

Lead from Firing Range and the Potential to Contaminate Drinking Water Supply

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Introduction

A Medical Health Officer for Vancouver Island Health Authority contacted the BC Centre for Disease Control (BCCDC) to request information concerning the potential for water contamination posed by lead from a firing range in close proximity to a drinking water supply inlet. The Courtenay and District Fish and Game Protective Association owns and manages the firing range and, in addition to current operations, is looking to expand.

To look at the potential for contamination, the following report presents literature concerning speciation and mobility of lead from lead shot in soils and water, originating from firing ranges.

Literature Search

An initial literature search of scholarly (peer-reviewed) journal articles was conducted for lead shot from firing ranges and contamination of soil and drinking water (or water) and exposure/health risk (human and/or ecological). Scientific literature was scoped, using the EBSCO and Web of Science databases. No date limitations were set; the search included all years. English language articles were the primary focus but did not limit the search, which also included some popular media. Search terms/strings included (firing range) AND lead or copper or metal. No additional terms were used in order to provide a wide net of papers. The results were manually reviewed and 63 citations were saved for initial review.

Results were further refined through abstract reviews, for direct relevance to outdoor firing ranges and lead mobility in soil and water. Since health effects and indoor firing ranges were not part of the main focus, this information was limited. Bibliographies of selected papers were reviewed for additional sources of information. Google search engine was used to locate regulatory, guideline, report, and governmental agency information pertaining to lead and small arms firing ranges.

Background

Lead in the environment can be naturally occurring and/or from anthropogenic sources. Naturally occurring lead in soils is normally less than 50 parts per million (ppm), with Canadian estimates ranging from 12-25 ppm,¹ but levels can increase due to lead introduced through

industrial activities or other anthropogenic sources.² Anthropogenic sources of lead from industrial processes (e.g., smelting or combustion) can cause a diffuse spread of lead; lead from shot, introduced by firing ranges, generally creates point sources of lead in and around the range environment. Point sources from shooting ranges can cause lead levels in the environment to be several orders of magnitude higher than normal.³ Lead bullets and fragments at firing ranges are usually contained within a relatively small, well-defined area, or volume of sand and/or soil but micro particles can become airborne and be deposited over a larger area. Skeet and trap ranges can have shot fall zones that cover 10–50 acres or more, depending upon the layout of the range.⁴

Lead shot contains lead (97%), antimony (2%), arsenic (0.5%), and sometimes nickel (0.5%); lead bullets are composed of lead (90-99%), antimony (1-10.5%), and copper (0.1%). According to the Interstate Technology and Regulatory Council (ITRC), Small Arms Firing Range Team's *Characterization and Remediation of Soils at Closed Small Arms Firing Ranges* states that small arms firing ranges may contain lead, antimony, copper, zinc, arsenic, and polycyclic aromatic hydrocarbons (PAHs) from non-exploding (non-energetic) bullets and fragments, bullet jackets, and related sporting material (e.g., clay targets), but lead is the "primary risk driver."

Lead can be toxic to soil fauna, vascular plants and small mammals. Many studies have looked at the toxicity of lead in the environment; the highest risks are predicted for small mammals and birds ingesting lead shot while feeding or using as grit to aid in digestion.⁵⁻⁸ In humans, lead exposure can affect the functioning of kidney, bone, the central nervous system, and have other health impacts. In children, lead exposure has been associated with developmental delays and reduced IQ. Plumbism, or lead poisoning, has long been associated with occupational lead exposure, including inhalation of airborne lead particles at a firing range.⁹⁻¹⁷

Other routes of exposure can be through ingesting lead contaminated soil, sediment, food or drinking lead-contaminated groundwater. The ITRC guidelines for small arms firing ranges note that "lead dissolution and migration of lead to groundwater or through aerially (windblown) or hydraulically (erosion and deposition) dispersed particles can cause exposure and result in elevated levels of lead in the blood of humans and wildlife..."³

However, knowing the concentration of lead in the soil alone does not provide sufficient information concerning the fate and transport of lead in the environment. Lead solubility is dependant on speciation, soil, and water conditions. The following sections provide information from literature that focuses on lead speciation and mobility in soil and water.

Lead Speciation and Mobility

According to the U.S. Environmental Protection Agency (US EPA) guidance on health risk and shooting ranges, it is important to distinguish between high and low velocity ranges. Soil at high velocity ranges usually contains very fine particles of lead, in addition to bullets and bullet fragments. Soil at low velocity ranges tends to contain whole and partially decomposed lead pellets.⁴ Estimates for lead at shooting ranges show that 90% of lead is from intact shot, 5% is in corrosion products around lead shot, and 5% is in corrosion products in soil and fixed by soil.¹⁸

When interpreting analytical results, the presence of a bullet, bullet fragment or lead shot in a soil sample will result in a very high measurement of lead concentration which "may not yield an accurate prediction of risk for current exposure scenarios (but may be appropriate for future exposure scenarios)." Large lead fragments have shown to be relatively stable; once lead shot is deposited in the fall zone, it can remain relatively immobile. However, micro particles of lead from oxidation and other processes in the soil can become airborne and mobilize away from the fall zone.

The rate of weathering and oxidation is highly variable and site specific. Important variables governing speciation and solubility are pH and oxidation- reduction potential (redox or Eh). Metallic lead is stable in a very low redox potential condition, but typical soil conditions can have high level of redox potential, depending on composition.¹⁹

In general, lead exhibits its greatest solubility in acidic (pH < 4) solutions. Under acidic conditions, elemental lead will oxidize, releasing a hydrated cation, Pb⁺². Under alkaline conditions, elemental lead will oxidize under most circumstances to form a lead hydroxide complex. This influences mobility. Lead that exists in the dissolved state can be sorbed to negatively charged clay particle surfaces. According to an Army Corp of Engineers report (Larson et al. 2007), erosion and surface water transport of contaminated clays can be a major source of lead mobility in the environment.²⁰ This transport can be either attenuated or increased depending upon the mobility of the soil particles.²⁰

According to the US EPA Best Management Plan for firing ranges, there are five factors which most influence lead dissolution²¹:

- Annual precipitation rate
 High precipitation rates increase weathering rates; with increased contact time, there is a higher potential for lead to migrate off site.
- pH of rain and surface water and mineral content of soil
 The acidity of rainwater influences the dissolution of lead; however, if there are sufficient minerals such as calcium, magnesium, and iron present in the soil, the lead may precipitate out, thus reducing mobility
- Contact time
 Amount of time that acidic water is in contact with lead is a factor in the amount of lead that is dissolved.
- Soil cover
 Organic material absorbs lead and can remove it from solution.

The sections below present literature findings that concern lead speciation and mobility in soil and water.

Soil

Soil lead concentrations of greater than 10,000 mg lead/ kg soil have been reported at shooting ranges around the world, including new Zealand, U.S.A., England, Germany, and Scandinavia. Bennett et al. (2007) report that the range reported in other studies have been from near background to greater than 90,000 mg/kg at bullet pockets and back stop berms. ⁶

Canadian guidelines for lead in soil levels for residential/parkland is 140 mg/kg and 300 mg/kg, based on human health and environmental health, respectively. However, the bioavailability of lead in contaminated soils varies greatly depending upon its form in the soil. Because mobilization of lead is dependent on environmental conditions, many of the studies on lead from firing ranges are case studies concerning individual locations/ranges.

Duggan and Dhawan (2007) conducted a study of lead speciation and vertical distribution at a recreational firing range in Massachusetts and found that lead concentrations in soil showed a decreasing trend with sample depth; lead concentrations in soil decreased markedly across a 0-6 in (0-15.2 cm) depth profile. Weathered species, predominantly at the ground surface, exhibited minor vertical mobility through the soil column. Lead carbonates were the dominant non-shot form of lead present at all depths. Water-soluble lead species made up a minor fraction of the non-shot lead present in the samples. While the highest level of lead was found in the shot fall zone, the highest amount of extractable lead was found in the forest samples, which the authors attribute to the acidic conditions of the soil in that area.

In a study conducted in Florida, looking at environmental impacts of lead from a firing range, Ma et al. (2002) also noted that lead weathering products at the firing range were dependent upon soil properties and the amount of lead in the soil. Some of the weathered products entered the soil matrix and continued to transform in the soil environment, which is influenced most importantly by soil pH, buffering compounds, and organic material. Ma et al. (2002) also notes that soil with high clay, organic, and Fe and Al oxide content or high cation exchangeable capacity (CEC) tend to have low lead mobility and leachability. Additionally, soil with high pH tends to retard lead oxidation and dissolution.

According to the U.S. Army Corps of Engineers report on lead containment at active small arms firing ranges, lead entering a berm is initially present as metallic lead in either particulate or bullet form. As the berm system ages, the metallic lead undergoes corrosion processes that result in the release of lead cations. Depending on the environmental conditions within the berm, these lead ions can transport to groundwater as soluble lead, to surface water as soluble lead or sorb onto clay surfaces. It can also react with carbonates to produce lead carbonates which are soluble, associate with iron and manganese oxides and organic matter or react with compounds, such as phosphates, sulphates, and aluminum which will reduce mobility. ²⁰

Larson et al. (2007) writes that lead within un-amended sand berms could be susceptible to dissolution by acid rain, if there is a lack of adequate buffering.²⁰ The low buffering capacity of sand is a result of the sand's relative chemical inertness. This means that compared with most soils, sand has a lower capacity to reduce the mobility of soluble lead and, over time, lead ions can leach to either surface or groundwater.²⁰

Another study by Dermatas et al. (2006) investigated the effects of lead transformation products in soil on lead leachability by evaluating Toxicity Characteristic Leaching Procedure (TCLP) data from samples taken out of six different firing ranges in California and New Jersey. TCLP lead leachability levels and total lead concentrations in all of the soil samples tested were found to be above US EPA regulatory limits. However, the authors found that TCLP lead leachability levels did not always correlate well with total lead concentrations. Lead carbonate formation was found to play an important role in controlling lead leachability and precipitation was found to be an important mechanism immobilizing lead in all soils tested. Ma et al. (2002) also found that high calcium carbonate, iron, aluminum, and phosphorus contents were favourable for immobilizing lead in shooting ranges soil, resulting in low levels of lead in TCLP extracts and surface water.

Clausen et al. (2011), from the Engineer Research and Development Center for the U.S. Army, reviewed studies concerning lead migration in surface, pore, and groundwater at firing ranges. In their critical review, they write that lead accumulation in firing lines and target areas at shooting ranges was common and surface soil samples frequently exceeded regulatory limits. However, from the studies reviewed, Clausen et al. (2011) writes that subsurface migration was limited to approximately 1-3 meters and that many reported lead detections in groundwater were "sampling and/or analysis artefacts." In some cases, local soil characteristics will be adequate to retain lead within the range and maintain surface water or leachate water lead concentrations within applicable regulatory permit limits and/or action levels. Subsoil may also provide some degree of retardation. 18

Water

Lead can be transported via surface, ground, and pore water; not many studies deal with pore water since there is less monitoring of the vadose zone (layer between the ground surface and water zone) than of surface water or groundwater. The concentration of dissolved lead species in soil solution, surface water or groundwater is determined by pH, dissolution-precipitation as a function of redox environment, and sorption-desorption reactions. In a typical water body, dissolved lead forms precipitates of lead hydroxide [Pb(OH)₂], lead carbonate [PbCO₃, cerrusite], or basic lead carbonate [Pb₃(OH)₂ (CO₃)₂, hydrocerrusite].²⁰

Lead contamination can migrate from the site through weathering effects, storm water runoff, and through leachate (via groundwater) transport. Storm water runoff may erode the lead-contaminated berms and carry lead and contaminated soil particles away from the site into the surrounding environment. Rainfall intensity, ground slope, and soil type strongly influence the potential transport of lead away from the firing range. ^{20,21,24} Additionally, acid rain, which can have a pH of less than 5, can dissolve lead and transport it to nearby ground or surface waters."

Clausen et al. (2011) noted that investigations at eight target ranges reported elevated lead levels in surface water. Weathering of lead pellets resulted in elevated concentrations of waterborne lead (0.004- 0.84 mg/L) vs 0.007 mg/L at control sites. Cao et al. (2003) also reported that elevated total lead in surface water (up to 289 μ g/L) were observed in some ranges. For reference purposes, Health Canada Drinking Water Guidelines state the maximum acceptable concentration (MAC) for lead in drinking water is 0.010 mg/L or 10 μ g/L.

The runoff from Barksdale Air Force Base in Louisiana, U.S. was tested for lead levels and was found to have exceeded the maximum allowable lead levels in surface water during periods of heavy rain; the majority was insoluble lead adsorbed to suspended colloidal soil particles.²⁰

Lebare et al. (2004) conducted an investigation to characterize the extent and speciation of lead contamination in water, oil, and biota at a small arms firing and skeet range in New York.²⁷ Elevated lead concentrations were detected in soil and sediment. The authors state that lead was not confined to the soil but was leaching into the nearby streams and into the sediment.²⁷

Duggan and Dhawan (2007) concluded that weathering of shot and fragments, wind-blown fines, and transport of fines via surface run-off are possible means for lead found outside the shotfall area. Sample results found down-slope also indicate that transport from run-off is a likely source at these locations. However, they also concluded that soluble species of lead were minor and concentrations vs depth results indicated that lead vertical mobility was limited and did not suggest a potential for groundwater contamination via leaching. The presence of

lead compounds with extremely low solubility, such as lead phosphates, influences the solubility and availability of lead in the environment. When lead is sequestered within these phosphate minerals, lead has been shown to be far less soluble and its bioavailability to soil organisms has been reduced.²⁰

Conclusions

The fate and transport of lead from firing ranges is dependant upon environmental conditions that include lead speciation, influenced by pH, and the compounds present in soil and water environment. The amount and intensity of precipitation will also affect weathering and transport; the Comox Lake area receives approximately 1,485 mm of rain annually.²⁸

Research has shown that, in general, soil pH seems to have the greatest effect of any single factor on solubility or retention of lead in soils, with greater retention and lower solubility occurring at high pH.⁵ Solubility is dependent on pH, redox conditions of precipitation, soil/pore water, and/or groundwater conditions.⁷ However, solubility at lower pH can be buffered when there are sufficient amounts of aluminum, manganese, phosphate or iron oxides that are present.^{5,20} Once lead oxidizes, the solubility of lead is increased and may become mobile, however, a layer of oxidation can also impede further oxidation of the pellet.⁷

Because environmental conditions are unique to a site, a number of studies have been conducted at various firing ranges to determine the extent of contamination, bioavailability of lead, and/or fate and mobility of lead in the environment. Several studies have shown that lead from lead shot can leach into the soil, groundwater or surface water if the environmental conditions are conducive, although a recent review article from Clausen et al. (2011) refutes the mobility of lead from firing ranges in groundwater unless directly under the influence of surface water, such as a wetland.⁵

If there is potential for contaminants to migrate offsite, sampling soil, groundwater, and surface water at the range and adjacent areas would provide the best indication and extent of contamination. Assessing contamination patterns and specific characteristics of the range are not only important for risk assessments but also for management of the range. Bennett et al. (2007) recommends periodic remediation at active ranges to help reduce the risk to fauna receptors. Also, studies emphasize the importance of preventing surface water runoff from shooting ranges. Duggan et al. (2007) recommends runoff controls be implemented to prevent transport of lead in fines from the shotfall area. EPA has developed a reference for best management practices. The ITRC Technical Guideline for Environmental Management at Operating Outdoor Small Arms Firing Ranges also includes a comprehensive template for an Environmental Management Plan which could be used by the firing range (See Appendix for decision tree).

The ITRC Guideline also provides the following questions to assist range operators in collecting and assessing information necessary to understand the potential for lead and other heavy metal transport²⁴:

- Where are the property boundaries and do any rounds or shot fall beyond them?
- How is the metal distributed over the range property?

- In areas of soil, is the metal disturbed by bullet impact (e.g., bullet pockets)? Pulverized soil with high concentrations of lead and lacking vegetative cover is most susceptible to surface water transport.
- Can and has the mass of shot or bullets fired at each range been calculated on a regular basis?
- Are there hot spots (areas of concentrated lead)?
- Does wind or water erosion occur near these concentrated areas? If yes, then surface water and/or wind transport may be an issue.
- How deep is groundwater below the concentrated areas? If shallow (<10 feet, depending on soil type, pH, and mass of lead), groundwater transport may be an issue.
- Are site soils sandy and highly permeable or do the soils contain significant quantities of clay or organic matter? Sandy soils may allow vertical migration of dissolved lead, while clay or organic rich soils may adsorb the lead. Adsorbed lead may still be transported off site by surface water and wind erosion.
- Does slope of the ground surface encourage surface water runoff? Rills and gullies are an indication of erosion.
- Does the pH of the soil impact areas fall within the low solubility values (~6.0–9.0)³? If the pH is outside this range, there is higher potential for lead migration in surface water or groundwater.
- Is the impact area in a surface water body or wetland?
- How far is the impact area from flowing or non-flowing streambeds?

The BC Ministry of the Environment also recommends that operators/owners of the site implement practices that minimize the impact of activities which include developing plans to manage and minimize impact of activities on human health and the environment.²⁹ They also require that neighbouring sites be notified if contamination has or is likely to migrate offsite.

In cases where the local soils have undesirable properties, such as high acidity for range management purposes or easily eroded from the berm surfaces, partial replacement of the berm soil with a more suitable ballistic material may provide an economical alternative. Naturally occurring silicate mineral sands are relatively low cost materials that are widely available, due to their extensive use in construction. Impact berms made from sand are able to decelerate fired projectiles safely with minimal bullet fragmentation and lead-dust generation, as compared to compacted or coarse-grained soils.²⁰

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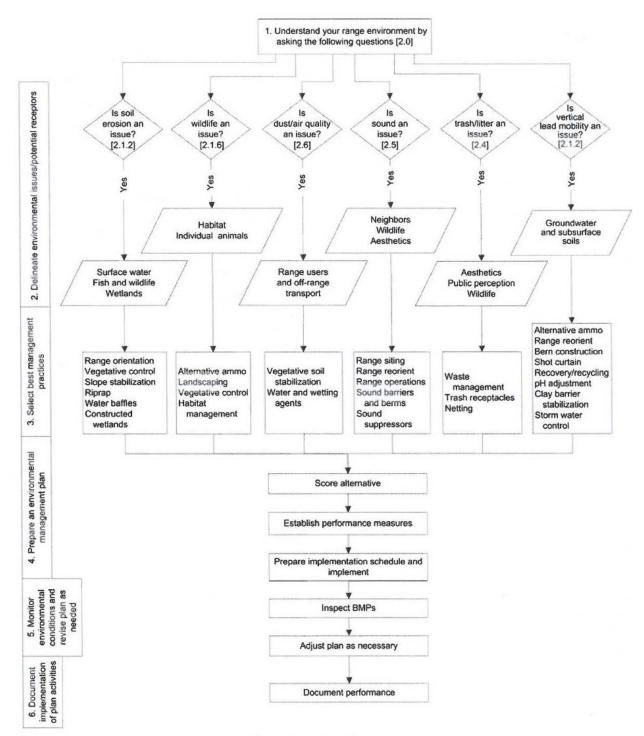
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Appendix Decision Tree for evaluating range and preventing potential surface water, groundwater, or air transport



Adapted from ITRC 2005 (numbers in tree refer to sections in the document)