJDEP giving proof of cleanup or arrange for an environmental site assessment verifying that no cleanup is necessary. By requiring this letter, banks have identified contamination at

agricultural sites undergoing development.

Wisconsin

The Wisconsin Department of Agriculture, Trade, and Consumer Protection (DATCP) oversees the cleanup of sites with lead and arsenic contamination from historical mixing, loading, and application of pesticides (Ibid, 2003d). Wisconsin's Lead Arsenate Program, now housed within DATCP, is developing and implementing a proactive approach to prevent contact with contaminated soils at lead arsenate sites. The Lead Arsenate Program is largely funded by an EPA grant to identify old orchard sites, develop a GIS database and Internet map server, and conduct public education and outreach.

After the land use has changed from agricultural to nonagricultural, properties contaminated with arsenic and lead are typically cleaned up through a state voluntary program. In 1994, DATCP (DATCP, 2007) set guidelines for site assessments to help property owners and developers identify areas of potential contamination. The guidelines are based on background levels (naturally occurring sites), pesticide-use level (orchards and other sites), and priority levels (spill, mixing, and loading sites). The cleanup action levels stated by the Wisconsin Lead Arsenate Program for priority-level pesticide sites are 100 ppm for arsenic and 400 ppm for lead.

Summary and Conclusions

Standards for arsenic and lead in the environment

In the United States, very few uniform standards exist for evaluating toxic action levels for chemicals in soil. What concentration levels are considered safe

in soil for a specific chemical? A specific chemical is toxic to a certain extent for different species. However, soil types and environmental conditions also affect the toxicity of the chemical. Eventually, uniform soil quality criteria may need to be developed for regulatory purposes, similar to water quality standards. Bioremediation of soils would be considerably simpler with standards that would provide more straightforward and consistent guidelines to evaluate toxic levels in soils (Arthur and Coats, 1998).

In the North Carolina reports of Barber Orchard, action levels for cleanup of that site were set at 200 ppm for lead and 40 ppm for arsenic. These numbers were determined by using several factors but were mainly based on the background levels found in the soil. Because lead and arsenic are naturally occurring in soil, a certain amount of these elements will exist as the "background level."

In Virginia, background levels for arsenic found in soil ranged from 2 to 10 ppm; for lead, 13 to 39 ppm. These numbers were based on the lowest levels of arsenic and lead found on the sites sampled by Schooley (2006). To date, no studies in Virginia have pinpointed the exact background levels in the commonwealth. A study from Virginia Tech (Jones and Thomas, 1999) looked at background levels in three areas of Virginia; the Coastal, Piedmont, and Valley and Ridge areas. However, the study was incomplete. It listed background levels for only a few soil series. Schooley (2006) took samples from the Valley and Ridge area of Virginia. The two soil series listed by Jones and Thomas (1999) for the Valley and Ridge area are Carbo and Frederick. According to their study, background levels for arsenic in Carbo soils (A horizon) are approximately 25

ppm, while lead levels are approximately 70 ppm. Background arsenic levels for Frederick soils (A horizon) are about 30 ppm, while lead levels are about 60 ppm.

Comparing lead and arsenic levels found at two Virginia orchard sites, Mint Springs Recreational Park and Snead Farm, showed results well above the background levels documented by Schooley and by Jones and Thomas. These levels were also higher than the predetermined standards for cleanup set by North Carolina and EPA.

Mint Springs Park showed soil contamination with lead levels as high as 494 ppm and arsenic levels as high as 130 ppm. Snead Farm had levels ranging from moderate to high levels of contamination with lead as high as 774 ppm and arsenic as high as 286 ppm.

Both orchards suggest heavy use of lead arsenate during the years of commercial production. There is a correlation with the high levels of arsenic and lead and those sites sampled that were confirmed commercial orchards. The data show that orchards located on small farms, not for commercial production, had low to moderate levels of arsenic and lead in the soil, indicating that smaller amounts of lead arsenate were applied to these sites.

The highest levels of arsenic and lead were found at Snead Farm. This former commercial orchard is very similar to Barber Orchard. Both orchards used an underground piping system to deliver water throughout the orchard. Barber Orchard also used this system to pipe pesticides through the orchard, possibly contributing to groundwater contamination. There is no proof,

however, that such practices were in place at Snead Farm.

In all cases, uniform soil standards for lead and arsenic would provide a valuable guideline to determine if remedial action is needed to clean up contaminated areas.

Practical Solutions and Recommendations for Landowners

Landowners who find themselves with soils contaminated with either high levels of lead or arsenic have several options. Unless large quantities of grading are required, soil removal is usually not practical. Phytoremediation is probably not realistic either. Here are some practical alternatives:

1. Do nothing. Homeowners can deal with soil residues by leaving the soil undisturbed or avoid contact with the soil altogether.
2. If planting vegetable gardens or flowerbeds, raise the beds by bringing in clean topsoil. These beds should be lined with heavy plastic to prevent mixing new soil with contaminated soils. This can occur very easily if the beds are mixed or cultivated during planting. Avoid direct contact of plants with contaminated soils by covering the soils with plastic mulch, organic mulch, or turfgrass. Always wash food plants harvested from these sites thoroughly before cooking to be safe. Vertical gardening techniques such as pole or trellis growing may also lessen the contact between fruit and soil.
3. Avoid using phosphate fertilizers on any soil that may have high levels of arsenic. Phosphate will compete with the arsenic in the

soil, allowing arsenic to become more bioavailable. This could make arsenic more likely to be taken up by plants, to leach, or to move offsite.

4. Monitor children's activity to help avoid unnecessary contact with soil. Washing after soil contact will minimize the chance of ingesting lead and arsenic residues. Growing grass is a sufficient barrier for family safety.

An excellent reference for landowners regarding gardening on lead and arsenic contaminated soils was published by Washington State University (Peryea, 2001).

Further Research Needed

The Virginia study (Schooley, 2006) provided a brief history of the use of lead arsenate in commercial apple orchards and a discussion of problems that can result from use. More extensive studies are needed including research on:

- Movement offsite (water contamination) – Contamination can spread through surface runoff or through subsurface water systems. For example, much of Virginia that was previously covered with commercial orchards has Karst topography. Karst strata can provide a direct conduit for surface runoff to move to groundwater. Surface runoff can be a serious concern because of the availability of the contaminants in the topsoil. The effects of runoff and strata on movement of lead and arsenic into groundwater needs to be examined.

- Leaching (water contamination) – The full implication of leaching needs further examination. The effect of other chemicals being applied to contaminated soil may cause lead arsenate to leach further into the soil and contaminate groundwater. Since many communities affected by lead arsenate use well systems, a thorough hydrologic analysis is needed to estimate the likelihood that of lead and arsenic will contaminate groundwater through leaching.
- Uptake in plant materials – The possible uptake of arsenic and lead by leaves and grass clippings requires further examination. Leaves and grass clippings are often collected by municipalities and redistributed as compost. If this compost is contaminated by lead and arsenic from one or two sites, it could easily be redistributed to previously uncontaminated sites thus compounding the problem. Uptake of lead and arsenic into edible plants could potentially impact food safety. Further study of lead and arsenic uptake by a broad range of species is necessary to determine the potential for food and feed contamination.
- Bioindicators – Fauna, grasses, and other plants may serve as indicators of the presence of arsenic and lead in soils. If a specific plant or other organism is present in a known contaminated area, it could be studied to determine if it has any properties that allow it to flourish in high levels of arsenic or lead. The

converse may also be true: plants that could be grown for aesthetic or productive benefits may not grow in highly contaminated soils. Other organisms and the population or diversity of these organisms on contaminated sites may be completely altered compared to uncontaminated sites. If this were true, these bioindicators could show the effects high levels of lead and arsenic may have on the ecosystem. They may even provide evidence as to the level of contamination in an area. These indicators might also be used to spot areas where contamination has occurred without having to do thorough soil sampling.

- Snead Farm (Virginia) – With high levels of lead and arsenic found at this site as well as it's similarities to Barber Orchard, more research is needed on the Snead farm. It should be determined if chemigation was used on the site, if there is groundwater contamination, and what effects these high levels of arsenic and lead have on the ecosystem. The results of such a study may be highly beneficial to other areas affected by lead and arsenic contamination.

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