

## Impact of Land Disturbance on the Fate of Arsenical Pesticides

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Elizabeth S. Winston, Roxanne Karimi, Carol L. Folt, and Celia Y. Chen

## ABSTRACT

Increasing development of historic farmlands raises questions regarding the fate of pesticides applied when these lands were in cultivation. We quantified As and Pb budgets in field soils in two orchards where arsenical pesticides were applied in the early 20th century and a third uncontaminated control field. Sequential extractions and X-ray analyses also were used to determine mineral phases. In addition, we measured metal loads in drainages adjacent to the fields and in two common macroinvertebrate taxa within the wetland at the outlet of the drainages. We find that the applied As and Pb have undergone little vertical redistribution; concentrations of As and Pb in the top 25 cm of contaminated orchard soils are higher than in the uncontaminated control field. However, none of the applied lead arsenate ( $PbH_2AsO_4$ ) remains in its original mineral phase. Instead, the metals are more primarily adsorbed onto fine silt and clay-sized amorphous oxides and organic matter. Further, physical erosion associated with tilling and replanting appears to have mobilized the fine-particulate-bound As and Pb in one orchard. The remobilized metals are found in sediments in the stream channel draining the tilled orchard. It is unclear if the As and Pb transported sediments are biologically active; average macroinvertebrate metal burdens in the wetland are not elevated above those observed elsewhere in the region. However, little of the mobilized metals may have reached the wetland. These results demonstrate that land use change can significantly impact the retention of arsenical pesticides.

**D**URING THE 1930S AND 1940S an average of more than 40 Mg of arsenical pesticides were applied annually to U.S. farmland (Murphy and Aucott, 1998). Growing recognition of the health risks (U.S. Public Health Service, 2000; Karagas et al., 2002; Smith et al., 1992) associated with long-term exposure to the low levels ( $1\text{--}50\text{ }\mu\text{g L}^{-1}$ ) of As in drinking water has focused attention on the fate of these pesticides.

The use of arsenical pesticides was particularly prevalent in apple (*Malus sylvestris* Mill.) orchards before the invention of 1,1,1-trichloro-2,2-bis(4-chlorophenyl)ethane (DDT) in 1947 (Welch et al., 2000). Many studies have concluded that the redistribution of surface-applied Pb and As within orchard soils is limited; decades after the application of arsenical pesticides, the highest concentrations of Pb and As generally remain in the top 25 cm of contaminated soils (Aron et al., 1980; Benson, 1976; Jones and Hatch, 1937; Peryea and Creger, 1994; Veneman et al., 1983). Arsenic is somewhat more mobile

than Pb (Elfvig et al., 1994; Merry et al., 1983; Peryea and Creger, 1994), particularly in sandy soils (Veneman et al., 1983). In some cases the mobility of As may be significantly increased by the application of phosphate-rich fertilizers (Johnson and Barnard, 1979; Murphy and Aucott, 1998). Phosphate and As exhibit similar physiochemical behavior in soils and compete directly for specific adsorption sites on soil particles (Woolson et al., 1973).

The limited redistribution of As and Pb implies that these metals are strongly retained in the soil. However, if the metals are retained on fine particles, then the retention of the As and Pb may be impacted by physical erosion (Cooper and Gillespie, 2001), particularly if the upper soil is physically disturbed such as during tilling or land development (Wu et al., 2004). More than 2500 km<sup>2</sup> of U.S. farmland are developed each year (USDA, 2003b), suggesting the possibility that land development might mobilize significant amounts of Pb and As in lands where arsenical pesticides were used.

To assess the potential for land disturbance to mobilize arsenical pesticides, we compared the Pb and As budgets of two orchards in southern New Hampshire having similar historical applications of arsenical pesticides during the early 1900s. Trees more than 80 yr old continue to grow in the first orchard, while the original trees planted in the second orchard were removed when the field was tilled and replanted in 1992. A third orchard where lead arsenate was never applied provided a control for our study. We also determined the masses of As and Pb in sediments within streams that drain each field; much of any soil that eroded from these fields would likely be deposited in these channels. Finally, to assess the bioavailability of any Pb and As mobilized by the tilling, we measured metal body burdens in macroinvertebrates inhabiting the wetland ecosystem at the outlet of the stream draining the tilled field.

## Site Description

The field site for this study is a commercial orchard located in southern New Hampshire. The orchard is still in production and has been owned and managed by the same family for >80 yr. Active orcharding at the site is indicated on the 1953 U.S. Geological Survey topographic map and the New Hampshire State Blister Rust survey maps from 1936, 1938, and 1960.

Average monthly precipitation at the site is  $8.4\text{ cm mo}^{-1}$  and is approximately uniformly distributed throughout the year. The site receives ~25% of its precipitation in the form of snow. About half the total annual precipitation in this region is lost to evapotranspiration (Randall, 1996). Orchard soils are well drained, shallow to medium depth, gravelly to fine, sandy

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**Abbreviations:** ICP-MS, inductively coupled plasma mass spectrometer; RUSLE, Revised Universal Soil Loss Equation; SEM, scanning electron microscopy; XRD, X-ray diffraction.

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metals has been concerned in the past with effects that produced clinical signs and symptoms However this view of metal  
toxicology has expanded in recent years due principally to two advances There has been a considerable increase in our  
knowledge of the biochemical effects of metals In addition biomarkers of toxicity can now be recognized that identify toxicity

at levels of exposure that do not produce overt clinical effects Thus the toxicology of metals is now focused on nonclinical events that reflect adverse health effects This new awareness has produced the challenge of determining the lowest adverse level of exposure With increasing analytical sensitivity and methodologies to detect small changes at the molecular level the lowest level of exposure of some toxic metals like lead is very small Indeed for metals in which there is no biologic requirement it may be questioned whether there is a level of exposure that does not produce some degree of toxicity For essential metals the question is being asked as to the levels at which exposure exceeds biologic requirements and excess exposure becomes toxic The appropriateness of health decisions and the formation of public policy are dependent on the availability of current scientific information that addresses these questions The information in this volume is intended to be a resource for this purpose as well as a reference for students of toxicology and other health professionals

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