ASSESSMENT OF THE POTENTIAL FOR THE USE OF LEAD AMMUNITION AT SHOOTING RANGES TO CONTAMINATE GROUNDWATER AND DRINKING WATER

FINAL REPORT TO ECHA FROM WCA

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EXECUTIVE SUMMARY

In this brief review to support ECHA's proposed restriction of lead ammunition at shooting ranges we have investigated recent evidence, predominantly from 2011 onwards, to assess the potential for lead to contaminate underlying groundwater that may be abstracted for drinking water. An initial conceptual model has been developed based on the source-pathway-receptor approach; this has been used to structure the report and then refined to highlight the characteristics of a shooting range site that could indicate a higher susceptibility to contamination of groundwater.

The upper layers of soil at shooting ranges carry a very large load of lead compared with local ambient background concentrations and concentrations of up to 30-40% Pb has been observed in surface soils. The weathering of shot and bullets to more soluble forms of lead has been well studied and shown to be greater under acidic soil conditions, and under vegetation, especially trees. Soil concentrations of lead generally decrease rapidly with depth and there are limited data that show concentrations of lead in subsurface layers and in groundwaters. Mobility of lead in soil occurs to a greater extent in conditions where processes of natural attenuation are reduced and loading of lead is relatively high. The factors that promote movement of lead from surface layers are also those that accelerate the weathering of the metallic lead shot and bullets and include acidic and organic rich soils with coarse soil texture, low iron, manganese and phosphate content.

Soil water concentrations of lead in subsurface layers have been measured at concentrations in the low mg L^{-1} range (up to 12.6 mg L^{-1} has been reported), which is several orders of magnitude above the drinking water standard for lead (10 µg L^{-1}). The concentration of lead in soil water has though been observed to rapidly decrease with depth and to date only a few studies have measured elevated lead in groundwater, with these being in near surface groundwaters under acidic soils (in areas that would generally be considered as wetlands).

Lead can be transported in soils, the vadose zone and in groundwater as either particles (e.g. small lead fragments from projectiles or lead sorbed to mobilised soil particles and organic colloids) or in solution. The connectivity between near surface soil, the vadose zone and underlying groundwaters is dependent on a combination of local factors, such as soil texture, soil chemistry and organic matter content, soil structure (particularly the existence of preferential flow pathways[1]), local geology, topography, climate, and the depth to the saturated zone. Shallow groundwater (e.g. on floodplains) is especially vulnerable to contamination but may also enhance lead mobility in the soil if water tables seasonally inundate near-surface horizons where the soil contains high lead concentrations. This issue may be exacerbated in the future in parts of Europe which are predicted to have wetter winters under climate change.

[1] Preferential flow pathways are marcopores, cracks, fissures and biopores (such as earthworm burrows and root channels) in soils that lead to the rapid transport of water and solutes.

Based on the consideration of soil chemistry and groundwater vulnerability, the key factors influencing the mobilisation of lead and its potential to migrate through the vadose zone to groundwater are:

- Acidic soil (pH <6) with relatively high organic matter content and low iron, manganese and phosphate content;
- Coarse (usually sandy) soils that allow vertical migration of dissolved or fine particulate lead;
- Preferential flow pathways, including ‘fingered flow’ in the soil matrix but more importantly macropore flow down soil cracks, plant root channels and animal burrows, along with fissured flow in the underlying vadose and saturated zone;
- Shallow depth to groundwater.

Specific groundwater vulnerability will also, clearly, be affected by the lead emission rate and historical loadings, driven by usage (e.g. number of rounds or amount of lead shot per day and the time over which the area has been used as a range). Several of the conditions enhancing the mobilisation and downward migration of lead in shooting range soils (e.g. acidic pH and preferential pathways such as tree roots) are found in forested areas, which have been identified in a number of the studies on shooting ranges reviewed for this report. Groundwater is commonly abstracted for domestic drinking water across the EU and also from private wells, these may be located in close proximity to shooting ranges and as such represent a particularly high risk because they are often taking relatively shallow groundwater and are not routinely checked for water quality.

It is difficult to estimate the prevalence and extent of groundwater vulnerability to lead contamination at shooting ranges at European, national or even regional levels because many of the contributing factors are local and difficult to predict at wider geographical scales. These local factors will always influence potential risks more than generic considerations but areas with high intrinsic vulnerability are likely to occur in all EU member states, although to differing extents. Detailed GIS analysis would be required to estimate the areas with high vulnerability which are also areas classed as aquifers. Information on the fraction of shooting ranges occurring in these high vulnerability areas would provide quantitative information on the extent of the risk and requirements for associated risk management.

Based on our review it is considered that migration of lead to groundwater is probably more likely to occur at clay target shooting ranges because the shot discharges over a wider area resulting in more widespread areas of contamination with highly elevated concentrations (estimated site loadings of up to 40 t of lead over the operational lifetime of clay target shooting ranges have been reported). Furthermore, the lead shot can become entrained in the soil surface, ultimately becoming buried due to the accumulation of organic matter, particularly in forest areas. This is in contrast to rifle and pistol ranges where shooting activity is more focussed on fixed targets using bullets that have a smaller surface area:mass ratio, although this can increase due to fragmentation. In addition, bullets and associated fragments at small arms firing ranges are generally retained in bullet traps or specific sand traps might already be in place that prevent lead from leaching to soil.

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1 INTRODUCTION

This assessment is to support ECHA's restriction proposal on the use of lead in ammunition for sports shooting, hunting and in fishing tackle (ECHA 2021), specifically to provide a technical review of recent evidence on the risks posed to groundwater by the use of lead ammunition at shooting ranges. The potential for lead in ammunition at shooting ranges to contaminate groundwater has been the subject of a considerable amount of academic interest in recent decades and there is existing guidance that states the need to protect groundwater from this source of lead (e.g. US EPA 2005; Bavarian LfU 2014; Kajander and Parri 2014).

Lead (Pb) from spent ammunition at shooting ranges is deposited on or into soils. It has been stated that shooting ranges are the second largest source of Pb pollution after the battery industry (Dinake et al. 2019). Shooting ranges tend to be characterised by high-use activities so the lead deposited leads to relatively high soil concentrations, over relatively small areas. Once deposited the original form of metallic lead is subjected to weathering processes that can result in its conversion to more soluble forms that have the potential to leach from the soil to groundwater below. This potential exposure pathway is of significant concern because a significant proportion of drinking water is abstracted from groundwater in many EU member states (EC 2021a).

The extent to which the use of lead ammunition at shooting ranges could lead to the contamination of groundwater and drinking water is uncertain and is the subject of debate in the scientific literature. Some authors suggest extremely low concentrations of lead (sub-microgram per litre) may be present in soil solution for potential transport to groundwater at shooting ranges (Clausen and Korte 2009; Dortch et al. 2013; Clausen et al. 2014). However, other studies have identified elevated concentrations of lead in soil solutions at shooting ranges and highlighted the influence of soil and vegetation factors upon potential transport down soil profiles (e.g. Selonen et al. 2012; Kelebemang et al. 2017) and preferential flow pathways in some conditions (Garrido and Helmhart 2012; Knechtenhofer et al. 2003).

1.1 Aims and objectives

The specific aims of this report are to:

a) Undertake a literature review to identify and summarise information on the potential for contamination of groundwater from the use of lead ammunition on shooting ranges; focussing on literature from 2011 onwards. This literature survey will be supplemented by a brief stakeholder survey, contacting operators of shooting ranges across Europe. This literature survey and stakeholder consultation will seek to identify existing risk management practices for the protection of groundwater and their approximate implementation costs.

b) Provide a description of the characteristics of groundwater (and specifically aquifers) in the EU that could be susceptible to lead pollution from shooting ranges, identifying the relevant exposure pathways and environmental fate and transformation processes of lead that could affect its migration to groundwater.

This includes a discussion of 'natural attenuation' and potential timescales over which contamination could occur.

c) Provide a summary of the risks to EU groundwater from lead ammunition at shooting ranges in the short, medium, and long term and attempt to differentiate the potential for risks arising from the use of lead shot versus bullets.

1.2 Conceptual site model

To provide a framework in which to undertake this review, a brief conceptual site model is utilised, adapted from the work performed by Dortch et al. (2013; see Figure 1.1). This considers the influence of sources of lead ammunition, and more critically, the influence of soil chemistry and other properties that affect the behaviour and fate of lead, specifically with regard to leaching from the soil and transfer through the vadose zone to groundwater. Not included in Figure 1.1 and beyond the scope of this report, are connections to surface waters through soil interflow, groundwater discharge and directly through surface run off.

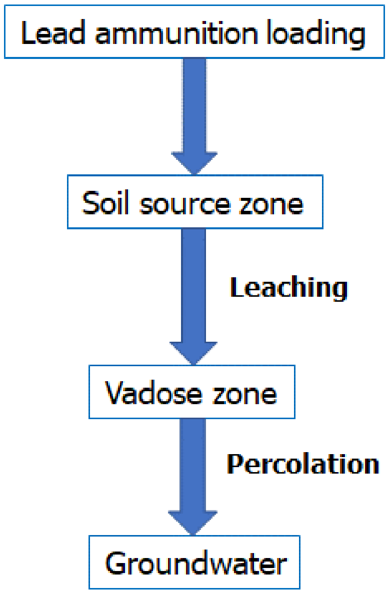

Figure 1.1 Conceptual model focused on the fate and transport of munitions constituents from firing ranges (adapted from: Dortch et al. 2013).

1.3 Report structure

After this brief introduction, Section 2 of this report details the literature and information search strategy, including the stakeholder consultation exercise. The information we have identified and screened in Section 2 is reviewed in Section 3, using the conceptual site model and source-pathway-receptor framework to structure the review, i.e. focussing on

- sources of lead to soil, soil chemistry relating to lead fate and behaviour;
- mobility of lead in the unsaturated zone and pathways to groundwater; and

- hydrogeological characteristics that make groundwater susceptible to contamination by lead from shooting ranges

In Section 4 we identify potential risk scenarios and provide a discussion on potential timeframe and assess the use of models verses monitoring programmes to predict the risks to aquifers. Mitigation measures are covered in Section 5 and finally, some overarching conclusions are provided in Section 6. Appendices at the end of the report give details of the stakeholder consultation and provide a table of recent site investigations undertaken at shooting ranges identified from the literature review.

2 LITERATURE AND INFORMATION GATHERING

This section details how the literature search strategy was undertaken to identify relevant publications on the potential for the use of lead ammunition at shooting ranges to contaminate groundwater and drinking water. It also includes the procedure followed for the stakeholder consultation, including the identification of relevant contacts.

2.1 Search strategy

At project initiation ECHA provided wca with the Annex XV report and additional references that they deemed relevant to the project aims. Literature searches were undertaken using two literature databases, SciFinder and Derwent Innovation to identify any additional published information available since 2011 (i.e. since the publication of the detailed review by Clausen et al 2011).

Two distinct search strategies were utilised due to the differing nature of the operating inputs of the two databases. Scifinder does not allow for the inputting of search strings, therefore an initial general screen was conducted using a specific phrase “Aquifer contamination from lead shot” (Table 2.1); this was selected to provide a rough determination of the available publications. Derwent Innovation allows the use of search strings, so several were prepared based on the key aspects of the project to identify potentially relevant literature.

A search was also conducted to identify any potentially relevant grey literature produced by local and national regulatory bodies. This has included searching for German language reports (based on initial discussion with the ECHA project team, it was indicated that there were potentially relevant reports produced by the Bavarian and Swiss authorities), which have been translated as required. Additionally, all references cited in the invitation to tender were also obtained and reviewed.

2.2 Screening outcomes

The search strings and phrases developed and run through SciFinder and Derwent Innovation are detailed in Table 2.1, and the number of hits per strings are detailed.

Table 2.1 Search strings and results from the literature searching

Search term	SciFinder[2]	Derwent Innovation[3]
Aquifer contamination from lead shot	0	N.A.
((7439-92-1 OR Lead) AND (Groundwater OR Aquifer OR Drinking water) AND (Ammunition OR Shooting))	N.A.	80
((7439-92-1 OR Lead) AND (Groundwater OR Aquifer OR Soil) AND (Ammunition OR Shooting))	N.A.	2675
((7439-92-1 OR Lead) AND (Drinking Water) AND (Ammunition OR Shooting)) AND (Risk assessment)	N.A.	1
((7439-92-1 OR Lead) AND (Groundwater OR Aquifer) AND (Shot OR Bullets))	N.A.	59

[2] https://www.cas.org/products/scifinder
[3] https://clarivate.com/products/derwent-innovation

Search term	SciFinder[2]	Derwent Innovation[3]
((7439-92-1 OR Lead) AND (Groundwater OR Aquifer) AND (Fate OR Transport OR Attenuation)) AND (Soil)	N.A.	717
((7439-92-1 OR Lead) AND (Groundwater OR Aquifer) AND (Firing Range OR Shooting Range))	N.A.	45
((7439-92-1 OR Lead) AND (Groundwater OR Aquifer) AND (Shotgun OR Rifle))	N.A.	15
((7439-92-1 OR Lead) AND (Groundwater OR Aquifer) AND (Shot OR Bullets)) AND (Soil)	N.A.	50

N.A.: not applicable

After conducting the searches, the results for each string were combined and the duplicates removed, resulting in 3420 unique papers. The abstracts of the publications were then reviewed to assess applicability prior to obtaining the paper for further analysis and review, which resulted in 65 scientific papers being obtained for detailed consideration.

2.3 Stakeholder consultation

Information on the potential for groundwater contamination from lead ammunition and its management at individual sites was also obtained from a targeted consultation with organisations offering outdoor sport shooting activities, including commercial recreational centres, training institutions and local and regional sports clubs. Searches for contacts were conducted using Google and survey participants were selected based on the scope of activities offered. The full list of contacted stakeholders is available in Appendix 1.

Information was sought on the extent of knowledge on lead contamination due to shooting activities and types and costs of risk management measures employed. A questionnaire was prepared and was offered in English, German, French and Polish. An example of the questionnaire (in English) is attached in Appendix 2. Selected contacts were emailed on Monday 26th June 2021 and given until Tuesday 6th July to respond to the questionnaire (deadline extended until Monday 12th July 2021 for some respondents).

Thirty-seven stakeholders from Belgium, Denmark, Finland, France, Germany, Ireland, Italy, the Netherlands, Poland, Spain and Sweden were contacted. No direct responses to the questionnaire were received from individual shooting ranges although FITASC (Federation Internationale de Tir aux Armes Sportives de Chasse) did reply to reiterate their previous positions in relation the Annex XV report and stakeholder consultation on proposed restrictions.

Although no questionnaire responses were received on the measures employed at individual sites, some European shooting clubs acknowledged the potential for groundwater contamination from lead shot on their websites and addressed their mitigation measures. As Denmark and the Netherlands have banned lead shot for recreational shooting, one shooting ground in the Netherlands noted the use of steel shot for skeet shooting[4]. An article in a recreational shooting magazine discusses the Danish ban and the user experience in switching away from lead shot[5]. A shooting ground in Germany has also voluntarily banned lead shot

[4] https://hetweideke.nl/
[5] https://www.sportingshooter.co.uk/shooting/shooting-with-steel-instead-of-lead-shot-6311654

on their grounds due to the potential for groundwater contamination and offers alternative types of shot (not specified)[6]. Additionally, a novel technology developed in Sweden, STAPP®, was identified to "capture" bullets and prevent their exposure to water[7]. The STAPP® website specifically mentions the technology as a solution to manage the leaching of lead from bullets and projectiles.

[6] https://www.schiesssportanlage-werlte.de/cms/page/posts/achtung-stopp-kein-bleischrot-14.php
[7] https://www.stapp.se/

3 LITERATURE REVIEW

This review has focussed upon the sources and characteristics of lead at shooting ranges, the transport, behaviour, and fate of lead following deposition onto and into soils and the subsequent exposure pathway from shooting ranges to groundwaters (Figure 1.1). We have attempted to identify the form and magnitude of potential risk conditions and the timeframes over which these risks may occur.

3.1 Sources

The use of ammunition at fixed shooting ranges generally results in the input of metallic lead to a relatively well-defined area of land[8]. The lead is either in the form of lead shot (pellets) from the firing of shotguns or bullets fired from pistols or rifles, with pistol and rifle firing ranges 4 times more prevalent in the EU (ECHA 2021). The distribution of lead will be different at these two types of shooting range and the resulting lead contamination likely to have different characteristics. For example, lead shot will usually be more widely distributed at clay target ranges, and it is smaller with a greater surface area:mass ratio that may make it more susceptible to weathering (Reigosa-Alonso et al. 2021). Lead shot tends to be spread across the soil surface at clay target ranges, where it has been found to make up to 30-40% of top layer of soil (VanBon and Boersma 1988; Austrian UBA 2002); at a clay target site in Austria it was estimated that shotgun firing discharged 1 tonne of lead per year to the site resulting in 40 tonnes being deposited over the 40-year lifetime of the range (Austrian UBA 2002). At another clay target range 25 t of lead shot was recovered during remediation (Bavarian LfU 2014).

Pistol and rifle shooting ranges are divided into the firing bay or line from where weapons are discharged, firing lane, the target line (where targets are placed) and berm (stop butt) behind the target line[9]. At pistol and rifle ranges the highest concentrations of Pb are found in the berm as this is where the bullets are captured after penetrating the targets (Dinake et al. 2019). However, bullets may be spread over large areas depending on the targets used (Okkenhaug et al. 2018) and Pb may also be elevated at the firing and target lines because of shooting activities (Sanderson et al. 2018; Sehube et al. 2017).

The use of lead in ammunition and where it ends up also varies according to user-groups and the type of shooting being undertaken (e.g. military, police, off-duty shooting, indoor ranges, short-distance ranges and shotgun ranges; BAFU 2020). It should be noted that different types of shooting are permitted in different EU members states, e.g. there are full bans on the use of lead shot in Netherlands and Denmark (ECHA 2021) so only rifle and pistol shooting is permitted to use lead ammunition in those countries.

Lead shot used in shotguns is generally 1-2 mm diameter (Soeder and Miller 2003) and contains lead (97%), antimony (2%), arsenic (0.5%), and sometimes nickel (0.5%) whereas

[8] Shotgun ranges can also be temporary or even transient i.e. annual or one-off shooting competitions in agricultural areas. The volumes of lead are consequently much lower than at permanent ranges and are unlikely to present a risk so this scenario is not considered further within this report

[9] Backstops/berms are small slopes usually made of sand or soil from of the surroundings. They are about 5-7 meters high and located at the end of the field, where remains of ammunition, usually fragmented, will accumulate (Rodriguez-Seijo et al. 2016)

lead bullets used in rifles are composed of lead (90-99%), antimony (1-10.5%), and copper (0.1%) (Dinake et al. 2019; Barker et al. 2020). The projectile from pistol ammunition is distinctly different as it contains only 52% lead (Dinake et al. 2019; Sehube et al. 2017). Bullets in the berm behind targets at small arms firing ranges (SAFRs) will have been subject to abrasion as they enter the backstop soil; the abrasion forms fine particles of metallic Pb that can rapidly be transformed into lead compounds that are more soluble than the initial metallic Pb (Laporte-Saumure et al. 2012). Fragmentation of spent ammunition is also very important and is specifically due to bullet-on-bullet impact, which generates a higher proportion of lower size fractions (e.g. <1mm and <250µm; Sanderson et al. 2018). Fayiga et al. (2011) found that soil at 3 rifle shooting ranges in Florida, USA had the most accumulation of Pb (60-70%) in the very coarse sand fraction (1-2mm), this was suggested to be due to bullet-on-bullet impacts leading to ongoing fragmentation and resulting in bullet fragments in the coarse fraction of soil.

The age of shooting ranges is also important when considering the make-up of the contamination source as older, historic small arms ammunition (pre WWI) was often almost pure lead (Larson et al. 2011). Historic ammunition tends to be in larger fragments compared with modern ammunition and shows slower dissolution rates after being deposited into soil (Larson et al. 2011); this is due to less fragmentation of the older ammunition which was softer due to its higher lead content; modern ammunition by contrast is harder (due to the addition of antimony), is fired at a higher velocity and modern rounds have a higher surface area: mass ratio. The fragmentation of modern bullets is also affected by soil type and distance from the target (Larson et al. 2011), i.e. larger bullet fragments are produced when the impact area is composed of soft, silty or clay soils and when the firing distance is shorter. A Canadian investigation (BCCDC 2011) also found that high velocity ranges deposit very fine particles of lead in addition to bullets and bullet fragments whereas soil at low velocity ranges tends to contain whole and only partially decomposed bullets/pellets.

Shooting at historic sites is sometimes still ongoing. This means that the contamination will have different characteristics in different parts of the site, e.g. a shooting range at Glanegg, Austria has been used by the Austrian military since the 19th century and was the subject of numerous investigations from 2010-15, which found it to be significantly polluted by lead with numerous hotspots (Austrian UBA 2018); even in the old part of the site with historic contamination, highly elevated soil lead concentrations of up to 28,000 mg kg^{-1} were detected.

The highest concentrations of lead at rifle and pistol ranges are invariably reported in the bullet impact berms (e.g. Sanderson et al. 2018). Pb concentrations in the range of 10-100,000 mg kg^{-1} have been reported in berm soils of numerous European firing ranges (e.g. Dinake et al. 2019; data from individual sites are included in Appendix 3 of this report). Similar concentrations of Pb in berm soil are reported in the USA (e.g Clausen et al. 2011; Fayiga et al. 2011[10]) and Africa (Sehube et al. 2017; Kelebemang et al. 2017). These very high concentrations of soil lead are measured within the berm soil or in the near surface soil (in front of the berm at SAFRs or in the drop zone where lead shot accumulates at a clay target shooting range), with a very sharp decrease in Pb concentration with depth, e.g. Laport-

[10] Pb concentrations from 3 shooting ranges in Florida were reported ranging from 10,000 to 70,000 mg kg^{-1}.

Samure et al. (2012) measured concentrations of ~12,000-67,000 mg kg^{-1} from 0-15 cm in the front of an SAFR berm and 423 mg kg^{-1} at 50-90 cm.

The spent ammunition at SAFRs may be retained by using bullet traps or specific sand berms. However, soil berms are still used in the EU for which lead migration be possible although a function of berms is to retain bullets and bullet fragments above the ground surface and this may assist in decreasing the potential migration of lead to groundwater. In contrast,, lead shot at clay target shooting ranges can become buried by the action of frost and the accumulation of new organic matter on the soil surface (Selonen et al. 2012); this accumulation will be most marked at ranges in forested areas. Selonen et al. (2012) investigated a clay target shooting range in a boreal pine forest in Finland; in the operational part of site the concentration of lead was higher in the surface layer of soil compared to the humus layer, whereas at the abandoned part of the site Pb in the humus layer was over twice as high as the concentration in the surface layer. The downward migration of Pb in this area is estimated to be 2 to 3mm per year.

Most data at shooting ranges are reported for total lead in soil following the removal of larger fragments of metallic lead (e.g. analysis is generally of soil particles <2mm) but it is the proportion of lead that is mobile or potentially mobile that is most relevant for assessment of the potential risk to groundwater. The weathering processes that result in the formation of more soluble forms of lead are covered in detail in Section 3.2 on soil chemistry.

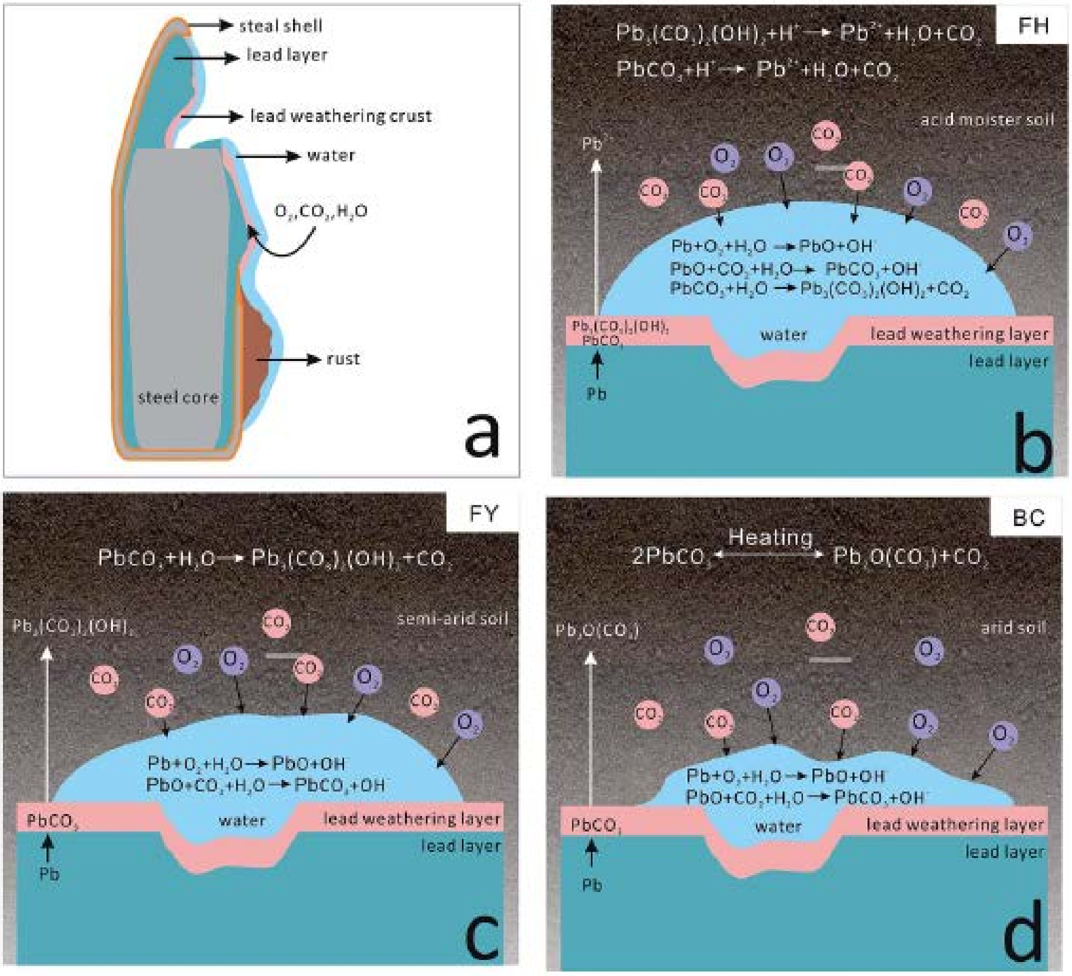

Figure 3.1 Schematic diagram of the weathering mechanism of a bullet exposed to air and water in three studied shooting ranges under different climatic conditions (from: Li et al. 2015)

3.2 Soil chemistry and lead fate and behaviour

Following deposition to soil lead shot and bullets will be subjected to weathering processes, the extent and outcome of which are determined by local soil and environmental conditions. In this subsection we build upon the discussions in the Annex XV Dossier (ECHA 2021) and attempt to detail some of the key soil chemistry factors that influence the behaviour and fate of lead at shooting ranges.

There are many studies focussed upon the weathering of lead in soils at historic and modern shooting ranges. It has long been established that total metal concentrations are generally poor measures of behaviour, fate and the potential risk of metals in terrestrial ecosystems (e.g. McLaughlin et al. 2000). Characterisation of the lead pellets, bullets, and fragments thereof, using solid-state speciation techniques such XRD, XRF, XANES, SEM in shooting range soils have identified the most common weathering products as cerussite, hydrocerussite, pyromorphite and anglesite (Fayiga et al. 2011; Sanderson et al. 2012; Barker et al 2020). Li et al. (2015) noted that crystal weathering phases of lead bullets could be readily identified as cerussite ($PbCO_3$), and hydrocerussite ($Pb_3(CO_3)_2(OH)_2$) in soils that where alkaline or circumneutral pH, but for the acidic soils, few if any crystal phases were identified (Figure 3.1). Solid-state methods can provide an understanding of the macro forms of lead from pellets and bullets in soils post deposition and particularly identify the mineral forms and characterise the weather products at submicron scale. However, these methods are not capable of providing the resolution required to assess the relatively low lead concentrations from weathering processes that may migrate from the upper horizons of soils.

In addition to the well-studied crystalline weathering forms of lead pellets and bullets in soils, lead released from these sources will be subject to a range of soil processes that will reduce its availability to biological organisms and for transport from the soil surface layers. Figure 3.2 shows the processes that collectively may be considered to represent natural attenuation. Under steady-state conditions the concentration of lead in the soil solution is buffered by the lead that is weakly bound, or exchangeable, on the soil surfaces. With increased loading of lead, this solution lead may remain relatively constant, while exchange surfaces and binding sites remain available. Soil factors are obviously hugely influential in determining this attenuation of lead and the concentrations in soil solutions. Janik et al. (2015) measured the solid-solution partitioning of trace metals, including lead, in 481 spatially representative soils from across Europe from the Geochemical Mapping of Agricultural Soils (GEMAS) program. A high distribution coefficient (K_d) indicates a greater association of metal with soil solid phases and lower values, that a greater proportion of the metal will be in soil solution. Metal retention in soils is often linear in relation to solution metal concentrations under low metal loadings, but curvilinear as loadings increase as high affinity sites are filled and only lower affinity sites remain. Janik et al. (2015) measured distribution coefficients for lead ranging from 10 to 339,624 L kg^{-1} (n = 481), with a median value of 32,284 L kg^{-1} and standard deviation of 45,406 L kg^{-1}. This relatively large variation demonstrates why default or ‘representative’ K_d values for metals are of limited worth in the development of continental and regional scale risk assessment scenarios and that local considerations of soil properties are key.

Empirical research focussed on ecotoxicological assessment of lead in soils, primarily in the form of lead salts, identified effective cation exchange capacity (eCEC), and those soil properties that influence eCEC (e.g. total C, exchangeable calcium and magnesium, clay content) as the driving effects on bioaccumulation (e.g. Smolders et al. 2009; Lanno et al. 2019). Experimental evidence supports the importance of the eCEC in affecting the bioavailability and biological uptake of lead from soils and suggests that lead from dilute chemical extractants, such as calcium chloride (0.01M), and in soil pore water could be used as predictors of lead uptake and availability (Zhang et al. 2019).

Operationally defined chemical extractants, including dilute chemical extractants and multiple stage soil sequential extraction methods have been widely used to assess the possible forms of lead in shooting range soils (Fayiga et al. 2011; Islam et al. 2016; Sehube et al. 2017; Kelebemang et al. 2017). These techniques utilise chemical reagents of differing severity to extract trace metals and equate the severity (and apparent reactant selectivity) with forms of the metal in the soil. For example, dilute extractants such as calcium chloride solutions or ammonium nitrate may be described by authors as readily exchangeable (i.e. the metal in solution and bound weakly to soil surfaces in Figure 3.2). Metals extracted from the remaining solid material using for example hydrogen peroxide may be bound to organic matter or using hydroxylamine hydrochloride as being bound to iron and manganese oxides. It is generally understood that this level of specificity regarding extractants removing particular ‘forms’ of the metal are somewhat ambitious and what these methods are generally useful for is assessing relative changes in similar soil types. The relevance of sequential extraction data in this assessment is therefore somewhat limited.

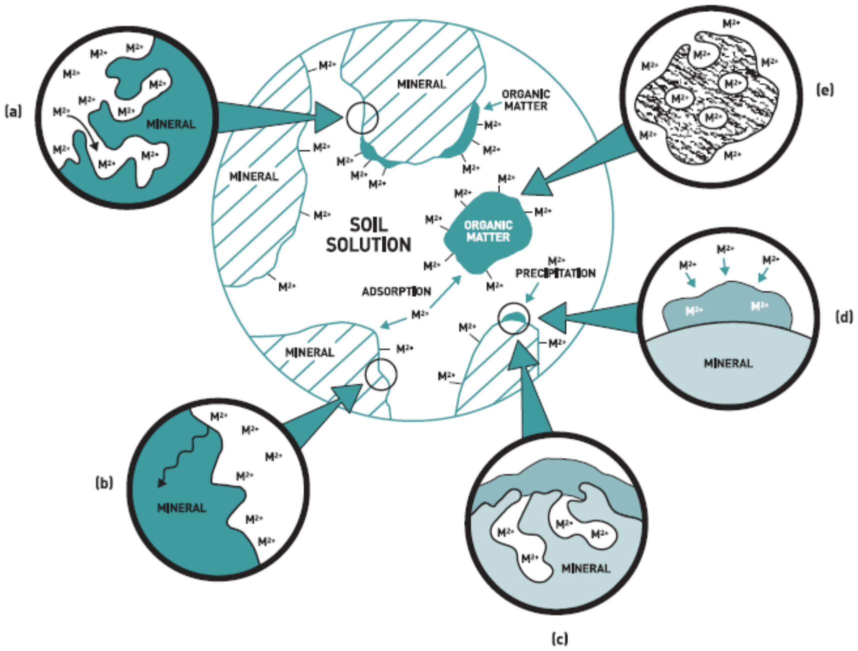

Figure 3.2. Adsorption, precipitation, and ageing processes in soil that may reduce the availability and transport of lead (ICMM 2007).

Mobility describes the tendency of metals such as lead to move through the soil profile and is assessed by a wide variety of techniques in the studies reviewed for this report, with the different (again operationally defined) test methods and ways in which the data are presented making comparisons extremely difficult or impossible. It is also questionable whether some of the methodologies commonly used are appropriate for determining the potential for Pb to leach to groundwater. For example, the commonly quoted USEPA TCLP test[11] is designed to determine waste classification and seeks to simulate the aggressive conditions of a landfill. In contrast another EPA method, the SPLP test[12], is specifically designed to assess leaching and the risk to groundwater and may therefore be more relevant. Results for ‘water soluble’ lead

[11] Toxicity characteristic leaching procedure (TCLP) involves a simulation of leaching through a landfill and soils having TCLP >5mg/L are classified as hazardous materials. US EPA Method 1311 (USEPA 1995[11]), uses ‘extraction fluid 1’; acetic acid, adjusted to pH 4.93 using 1M NaOH (or ‘extraction fluid 2’: acetic acid at pH 2.88 for alkaline solids)

[12] Synthetic preparation leaching procedure (SPLP) is used to assess mobility in different soil; it simulates unbuffered acid rain at pH 4 or 5 (prepared by adding sulphuric and nitric acid to water). Soils with a SPLP >15ppb are considered by USEPA to have potential to cause groundwater contamination (USEPA 1994 [12] https://www.epa.gov/sites/production/files/2015-12/documents/1312.pdf)

and SPLP data are reported in the summary of site investigations detailed in Appendix 3. As is often the case with these types of purposive simulation extraction procedure, field-based validation and evidence-based linkage to what the test is designed to reflect is not available or gives mixed outcomes, making interpretation challenging (e.g. Townsend et al. 2006).

There is no doubt that compared to the total concentrations of lead in the soil, relatively low concentrations will be water soluble, or potentially leached to lower parts of the soil horizon. In a study of metal mobility in two slightly acidic (pH 6-6.8), sandy loam, shooting range soils in Korea, Islam et al. (2018) noted that just 0.06 and 0.01 percent of the total lead content was water soluble. The lead concentrations at these low total percentages were between 0.4 – 11 µg L^{-1}. Further experimental results, specifically water extractions of the soil have been used to reflect the concentration of lead in shooting range that may be readily leached from the upper soil horizons. Hui et al. (2011) and Selonen et al. (2012) demonstrated elevated concentrations of lead in water soluble extractions from historic and current shooting ranges in Finland when compared to unimpacted sites. Soils were under coniferous vegetation and had relatively low pH values (≈ 3.5) but gave water soluble concentrations of between 110 – 120 mg kg^{-1} (3.2 mg kg^{-1} for unimpacted soils). Soil pH values of less than 6 are identified to accelerate weathering process of the lead shot and bullets (e.g. Li et al. 2015) and depending on the soil chemistry reduce the capacity of lead sorption process (Figure 3.2, e.g. Ogawa et al. 2014; Okkenhaug et al. 2018). For example, an investigation of a rifle range in Switzerland (ETH 2002) where soils had pH values of 3.7-7, observed that the proportion of water soluble lead correlated with the acidity of the soil, ranging from 0.002% of the total lead in the circumneutral soils to 1.7% of the total lead in more acidic soil found in an area of forest.

Although freely dissolved lead can potentially move in pore water, the importance of this process is reduced by reactions between Pb and mineral and organic components of the solid phase. Movement of lead is more commonly observed if it is associated (in ligands) with dissolved organic matter (DOM) or anions such as chloride or sulfate (Clausen et al. 2011 and cited references therein). Ligand associations with DOM (e.g. humic colloids) are believed to be especially important in controlling Pb mobility in soils because they can effectively maintain lead in solution under conditions in which it would normally bind or precipitate. The binding of lead to humic acids from peaty soils is especially effective in facilitating transport from areas of lead deposits to the extent it has been suggested for use in remediation and metal recovery (e.g. Lodygin 2020). The predicted precipitated species can control lead solubility and it has been noted that DOC has a role in maintaining soluble concentrations (and also transforming the metallic lead in to weathered forms). Yet, it is clear that because of the relatively high loadings of lead, dissolved concentrations at or above current drinking water levels are possible.

Reigosa-Alonso et al. (2021) used a several chemical extractants on soils from a historic shooting range in North West Spain to identify the form that lead was present in, whether that form was mobile and would reach subsurface layers (> 30 cm). Soil properties and possibly vegetation was considered by the authors to drive the mobility of lead, specifically the coarse texture of the soil, relatively low pH (4.8 – 6.6) and low iron (15 g kg^{-1}), manganese (0.5 g kg^{-1}) and aluminium (3.5 g kg^{-1}) content reduce the capacity for lead attenuation. The role of vegetation in mobilization of lead from shooting ranges was investigated by Fayiga and

Saha (2016) in a column experiment using three sandy textured (> 85% sand), slightly acidic (pH 6-6.8) shooting range berm soils from Florida, with treatments where the bullets removed through sieving, and grassed. The effect of the vegetation on the characteristics of the leachate are shown in Table 3.1. The lead leached decreased for two of the soils, between the grassed and control soils and the authors considered this likely due to greater levels of root uptake in those soils.

Table 3.1 Effect of sieving and vegetation on leachate characteristics in shooting range soils (from: Fayiga and Saha 2016).

Range	SR 1		SR 2		SR 3	
	Grass	Control	Grass	Control	Grass	Control
Total leachate Pb (mg/L)						
Unsieved	11.4 ± 1.89a	13.9 ± 3.00a	1.86 ± 0.45a	1.42 ± 0.04a	40.0 ± 13.6a	129 ± 32.0a
Sieved	17.5 ± 3.17b	19.9 ± 1.94b	1.30 ± 0.12b	1.10 ± 0.10b	61.0 ± 17.0b	163 ± 26.0b
Total DOC (mg/L)						
Unsieved	590 ± 4.44a	634 ± 50.4a	700 ± 7.14a	989 ± 73.5a	481 ± 15.2a	583 ± 16.2a
Sieved	777 ± 8.53b	818 ± 19.6b	777 ± 36.8b	1032 ± 136a	526 ± 35.8b	777 ± 26.1b
Leachate pH						
Unsieved	6.75 ± 0.54a	6.24 ± 0.14a	7.58 ± 0.10a	7.56 ± 0.23a	6.96 ± 0.22a	5.51 ± 0.44a
Sieved	7.32 ± 0.11b	6.15 ± 0.28a	7.68 ± 0.25a	7.61 ± 0.13a	6.12 ± 0.34b	5.70 ± 0.43a

Values are means ± standard error, n = 4, Error includes instrumental error, instrumental drift error, analytical error.
Treatments with different letters are significantly different at $\alpha = 0.05$; SR – Shooting range, control – un-vegetated soils.

The importance of the formation of crystalline lead forms controlling dissolved concentrations have been investigated by Jurgens et al. (2019) through an assessment of the potential of lead exposure via drinking water from untreated groundwater sources in the U.S. The lead source was the plumbing pipes and fixtures from the well, to the dwelling. The authors utilised the geochemical speciation model PHREEQC (v3) to estimate the lead solubility potential in 8,300 untreated groundwater samples collected nationally from 2000 to 2016. The model calculated the concentration of lead that could be in solution before the formation of a lead-bearing precipitate would form given the water chemistry conditions of the groundwater. Importantly, one of the aims of the work was to identify the conditions that may result in potential increases in lead exposures in drinking water. Highly susceptible groundwaters that would lead to elevated lead concentrations (7.5-15 µg L^{-1}) were those that had geochemical characteristics that would limit lead precipitating as a solid or mineral phase. Specifically, these were:

- Acidic (~ pH 5.1);
- Low concentrations of alkalinity (8.0 mg L^{-1});
- Low concentrations of orthophosphate (<0.01 mg L^{-1}).

These findings indicate that hydrogeological conditions typically control the potential for transport of lead through the vadose zone and into groundwaters.

From the literature we have reviewed here we may summarise that:

- Typically, lead is retained close to the soil surface. However, under the extreme loadings at shooting ranges, evidence indicates that lead may percolate down the soil profile.

- Once deposited in or on the soil the behaviour and fate of the lead from pellets or bullets will be determined by soil properties, climate, and management practices (Sanderson et al. 2012).
- Natural attenuation, at such relatively high lead loadings, for some soil types will not be effective in the long-term (taking 000's of years) to retard downward migration of lead.
- The key soil properties to that may prompt lead movement from the soil surface to underlying layers are low pH (<6), coarse textured and freely draining soils with relatively high levels of dissolved organic carbon and low iron and manganese content.
- Other factors of importance are likely to include elevated rainfall (where precipitation is much great than evapotranspiration), and presence of shallow groundwater (< 3m).

3.3 Pathways

Following the weathering of lead ammunition discussed in Section 3.2 the initial step in the pathway from soil to groundwater is the movement of lead into 'soil water' through the vadose zone. It could be reasonably suggested that the closest estimates of what concentrations of lead may be present in soil waters could be those that have been measured in porous cup lysimeters and root zone samplers, although the practical difficulties of measuring soil water chemistry are well known (e.g. Watmough et al. 2013).

Clausen et al. (2011) has suggested that lead subsurface migration would be limited to 1-3 metres, and that detection of lead beyond this was probably reflective of experimental artifacts. Yet, in an earlier paper also by Clausen (Clausen and Korte 2009) concentrations of lead of 50-670 µg L^{-1} were measured in soil pore waters in soils at three U.S. military training facilities using ceramic suction-cup lysimeters (the LoQ for lead was 1 µg L^{-1}, using ICP-MS). Using similar lysimeters to Clausen, in the vadose zone beneath a calcareous sandy berm with slightly alkaline pH (7.6-8.0) at a small arms shooting range in Canada Laporte-Saumure et al. (2012) measured much lower concentrations of lead in the region of 10 µg L^{-1}. The groundwater table was at a depth of 6.5 m below the berm and concentrations of lead were determined to be at background levels in the groundwater.

Soil solution concentrations of lead were also measured in calcareous alluvial soil from a shooting range in Switzerland using large scale lysimeters (17.5 m^2) over several months through different water holding conditions by Hockmann et al. (2018). Lead concentrations were unaffected by changes in redox conditions induced by waterlogging but did show a clear seasonal trend reflecting lack of percolation during deep winter (Figure 3.3). While it is known that lead is not redox active, the sorptive phases responsible for reducing lead concentrations in solution are, e.g. ferric (hydr)oxides and sulphides (shown above in Figure 3.2). Therefore, it is reasonable to expect that lead concentrations in pore water may increase under reducing conditions. Work by Dewey et al. (2021) suggests that a potential reason for this limited release of lead into solution phases is binding to particulate organic matter across the critical redox transitions. These authors assessed lead mobility in a floodplain soil contaminated by historic metalliferous mining upstream; the soil was of circumneutral pH with less than 3% carbon content, with seasonally varying groundwater levels; this resulted in elevated but relatively low lead concentrations (< 500 mg kg^{-1}). Dissolved lead concentrations in the soil porewater were measured at less than 17 µg L^{-1} and were considered by the authors to remain as such provided there was sufficient particulate organic matter and sulphur (the source of lead was galena, lead sulphide ore) to ensure binding to solid phases during the fluctuating redox cycles.

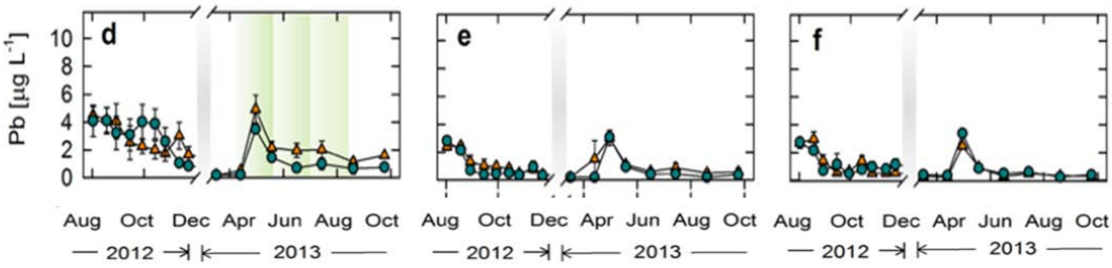

Figure 3.3 Concentrations of dissolved lead in the soil solutions from the drained (triangles) and waterlogged lysimeters (circles) at 20 cm (d), 37 cm (e) and 54 cm (f) depth. Error bars represent standard errors (n = 4-6) (from: Hockmann et al. 2018)

The Annex XV Dossier (ECHA 2021) details the case of a shooting range in Germany (Mainbullau) with 40 years of activity. In 2019 lead concentrations in soil pore water, as sampled by suction lysimeters at 70cm depth, were measured at five different locations as 44.5, 1,460, 198, 64.4, and 12.9 µg L^{-1}. Two of five of these measurements exceeded the Phase 2 action level of 100 µg L^{-1}, requiring remediation (Bavarian WWA Aschaffenburg, 2019). A detailed investigation of this site was requested by authorities, with reference to Bavarian soil protection laws, due to insufficient knowledge of the location's geology and mobility of the contaminating materials (arsenic, antimony, and lead).

At the Glanegg shooting range in Austria (Austrian UBA 2018), a sloping area that formed a natural barrier for the capture of lead shot had measured lead concentrations of up to 1,000 mg kg^{-1}. It was considered that the lead was of relatively high mobility as leaching studies indicated >0.1% mobile lead. This mobility was confirmed by the detection of elevated lead at the deepest lysimeter depths of 90cm and some percolation waters were considered highly contaminated (although no data are provided). A soil water Pb concentration of 12.6 mg L^{-1} was measured at 5cm depth at the Kuchlmuhle clay target shooting range in Austria (Austrian UBA 2002) in a markedly acidic soil (pH 4.3 in humus layer). The concentration of Pb in soil water was observed to decrease significantly with depth, being 1.5 mg L^{-1} at 15cm and 0.004 mg L^{-1} at 40cm.

Okkenhaug et al. (2018) investigated lead mobility at a historic shooting range site in Norway over peatland soils (pH ≈ 5.3) with shallow groundwater. The authors measured relatively low concentrations of lead in pore waters extracted by suction lysimeters (~ 1 µg L^{-1}) and postulated that this was due to the elevated soil pH at the site, compared with other peat bog studies. Nevertheless, they did measure lead concentrations of 22 ± 5 µg L^{-1} in shallow groundwater and elevated lead concentrations in surface waters draining the site, supported by hydrogeological modelling, and noted strong correlations between lead and DOC concentrations. Soeder and Miller (2003) also measured highly elevated Pb (1 mg L^{-1}) in shallow groundwater (<1 m) at a trap-shooting range that was operated in the United States from 1962–1998. It was suggested that lead from a concentrated deposit of shotgun pellets on the at the site had been mobilized through a combination of acidic water conditions and a very sandy, shallow, unconfined aquifer. It should be flagged that both sites would be considered as wetlands and therefore not strictly within the scope of the restriction, but they illustrate the potential for significant quantities of lead to be mobilised and for a proportion of this to vertically migrate to groundwater under certain specific conditions.

From the previous subsections, it is apparent that local soil and environmental conditions may lead to elevated concentrations of lead to be found in soil pore waters at shooting ranges. However, it is important to assess the key potential linkages between the lead in soil/porewater and its movement to groundwater.

Lead can be transported in soils, the vadose zone and in groundwater as either particles (e.g. small lead fragments from projectiles or lead sorbed to mobilised soil particles) or in solution. In the case of particulate lead, smaller particles are generally associated with the highest potential mobility in both porous matrices and in surface runoff. Small particles can be transported through the soil and vadose zone matrix, although substantial displacement of particles is only likely to occur where there is preferential flow. Preferential flow can occur in homogeneous matrices (sometimes called “fingered” flow) but is often associated with macropores. The latter form of preferential flow is most likely to transport both dissolved (including Pb associated with humic colloids) and particulate Pb, because there is potentially less interaction between moving solute and the soil solid phase (Vanderborght et al. 2002).

Knechtenhofer et al. (2003) observed rapid reductions in lead concentrations with depth and a relatively uniform spatial pattern in an acidic soil at a shooting range in Switzerland even in the presence of pronounced near surface finger-type preferential water flow. However, below 20 cm, preferential flow associated with tree roots appeared to transport some lead deeper in the soil (and possibly breakthrough to groundwater). These deeper macropores are believed to be long-lived and may be responsible for small but potentially significant amounts of contaminant transport, despite the fact that most lead (>95%) appears to be very strongly retained in the upper 10cm of the soil. Garrido and Helmhart (2012) also showed that preferential flow paths were potentially important in the mobilisation of lead from contaminated roadside soils: soil sampled from zones with substantial preferential flow, as identified by dye tracing, had higher lead concentrations than soil identified as having predominantly matrix flow (i.e. little or no preferential flow). Soil in the preferential flow paths had lower pH and higher carbon content (organic and inorganic) compared with bulk matrix soil. This study suggests that, in this case, lead is mobilised and transported down preferential flow paths but is also retained along these paths, possibly in association with higher carbonate concentrations arising from the CO_2 generated from enhanced respiration in root channels. No evidence of preferential migration of lead out of the soil profile was presented in this case.

Clearly, the deeper the water table, the less likely it will be that continuous macropores (e.g. from active or old root channels or earthworm burrows) will connect the soil surface with the saturated zone. Thick unsaturated zones provide many opportunities for contaminants such as lead to come into contact with potentially reactive solid phase components; this means that the bulk of lead that is mobilised from the near-surface soil will be transported a relatively short distance before being immobilised. However, there may be situations in which there is some enhanced connectivity between the near-surface soil and the saturated zone, even in otherwise low permeability matrices - such as in fractured chalk (e.g. Nativ et al. 1995). Whilst there will be a tendency for solute moving down a vertical macropore wall to be imbibed by the matrix, the rate at which this occurs will depend on the water content (and, hence, the tension) and the hydraulic conductivity of the matrix. If the matric potential gradient at the wall is low (e.g. when the matrix is relatively wet) and or the hydraulic conductivity of the matrix is low, solute may penetrate the vadose zone via the macropore network (Beven and Germann 1982).

Soluble lead would be transported in porous media such as soil and rock via a combination of diffusion and advection. Advection, in which the contaminant is transported along with moving water, is generally much more important than diffusion. However, diffusion and hydrodynamic dispersion are important in spreading out a contaminant as it moves through the medium via advection. Water movement is driven by the gradient in potential energy (the hydraulic gradient) and is also limited by hydraulic conductivity (which increases steeply with increasing water content to a maximum value at saturation: K_{sat}). Both the energy gradient and the hydraulic conductivity are influenced by the pore size distribution (which is correlated with grain size). Water will only drain under the influence of gravity from larger pores. In small pores water is retained by capillary forces. Coarse textured materials have larger pores and tend to have higher K_{sat} values than fine textured media. They are, therefore, more easily drained and, hence, may transfer contaminants more readily under the same climatic conditions. However, substantial advective flow is possible in some fine textured media via macropores (cracks, root channels or burrows created by soil fauna; Beven and Germann, 1982). Significant drainage only typically occurs when soils are relatively wet (i.e. when larger pores become water filled). In Europe this occurs during late autumn, winter, and early spring, when precipitation or snow melt exceeds evapotranspiration. Net energy gradients are generally vertical in unsaturated soils. Below the soil, in the unsaturated (or vadose) zone, water and associated contaminants can continue to migrate vertically downwards until they reach a permeability discontinuity (e.g. a low permeability layer) or the water table. Unsaturated zone transport is relatively slow because the unsaturated hydraulic conductivity is low below saturation. In the saturated zone, hydraulic conductivity is not affected by the water content because the pore space is saturated, although K_{sat} for rock is often lower than that for soil and weathered regolith. Potential energy gradients in the saturated zone are driven by a combination of gravity and pore water pressure – with water moving down topographic gradients close to the phreatic surface but also moving from regions of high to low pore water pressure further away from the water table. This often drives water towards rivers, streams, springs and wells. If a well is present, a high rate of abstraction can result in the depression of the water table around the well (sometimes called a cone of depletion) which can accentuate the hydraulic gradient between the bulk aquifer and the well and, thus, increase the rate at which groundwater moves to into the well. If the groundwater is contaminated, this can increase the risk of well contamination.

One final point should be made about very shallow groundwater – for example in the riparian zones of streams. If the water table is seasonally close to the ground surface it can periodically interact with the soil – bringing the saturated zone into contact with potentially high soil lead concentrations, if present (lead levels at shooting ranges are likely to be highest close to the surface and to decrease quasi exponentially with depth, e.g. Knechtenhofer et al. 2003). This has the potential to "flush out" Pb from soil pores which would otherwise be immobile under unsaturated conditions. This scenario is probably not commonplace but could occur if all or part of a shooting range were situated in a riparian zone with shallow groundwater. It was considered by Jarsjo et al. (2020) in a modelling exercise assessing the effects of an elevation in the water table resulting from climate change in till soils. They found that a decrease in the depth to the water table (i.e. an increased water table elevation) of just 20 cm, relative to historical observations, resulted in substantially more predicted lead mobilisation and transport (in part, due to the much higher values of K_{sat} which are typically observed closer to the ground surface). That said, predicted lead transport under the current climate scenario was low and, although plausible, this enhanced risk under climate change remains somewhat speculative.

3.4 Receptor

The sensitive receptor in our conceptual source-pathway-receptor model is groundwater as many aquifers are abstracted in the EU for the supply of domestic drinking water, the EC (2021a) report 'Groundwater as a Resource' suggests that the proportion of EU households supplied from groundwater could be as high as 75%. Private wells are also drawn from groundwater and may be more susceptible as they are commonly drawn from shallow groundwater[13].

The potential for lead to contaminate groundwater is a major concern due to the highly toxic nature of lead compounds, which have been demonstrated to have a wide range of health effects including renal toxicity, cardiovascular effects and neurobehavioural effects in children (EFSA 2010). The adverse effects of lead are thought to have no threshold so exposure should be as low as reasonably practicable. The drinking water standard for lead has been lowered in recent years and is currently set at 10 µg L^{-1} with the stated objective to lower it to 5 ug/L over a transition period of ~10 years (EC 2020).

The term groundwater is usually used to refer to water in saturated rocks in the sub-surface. This water is commonly abstracted for use in irrigation, in industry (including the manufacture of food and beverages) and for (public and private) domestic water supply. Groundwater also maintains baseflow in rivers and streams during periods with little or no runoff and hence supports freshwater ecosystems. In terms of the protection of human health from exposure to harmful levels of Pb, wells used for domestic supply are clearly the priority receptors. Of these, private wells are often seen as being most at risk because they are commonly relatively shallow and are usually not as regularly checked for water quality, compared to those operated by municipal water suppliers. Groundwater which has a high enough porosity and permeability to allow either a significant flow or to allow the abstraction of significant quantities of water

[13] For example, at a military shooting range at Glanegg, Austria (Austrian UBA 2018) the subsoil was determined to be made up of quaternary gravel and sand deposits and the depth to groundwater is only 4-5m, with this aquifer supplying the local city of Salzburg. Also, within a radius of 500 m there are four further groundwater withdrawals, including two house wells and two utility water wells.

is termed an aquifer (EC 2016). Aquifers can be either porous (such as sandstone, which tend to predominantly exhibit inter-granular flow) or fissured (such as fractured chalk or karstic limestone, where most flow takes place via cracks and channels between the bulk rock matrix). Both types can represent important aquifers. Aquifers can also be designated as unconfined (which only have impervious layers below them) and confined (which have impervious layers both above and below the main saturated strata). Reliance of groundwater for water supply varies widely across Europe. According to the EEA (2019) about 24% of the total water abstraction in Europe was from groundwater (mainly for agriculture). However, the fraction of EU inhabitants which rely on groundwater for their water supply may be as high as 75% EC (2021a). Similarly, the fraction of the EU population which relies on private wells also varies widely by country. According to Hulsmann (2005), the fraction of EU citizens served by "very small" supplies was as high as 10% - predominantly in rural areas (and including community managed supplies of different types). However, the proportion of this fraction made up of private wells is not reported. WHO (2011) have collated available data for some individual European countries which provide useful *ad hoc* illustrations; for example, the fraction of the population reported to reply on private supplies (boreholes and wells) is 25% in Lithuania, 16% in Estonia, 10% in Finland and 7.6% in Czechia.

3.4.1 Groundwater Vulnerability

According to the EC (2021b) "groundwater vulnerability" refers to the system characteristics which influence the ease with which groundwater may be contaminated by human activities. This vulnerability can be either (i) "intrinsic", referring to characteristics of the hydrogeological setting which could affect the propensity of substances introduced at the ground surface to contaminate groundwater in general (Vrba and Zaporozec 1994) or (ii) "specific", referring to the vulnerability of groundwater to a particular contaminant or a group of contaminants.

3.4.2 Intrinsic Vulnerability

Aller at el. (1985) summarised seven factors which affect intrinsic vulnerability with the acronym DRASTIC:

(1) Depth to the water table (**D**): High vulnerability tends to occur where the unsaturated zone is shallow (i.e. superficial aquifers which have unsaturated zones less than 5 m thick). Thick unsaturated zones give more opportunity for contaminants, such as Pb, to come into contact and react with the soil & rock matrix and potentially be immobilised via a range of reactions.

(2) Net recharge (**R**): High rates of net recharge (*cet. par.*) are considered to enhance vulnerability because the rate of solute transport through the unsaturated zone is higher. However, very high recharge rates can also result in dilution of mobilised contaminants.

(3) The aquifer medium (**A**): High vulnerability occurs when a significant fraction of the total flow is via fractures (as opposed to via intergranular flow). Materials with high bulk permeability have higher vulnerabilities in general because there is a lower capacity for attenuation.

(4) The soil medium (**S**): Finer textured soils (such as silts or clays) are generally considered to be less vulnerable to pollutant transfer to the underlying layers than

coarse textured soils (such as sands), provided that shrink-swell clays do not create significant cracking which can cause preferential flow which can by-pass the soil matrix.

(5) Topography (**T**): Land surface topography affects near surface hydrological processes such as overland flow but is also sometimes indicative of the hydraulic gradient of the saturated zone at the water table surface. Highest vulnerability is assigned to low gradient areas because of the enhanced chance of contaminant infiltration, rather than runoff. However, steeper slopes infer higher hydraulic gradients and more rapid groundwater velocity which could increase vulnerability because attenuation times maybe reduced.

(6) The impact of the vadose zone (**I**): Vulnerability is strongly controlled by the nature of the materials forming the unsaturated zone. As for the aquifer itself, high groundwater vulnerability occurs when a significant fraction of the total flow in the unsaturated zone is via fractures (as opposed to via intergranular flow). Fine grained materials, such as clays, often increase unsaturated zone travel times and act as protective barriers, as long as they do not have significant fracturing.

(7) The hydraulic conductivity of the aquifer (**C**): This refers to the rate of saturated flow under unit hydraulic gradient. High vulnerability will occur when hydraulic conductivity is high because contaminants can move rapidly through the aquifer (e.g. from point of contamination to point of abstraction, e.g. for drinking water use).

It is important to note that the vulnerability of unconfined aquifers is generally considered to be higher than that of confined aquifers, although even in confined systems there may be some recharge through confining layers (Aller et al. 1985). In the original scheme and in most applications of DRASTIC, the factors are assigned weights (w) to reflect their general importance to overall intrinsic vulnerability (V). The weights are shown in Table 3.2. Each factor is also assigned a rating value (ρ) from 1 to 10, with the highest vulnerability assigned a rating of 10. For example, aquifers with a depth to the water table of < 1m will have a rating of 10, whereas a depth over 100m would have a rating of 1. Karstic aquifers (i.e. those with limestone geologies characterised by large fissures and caverns) have a rating of 10 whereas massive shales score only 2. For full details see Aller et al. (1985). Thus, overall vulnerability is:

$$V = \sum_{i=1}^{7} w_i . \rho_i \qquad (1)$$

where the index i is the DRASTIC factor and w and r are the weights and ratings, respectively.

Table 3.2 Weights assigned to each DRASTIC factor (from: Aller et al. 1985). The higher the weight the more important the factor.

Factor	Weight
D	5
R	4
A	3

S	2
T	1
I	5
C	3

Two contrasting cases of intrinsic vulnerability are illustrated in Figure 3.4; High vulnerability in settings with (for example) an unconfined aquifer with a shallow water table, fractured calcareous geology (e.g. Karst) and thin, high permeability soils and an example of low vulnerability where deep groundwater is overlain by thick, fine-textured soils and or glacial drift deposits, such as boulder clay (which has a very low hydraulic conductivity and can act as a barrier to the migration of pollutants through the unsaturated zone).

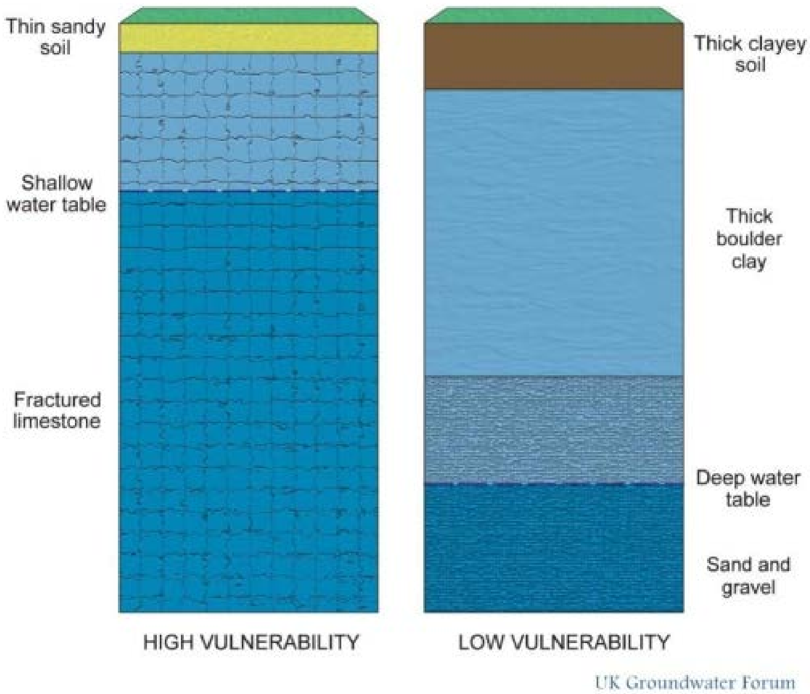

Figure 3.4 Illustration of two examples of intrinsic groundwater vulnerability. On the left, high vulnerability is predicted in settings in which an unconfined aquifer has a shallow water table, fractured calcareous geology (e.g. Karst) and thin high permeability soils. On the right, low vulnerability is predicted where deep groundwater is overlain by thick fine-textured soils and or glacial drift (e.g. boulder clay). (from: Carey et al. 2017, originally from the UK Groundwater Forum).

The DRASTIC methodology was applied by the European Commission (2021b) to map intrinsic groundwater vulnerability across Europe (including the EU, Norway, Switzerland, the UK, the Balkans, part of Turkey and Baltic Russia); the output is shown in Figure 3.5. Areas with high intrinsic vulnerability include the Po basin, the Puglia Region of Italy, parts of western, central and northern France (e.g. Aquitaine, Poitou-Charentes, the Ile de France, Picardie and the Nord Pas de Calais), parts of northern Germany, Belgium, the Netherlands and parts of the Baltic States. These areas have a variety of combinations of shallow groundwater levels, low slope angles, unconsolidated sedimentary geologies, and high recharge rates. High vulnerabilities in central Ireland and lower Bavaria are predicted due to Karstic geologies with shallow water tables. Lower intrinsic vulnerabilities are predicted in mountainous areas.

Karstic groundwater is considered to be especially vulnerable to contamination due to thin soils, very low permeability matrices and the rapid movement of water through large fissures both above and below the water table (e.g. Vias et al. 2006).

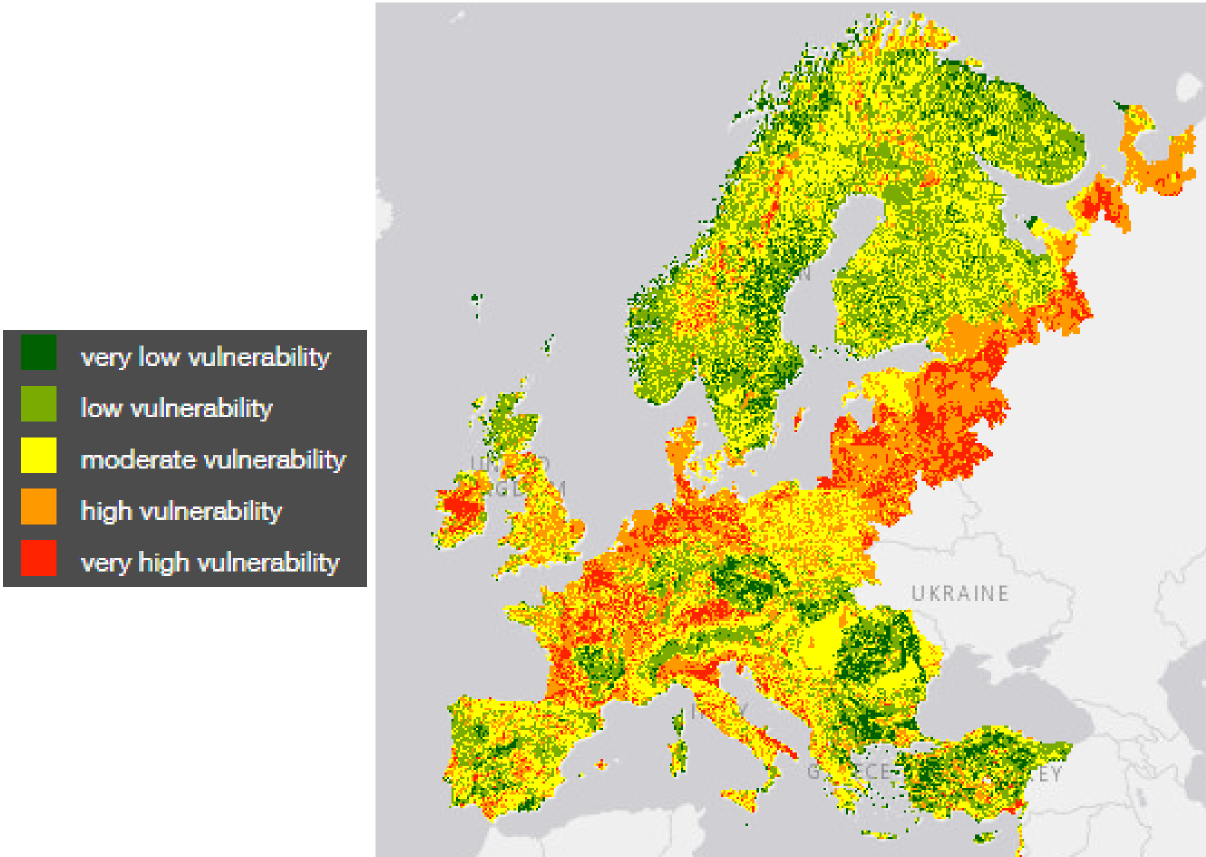

Figure 3.5 Intrinsic groundwater vulnerability map for Europe generated using the DRASTIC methodology by the European Commission (https://water.jrc.ec.europa.eu/).

Similar intrinsic vulnerability schemes have been applied elsewhere (e.g. in the UK: SNIFFER WFD 28, 2004; Carey et al. 2017 and in Europe for Karstic systems: Daly et al. 2002; Vias et al. 2006). In the case of the intrinsic vulnerability maps developed for England and Wales (Carey et al. 2017), scores are calculated on the basis of recharge rate, soil leaching class (assigned a priori to soil types by the National Soil Survey), glacial drift properties, unsaturated zone flow type and depth. Low scores indicate high vulnerability. Two map products have been produced (1) a combined map which separates groundwater vulnerability from aquifer designation status – allowing the propensity for pollution to be separated from the level of harm (e.g. to drinking water) and (2) a simplified map which gives the overall risk to groundwater e.g. from a pollution incident or activity, similar to the DRASTIC score. This distinction allows pollution risk mitigation measures to be prioritised in areas containing productive aquifers, which are likely to be utilised for water supply. Unproductive strata are not likely to be abstracted and, hence, have lower priority for protection. In the case of the COP methodology developed for Karst (Daly et al. 2002; Vias et al. 2006), vulnerability is calculated as the product of three factors representing (i) C: flow concentration (the potential to bypass overlying layers by, for example, swallow holes); (ii) O: the protection provided by overlying layers, including the thickness and nature of the unsaturated zone (soils, drift and bedrock) and (iii) P: vulnerability modification afforded by the annual amount and distribution of precipitation (vulnerability is assumed to increase as mean annual precipitation increases upto 1200 mm y-1; beyond 1200 mm y-1 vulnerability is assumed to decrease due to dilution effects). Clearly, schemes like DRASTIC and COP are indicative and cannot describe the detailed characteristics of individual sites.

3.4.3 Specific vulnerability

Specific vulnerability extends the intrinsic vulnerability of the hydrogeological setting to incorporate factors influencing the source strength of a particular contaminant. In the case of nitrate, a common groundwater pollutant, this could include land use and nitrogen fertiliser application rates in the recharge zone. Specific vulnerability may also be increased if net recharge is influenced by irrigation. In the case of lead from shooting ranges specific vulnerability might include the areal extent of the operation, the intensity of shooting activity and the length of time over which the range has been in operation. In addition to soil hydraulic characteristics affecting intrinsic vulnerability, soil properties on the range could also affect Pb speciation and mobility (see Section 3.2).

4 RISK ASSESSMENT

In this section we describe exposure related factors that may influence any assessment of potential risks to groundwaters from the use of lead shot and bullets. Specifically, we have focussed upon the types of groundwaters that may be vulnerable, the timeframes over which this may occur and the gaps and uncertainties in the assessment of sites to ascertain potential lead risks to groundwaters.

4.1 Specific groundwater vulnerability for lead and likely prevalence

Specific groundwater vulnerability for lead arising from shooting activities will be high where there is a combination of high lead emission rate, driven by usage (e.g. number of rounds or amount of lead shot per day) on acidic soils (which will enhance the dissolution of lead fragments) with moderately high organic carbon in zones with high intrinsic groundwater vulnerability (e.g. high DRASTIC V values: Equation 1).

One potential challenge with assessing specific vulnerability for spatially limited activities such as shooting ranges is the scale-mismatch between the typical size of firing ranges and spatial information about groundwater characteristics – which may only be available for large grid cells or regions much larger than the area of firing ranges. It is also likely that the most intensive firing activity will be concentrated in restricted areas (Section 3.1). This suggests that Pb concentrations in soil and the underlying vadose zone are likely to be very high in these high activity areas but much lower on average. Even within zones of high activity it is likely that the spatial variability in concentrations will be high (Clausen et al. 2010) and likely to be accentuated by preferential flow (e.g. Knechtenhofer et al. 2003). The risks associated with localised plumes of high Pb water are likely to be highly context-specific. It is also important to make the distinction between aquifers and non-aquifers (Section 3.4). Groundwater bodies with low yield (non-aquifers) are unlikely to be utilised for water abstraction and, hence, present a much lower risk to receptors such as humans from Pb contamination.

It is difficult to estimate the prevalence of these specific conditions across Europe. Whilst there are zones of high intrinsic groundwater vulnerability in some regions (Section 3.4.2 and Figure 3.5), there may be localised situations elsewhere which also present high risk. Thus, although high specific groundwater vulnerability for lead from firing ranges is probably limited to a small fraction of total sites in Europe, this fraction may not be insignificant. Detailed site-specific risk assessments would allow very high-risk activities to be more easily identified and managed.

Some areas with high intrinsic vulnerability are likely to occur in all EU member states, although to different extents (even within member states). Groundwater which is classed as an aquifer is clearly more important to protect than low yield groundwater that is not likely to be used for abstraction. If appropriate data were available, it would be possible to estimate the fraction of aquifers in each member state that have a high vulnerability (e.g. using spatially referenced data on DRASTIC class, or similar vulnerability indicator, and the spatial extent of major and minor aquifers). Detailed GIS analysis would be required to estimate the extent of areas with high vulnerability which are also areas classed as aquifers. Information on the fraction of shooting ranges occurring in these high vulnerability areas would provide quantitative information on the extent of the risk and requirements for associated risk management. Figure 4.1 shows the outcome of an attempt to identify shallow groundwater areas (water table < 10m below ground level) in the EU which are vulnerable to pesticide leaching (Negley et al., 2013). This primarily utilised topographic data assuming that shallow groundwater is more likely to occur in contiguous low gradient areas in river valleys.

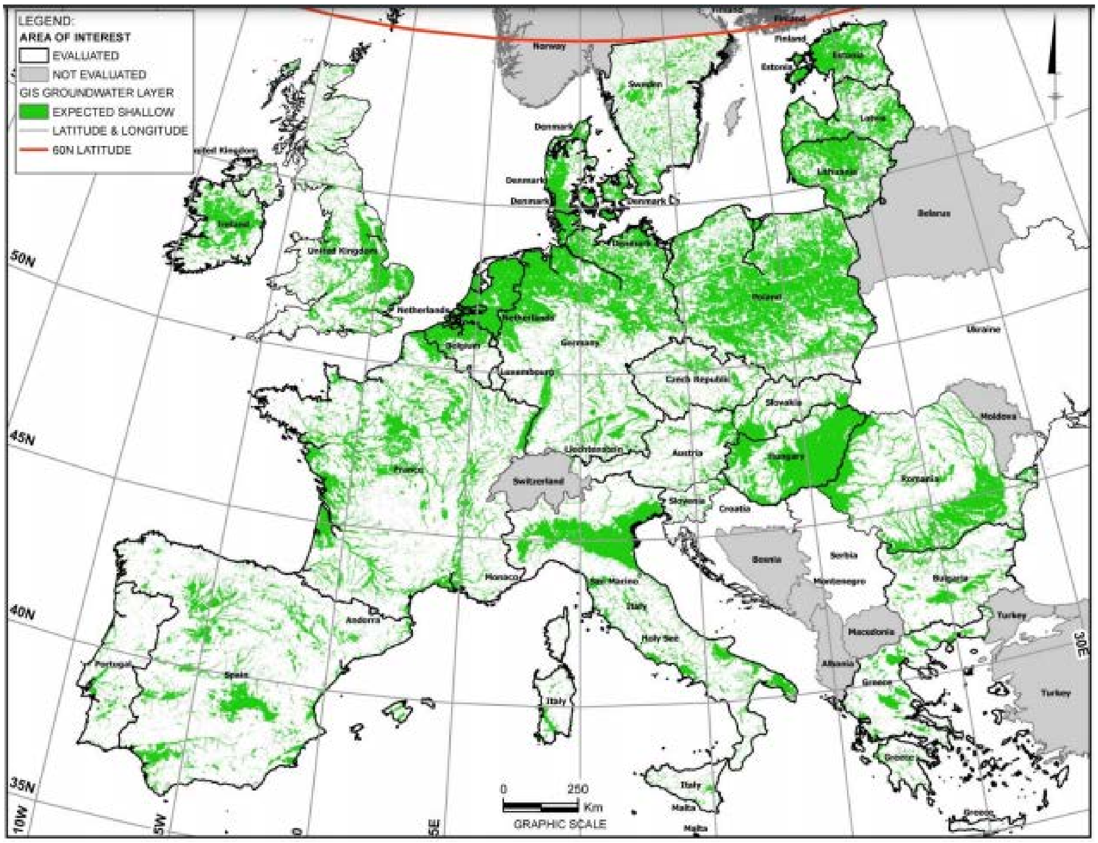

Figure 4.1 Estimated "pseudo-alluvial" areas which are predicted to have shallow groundwater according to a topographic analysis conducted by Negley et al. (2013)

4.2 Timeframes

Section 3.2 has provided details of the weathering processes undergone by lead shot and bullets on deposition to soil. Clausen and Korte (2009) have suggested that the capacity of soil for lead sorption is not infinite, but in most cases the mass of lead introduced into the environment and subsequently dissolved is negligible compared to the sorptive capacity of the soil, with the suggestion that lead migration is typically limited to a few metres in the vertical direction. However, it should be noted that the Clausen et al studies (2009, 2011) are specifically focussed on SAFRs and do not consider the use of lead shot at clay target ranges.

Furthermore, studies suggest the solubility of weathering products largely controls lead release into solution but the rate of weathering of the lead pellets and bullets is relatively slow. Importantly, the rate of weathering of pellets and bullets appears to be strongly soil pH dependent, for example it is estimated that in circumneutral soils just 4.8% of lead in the pellets has been transformed to lead carbonate and lead sulphate over 20-25 years at a Swedish shooting range (Lin et al. 1995). Over the same time period, on a site with more organic rich soils these authors estimated that 15.6% of the lead was transformed to secondary lead compounds. Regiosa-Alonso et al. (2021) suggests an even faster rate of weathering, citing three studies reporting a range of 5-17% over a relatively short time of 6-13 years, and from these observations estimate an annual weathering rate of the lead shot of 0.7-1.25% per year. Complete transformation of the lead pellets and bullets to weathered products has been estimated to take between 100 and 300 years (e.g. Rooney et al. 2007). Laporte-Saumure et al. (2012) estimated metal fluxes and modelled leaching rates of lead suggesting a long-term threat to ground waters at a Canadian SAFR (>100,000 years). Based on these estimates the authors calculated an annual metal leaching rate of only 0.0001%, but it should be noted that the slightly alkaline soil pH (7.6-8.0) are not conducive the mobilisation of lead. The long-term potential migration of lead from shooting range soils to groundwaters at relatively low concentrations, but above backgrounds, is not considered to be an isolated outcome (e.g. Martin et al. 2013). Figure 4.2 shows the range of estimated timeframes over which lead from shot and bullets may leach from the soil surface to deeper into the soil profile.

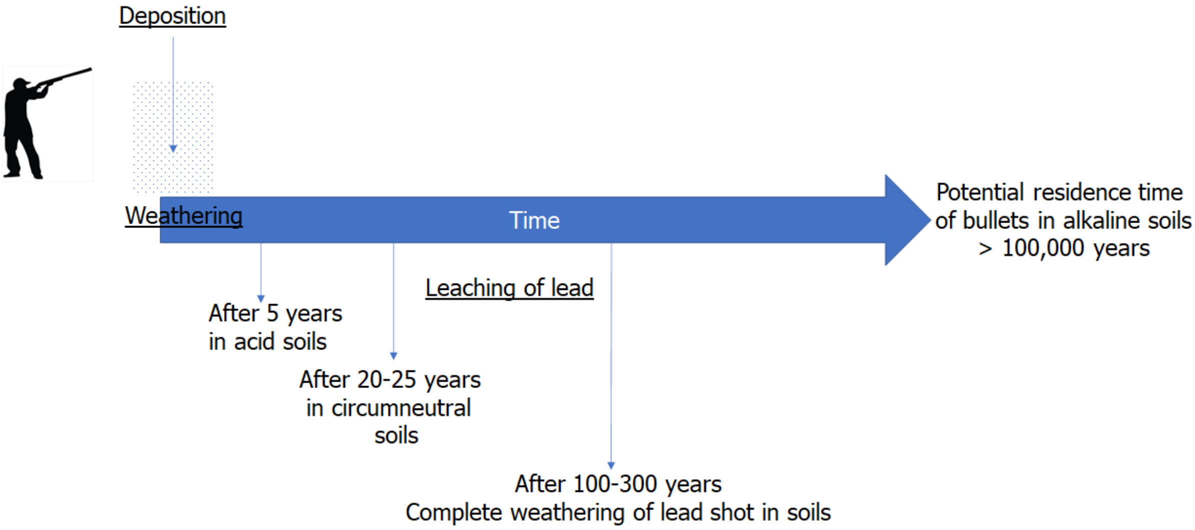

Figure 4.2 Estimated timeframe over which lead may be released from lead shot and bullets at shooting ranges.

Where elevated Pb leaching is expected, as indicated above substantial time lags may exist between mobilisation in the near-surface soil, plume breakthrough at the water table and migration of Pb in the saturated zone (e.g. to abstraction points). These lags may be of the order of several decades or substantially longer.

Lead bullets can fragment, and the smaller fragments may behave like lead shot. Generally, however, depending on the bullet construction (modern bullets tend to fragment more than older ammunition) the lead is present in larger forms than shot and so would be less exposed to weathering and potentially present a longer-term challenge (e.g. Barker et al. 2020).

In addition to the vertical migration of soluble lead and fine lead particles it is also important to consider the potential movement of weathering lead ammunition through the soil profile due to the accretion of organic matter at the soil surface. Selonen et al. (2012) report a downward migration rate of 2 to 3mm per year, but this biogeochemical recycling and movement is only likely in the most biologically active part of the soil profile (i.e. the upper horizons).

4.3 Site Assessment

Numerical modelling can, in principle, be used to describe the transport of Pb in different hydrogeological settings and, hence, predict groundwater vulnerability to contamination from shooting ranges. Different contaminant transport models exist from fairly simple conceptual representations (e.g. analytical solutions to the transport equations for simple scenarios: Lessoff and Indelman 2004) to highly mechanistic descriptions of water and solute dynamics in porous media (e.g. MODFLOW MT3D: Bedekar et al., 2016 and HYDRUS: Šimůnek et al., 2008; Trakal et al. 2013). Whilst such models can often simulate water transport phenomena reasonably well along with the transport of metals in simple scenarios (e.g. in lysimeters or in uniform materials), the accurate description of both the solute transport phenomena and the key geochemical processes which control Pb mobility (e.g. pH-dependent solubility and association with organic ligands) in field settings often remains challenging. This challenge may limit the extent to which models can describe dynamic interactions at specific sites with complex and variable hydrogeologies. Another limitation is that mechanistic models can be time consuming to set up and run for complex real-world scenarios and require considerable expertise and experience. Field monitoring is, therefore, still needed to provide firm underpinning evidence for the assessment of risks to groundwater receptors at sites with identified risk factors.

5 MITIGATION MEASURES

There are three main mitigation measure categories proposed in the literature as cited in the US EPA Best Management Practices (BMP) for Lead at Outdoor Shooting Ranges (US EPA 2005), and journal articles also follow these steps (for example Sanderson et al. 2018 and Dinake et al. 2019); control and containment, prevention of migration and removal and recycling.

A fourth step should also be implemented relating to an overarching environmental management plan, including documentation and record keeping and although this is not a mitigation in itself it is an important step in setting out and evaluating mitigation procedures. This section of the report will detail the mitigation measures covered by each sub-category, the reported effectiveness of these measures from literature and any adverse findings from their use. The Finnish Ministry of the Environment has also produced a report detailing Best Available Techniques (BAT) for the Management of the Environmental Impact of Shooting Ranges (Kajander and Parri 2014) and this is also considered.

5.1 Capture and containment

The main tenets of the capture and containment aspect of lead mitigation are in the setting up of the range appropriately, for example by the inclusion of backstops (berms), traps using shock absorbing concrete for bullets and shot containment zones.

In the US EPA (2005) guidelines it states that earthen backstops should be between 15 and 20 feet high with an angled slope as steel as possible, though no explicit recommendations are made regarding the material used though it does assume that soil is used[14]. The guidance does state however that if a backstop berm is to be replaced, or after soil clean-up, other alternative methods are suggested to be implemented prior to the reimposition of an earthen backstop (US EPA 2005). This recommendation is supported by the conclusion of ECHA (2021) who identified that an earthen berm is not sufficient on its own as a mitigation measure.

There have been recommendations to replace existing soil berms with sand, in particular, medium sand with no organic matter, as soil berms can increase weathering and mobility of lead (Sanderson et al. 2018; Fayiga et al. 2011) and the use of sand backstops is suggested as a measure by both the US EPA (2005) (Figure 5.1) and the Finnish Ministry of the Environment (Kajander and Parri 2014). Limestone or gravel has also been suggested as being included in the base of a backstop to break potential capillary action as a pathway for lead migration (US EPA 2005). Kajander and Parri (2014) have indicated that the use of a sand trap in conjunction with a liner comprising concrete, asphalt, bentonite or a plastic membrane can be more effective than an earthen backstop at mitigating water pollution. In addition, any lead-contaminated water can be collected allowing for monitoring of water quality and, if necessary, treatment. Replacement of sand from the top of the structure is possible; however, the implementation of this at existing ranges can be difficult (Kajander and Parri 2014). The use of a cover (or roof) over the backstop berm constructed from either metal, concrete or

[14] It does state that when using an earthen berm or backstop, ensure that the uppermost layer exposed to the shooting activity is free of large rocks and other debris.

wood has been shown to have positive impacts with respects to impacts on groundwater based on the inhibition of rainwater accessing the backstop, with the only negating factor for implementation being the increased cost of construction (Kajander and Parri 2014). There has also been suggestion that the use of paper-based targets would be beneficial over traditional metal targets, as it minimises the amount of course-sized fragments of bullets being produced (Fayiga et al. 2011).

The use of bullet traps is extensively discussed in the BAT report of the Finnish Ministry of the Environment (Kajander and Parri 2014). It evaluates the effectiveness of a variety of different bullet traps in comparison to a backstop berm for a variety of factors including recycling, this is summarised in Table 5.1.

Table 5.1 Comparison of bullet trap type to a backstop berm (from Kajander and Parri 2014)

	Dusting	Recycling	Noise	Ricochet hazard	Costs
Plate and pit	–	+	–	–	–
Venetian Blind	–	+	–	–	–
Escalator bullet trap	–	+	–	–	–
Snail trap	+	+	–	0	–
Total Containment trap	+	+	–	0	–
Simple steel bullet traps	–	+	–	–	+
Pipe trap	+	+	0	–	+
Container solution	+	+	+	–	+
Concrete bullet traps	+	–	+	+	–
Rubber grinding bullet traps	+	0	+	+	–

\+ feature in favour of implementation compared to backstop berm
– feature not in favour of implementation compared to backstop berm
0 no significant difference

The use of traps has been indicated as beneficial for the purposes of recycling; however, some traps increase the potential for 'dusting' and the majority of traps have an increased cost over a backstop berm though earthen backstop berms are not a sufficient mitigation measure on their own evidenced by the recommendations of US EPA (2005), Finnish BMP (Kajander and Parri 2014), and the recommendations of ECHA (2021). A summary of risk reduction measures at shooting ranges in Switzerland (detailed in BAFU 2020) describes the use of artificial bullet trap systems. In the artificial bullet trap system described by BAFU, the bullet penetrates the front plate of the bullet trap (usually made of plastic) and is braked inside a box containing a material such as rubber granulate that causes only a slight deformation of the projectiles, so that they remain largely intact. The bullets are then recycled after the granulate has been separated. In Switzerland it is estimated that around 2/3 of the lead fired with ammunition, which corresponds to 210 t per year, temporarily end up in artificial bullet traps. It is of note that the utilisation of bullet traps is now mandated in Switzerland if compensation for assisting in decontamination is sort (FOEN 2020) and was also proposed as the preferred RMM in the ECHA Annex XV restriction report (2021).

At clay target ranges the US EPA (2005) recommend that measures are undertaken to reduce shot fall zones. Kajander and Parri (2014) agree with this recommending the use of terrain contouring and backstop berms; as an example, at the Lonato shooting range 96 % of fired shot can be recovered. By implementing these zones, the area subjected to pollutant load is reduced, though the total load is unchanged unless shot is regularly removed (Kajander and Parri 2014). Nets and barriers have also been successfully implemented at shotgun ranges, although the commercial availability of these barriers was poor the time the report was produced (Kajander and Parri 2014). An example of this technique is at the Baden-Würtremberg site detailed in a summary of risk management measures provided by the Bavarian LfU (2014); this 'shotnet' system uses nets erected ~67m from the shooting area to capture the shot so that it falls into a tub. Partial cost-recovery comes from the sale of recovered lead shot to scrap dealers.

5.2 Prevention of Migration

5.2.1 Horizontal barriers

Horizontal barriers or membranes made of plastics, geotextiles or asphalt can be used at clay target shooting ranges to prevent lead shot becoming entrained in the soil surface and as part of a strategy for lead recovery (Bavarian LFU, 2014; Kajander and Parri 2014). Use of this type of impermeable barrier is only appropriate after contaminated soil has been removed as it does not prevent percolation and can encourage the development of anaerobic soil conditions (ECHA, 2021).

5.2.2 Chemical Stablisation

Chemical stabilisation is an important aspect of prevention of migration, and several procedures (such as phosphate application and adjustment of soil pH by liming) have been well documented with many years of application in the field. Other soil stabilisation techniques have been assessed in the field and in laboratory-based experiments and these are briefly covered below.

The US EPA has recommended the application of liming agents and phosphate to control the migration of lead at shooting ranges (US EPA 2005). An application rate of 15 to 20 pounds of phosphate per 1,000 square feet is recommended to effectively control lead, either via the application of "pure" phosphate or as part of a lawn fertiliser; this is proposed where lead is widely dispersed in soils across the range, when a range is closing or if there is a high potential for vertical lead transport to groundwater and in particular for sporting clay ranges and areas that are not easily accessible by reclamation equipment (US EPA 2005). Liming is recommended to adjust soil pH to the range of 6.5 – 8.5, with spreading of lime around the earthen backstops, sand traps, trap and skeet shotfall zones, sporting clays courses and any other areas where the bullets/shots or lead fragments/dust accumulate (US EPA 2005).

However, there have been several reviews that question the effectiveness of lime and phosphate stabilisation. Butkus and Johnson (2011) noted that variations in the form of phosphate and lead present in a system can affect the products formed and consequently their relative mobility in natural systems. In a series of column experiments PO_4(aq) retarded the mobility of Pb(aq) and particulate PbO principally due to the formation of pyromorphite; however, they note that the practice of phosphate application and liming may result in an insufficient reduction in lead transport. For example, the presence of particulate hydroxyapatite increased the mobility of PbO at pH 7.2 relative to the control (Butkus and

Johnson 2011). The authors note that due to batch experiments being used, it is plausible that the rate of transformation of HA treatments might be slower in the field, and this would exacerbate the limitations of HA; thus, they recommend that the practice of using HA in sandy firing range soils, under low to neutral pH conditions, be reconsidered (Butkus and Johnson 2011). Subsequently, further studies have been conducted on the use of phosphate for chemical stabilisation, and the results are summarised in Table 5.2.

Table 5.2 Summary of recent research on chemical stabilisation of shooting range soils with phosphate (modified from Sanderson et al 2018)

Amendment	Application Rate	Findings
Phosphate rock, phosphoric acid	4:1 P:Pb	TCLP Pb reduced from up to 800 mg L^{-1} to <1 mg L^{-1}
Phosphate alkaline residue	0 – 20 %	TCLP Pb reduced from >100 mg L^{-1} to <5 mg L^{-1} Bioaccessible Pb reduced 20–70%
Phosphate, lime, MgO, red mud	2:1 P:Pb, Lime, red mud and MgO 2%	Pb bioaccessibility reduced by 20–55%
Phosphoric acid, MgO	1% P, 10% MgO	XAS-pyromorphite formation up to 38%. P + MgO reduced bioaccessibility by up to 25%
Phosphate coating	Bullet surface coating	Leachable Pb was reduced by 77–98% by $FePO_4$ or $AlPO_4$ surface coating

It is also of note that phosphate amendments may themselves leach and migrate, contaminating areas off-site. This can occurif phosphate is applied in excess, and can potentially contaminate ground or surface water (Scheckel et al 2013).

The review of Sanderson et al (2018) also included studies that have been performed using amendments ranging from calcium phosphate nanoparticles, biochar, bone and other materials (Table 5.3).

Table 5.3 Summary of recent research on chemical stabilisation of shooting range soils with alternative amendments (modified from Sanderson et al 2018)

Amendment	Application Rate	Findings
$Ca_3(PO_4)_2$ nanoparticles	5 %	$CaCl_2$-extractable Pb by > 90%
Mussel shell, cow bone and biochar	5 %	Maize uptake reduced by up to 71% Pb
Hydroxyapatite and ferrihydrite	5 %	Water-soluble lead by 99.9 %
Ferric oxyhydroxide with limestone	1 - 4 %	Water- and 1 M NH_4NO_3-extractable Pb reduced by 89–99%
Red mud, zero valent iron, iron sulphate	1:19 Fe:Soil, 1:4 red mud:soil, 2% goethite	Pb leaching reduced from > 700 to < 10 µg kg^{-1}
Biochar, iron oxide, gibbsite, silver nanoparticles	5% biochar, 0.1% iron oxides and nanomaterials	Pb extractability reduced by 13 – 94%

Amendment	Application Rate	Findings
Biochar	10 %	Exchangeable Pb reduced by 88.08%
Cow bone powder, biochar, egg shell powder	5 %	Water-soluble Pb in amended soil significantly decreased with saline water irrigation
Biochar, carbon nanotubes	0 – 2.5 %	BC reduced the concentrations of Pb in the soil by 17.6 %

Table 5.3 shows that there have been promising findings from the use of alternatives to solely liming and phosphorous treatments, for example, biochar reduced exchangeable Pb by 88%. However, the studies detailed in Table 5.3, are typically pilot schemes or bench scale experiments and further research is required into the viability of these amendments on a large scale; though it is of note that some studies have been conducted for up to 4 years in a field based setting.

5.2.3 Water Management and Treatment

Best management techniques recommended by the Finnish Ministry of the Environment also recommend water management, and where required water treatment (Kajander and Parri 2014). The type of water management implemented and the type of treatment required depends on the layout and the permeability of the shooting range; for example at pistol and rifle ranges, this would include redirection of waters outside the range area past the range area, and the collection of water from the backstop berm and, in some cases, also the intermediate area and the firing stands though this is typically not required when bullet traps or a covered berm are used (Kajander and Parri 2014). At sites with low water permeability, water can be collected from around a traditional backstop berm with open ditches and underground drainage, where it can be monitored and treated as required; however, at locations with high water permeability collection from around a traditional backstop berm with open ditches is extremely difficult as the water is absorbed into the soil. In these situations, percolating water can only be collected by the inclusion of a sand trap that includes a watertight surface directed to underground storage (Kajander and Parri 2014). The Finnish Ministry of the Environment recommend that water can be treated via filtration or sedimentation though it does state that reliable research data is not available on the functional effectiveness of sedimentation basins, wells and ditch systems at shooting ranges (Kajander and Parri 2014); an example filtration system is presented in Figure 5.1.

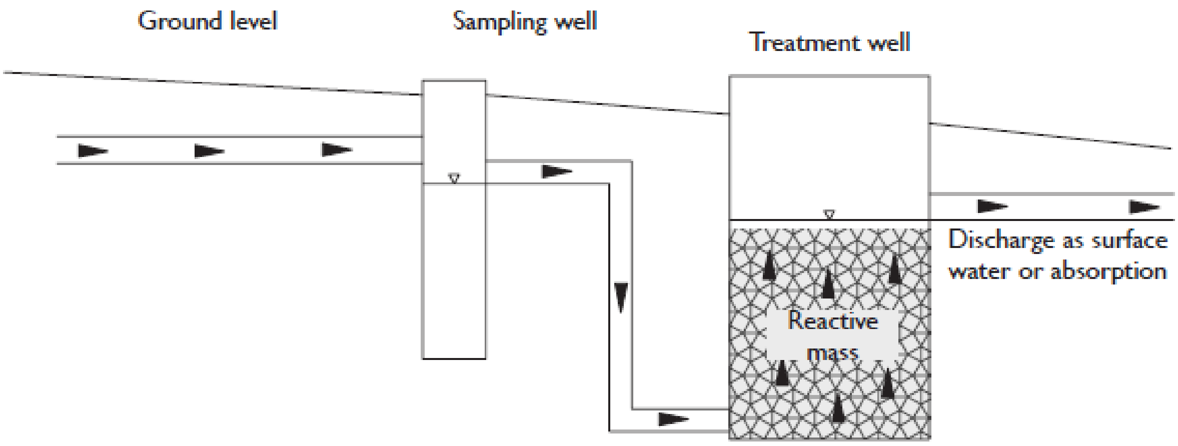

Figure 5.1 An example filtration system for water collection at pistol and rifle ranges (from: Kajander and Parri 2014)

Mariussen et al. (2012) performed several experiments using a variety of sorbents including charcoal and olivine sand added to iron powder in column experiments with water from a creek located at a shooting range in Norway. Charcoal was found to be the most effective means of reducing Pb and Cu in the drainage water; and they recommended combining sorbents in sequences to increase both the selectivity and efficiency of a sorbent as well as highlighting that Fe^0 also may be effective as a reactive material in an oxidative environment. The authors noted that the filters appeared to completely remove the episodic increase in the metal concentrations in the creek, though the sorption efficiency was reduced during low flow periods with low metal concentrations and that all of the sorbents tested increased the pH level in the outflow water (Mariussen et al. 2012).

5.3. Removal and recycling

The removal and recycling of bullets from shooting range soils has the potential to substantially reduce contaminant burden and environmental and human health risk; and removal is step 3 in the procedures recommended by the US EPA (Figure 5.1), examples of removal processes include raking, sifting, screening, vacuuming, soil washing, reclamation, and recycling (US EPA 2005). Screening is also recommended by the Finnish Ministry of the Environment, with berm renovation recommended after 10,000 rounds fired per shooting stand or every three to five years (Kajander and Parri 2014). The authors concluded that when correctly used, screening/sieving is an effective method of reducing the metal load in the range area and thus the risk of spreading, though they do state that there are some uncertainties inherent in the use of the method (Kajander and Parri 2014). However, caution should be exercised as mechanical removal of Pb may result in abrasion of Pb fragments and enrichment of Pb in soil (Sanderson et al 2018). Yi et al. (2010) investigated the effect of a sand berm, liming and bullet removal and observed that use of a sand berm and liming reduced Pb concentrations significantly due to lower moisture levels and organic content as well as higher pH although bullet removal transferred 2.5 times more total Pb to the soil due to abrasive action.

Other proposed solutions also include mass replacement at impact areas and removal of bullet scrap and soil in their entirety, these methods and a comparison to the soil screening approach are provided in Table 5.4.

Table 5.4 Summary of backstop berm renovation measures (from: Kajander and Parri 2014)

Technique	Description	Pollutant management (effectiveness and reliability, generation of water with pollutant content, generation of dust with pollutant content)	Availability / viability	Assessment on the method's suitability in the management of the environmental impact of shooting ranges
Mass replacement at impact areas	The soil in the impact areas containing the most bullet scrap is removed regularly. The removal interval depends on the number of shots, recommended 3...5 years.	Significantly reduces the load on the range structures. Particularly effective at new ranges when used regularly, allowing the removal of the most significant part of the bullets. At old ranges, some of the load is often deeper in the backstop berm and not affected by the technique.	Good. Mass replacement of the entire impact area may be difficult with regard to excavation technology and requires planning.	Suitable for pistol and rifle ranges where the bullets accumulate in the impact areas. Often expensive on the long term.
Screening of the impact areas	The soil in the impact areas containing the most bullet scrap is removed regularly. The screening interval depends on the number of shots, recommended 3...5 years. The bullets are screened out of the soil that can then be returned to the structure or disposed of as waste. The bullets can be recycled.	Effective at new ranges when used regularly, allowing the removal of the most significant part of the bullets. Questionably effectiveness at old ranges. Fine-grained metal remains in the berm, and disturbing the soil may increase the solubility of the metals. The spread of dust with metal content must be controlled.	Good. Can be carried out mechanically using different techniques, or manually. Screening of the entire impact area may be difficult with regard to excavation technology and requires planning.	Limited suitability for pistol and rifle ranges where the bullets accumulate in the impact areas. At old ranges, there is the risk of the metal particles attached to the soil become mobile. Most usable at new ranges at sites where the reduction of load is considered to be a sufficient measure.
Removal of bullet scrap and soil in their entirety	The contaminated soil containing bullet scrap is removed and transported away from the area. Requires quite extensive earthmoving work. The soil and bullet scrap can be separated by screening.	Effective management of pollutants. Eliminates the need of water management when carried out regularly. The mass replacement work causes some dust generation. Regularly causes the contamination of clean soil brought to the site.	Good/moderate. Requires a plan made by an expert. Mass replacement requires quite extensive earthmoving work.	As a risk management method, effective in principle, but an expensive solution that has poor eco-efficiency.

The study of Fayiga and Saha (2016) noted that bullet removal reduced total soil Pb in all cases except for one unvegetated soil; increased bioavailability of Pb in un-vegetated soils; and increased DOC concentration in the leachates based on a series of batch experiments in the laboratory. Variations were observed between soils, with vegetation reduced leaching of Pb in two soils and a combination of bullet removal and vegetation significantly reduced leaching of Pb in a third (Fayiga and Saha 2016). Vegetation increased water soluble Pb thus increasing Pb mobility in the soil and it was postulated that the addition of a chemical stabilizer to immobilize Pb may a potential solution as vegetation is needed as ground cover (Fayiga and Saha 2016). In the study utilising St Augustine grass, high concentrations of Pb were accumulated by increasing Pb availability in the rhizosphere with a very high proportion of the

Pb in the grass sequestered in the root zone. In the experiments conducted with unsieved soils with bullets the grass had higher plant biomass, which suggests a tolerance to Pb (Fayiga and Saha 2016).

Lafond et al. (2014) performed a lab study to evaluate the performance of a counter-current leaching process (CCLP) with leachate treatment to remove metals including Pb (3,368 mg Pb kg^{-1}) from a moderately contaminated shooting range soil determined an average removal yields of 92% for Pb. The authors concluded that the study showed that CCLP can be successively used for the remediation of moderately metal-polluted shooting range soils, though they admit that the technology would need trialling at a larger scale and using more heavily polluted soils (Lafond et al 2014).

5.4 Environment management plans

Management strategies are recommended by the both the US EPA and the Finnish Ministry of the Environment for the control and mitigation of potential contamination at shooting ranges. According to the US EPA the plans should document all best management practices implemented, including recycling of lead, including what was performed, when and by whom, and should be kept for the lifetime of the range (2005). The ITRC (2005) 'Technical Guideline for Environmental Management at Operating Outdoor Small Arms Firing Ranges' also includes a comprehensive template for an Environmental Management Plan that could be used by SAFRs.

Sanderson et al (2008) also state these plans should seek to reduce the weathering of bullets in the soil and limit the mobility and bioavailability of contaminants accounting for any site-specific characteristics. Both Sanderson et al. (2018) and Kajander and Parri (2014) state that the plan must include information required to meet the Finnish environmental permitting requirements and use the acceptable emission values for planning the required risk management measures.

5.5 Costs of remedial measures

No information was obtained from the stakeholder consultation on the costs of risk management measures applied at shooting ranges to prevent the migration of lead to groundwater due to a lack of response.

The costs associated with the implementation of mitigation measures are highly variable depending on the measures implemented. Kajander and Parri (2014) detail the approximate costs associated with implementing various techniques (Table 5.5), also detailed in the Annex XV report (ECHA 2021).

Table 5.5 Cost estimate for the implementation of mitigation measures at shooting ranges (from: Kajander and Parri 2014)

Mitigation Measure	Cost (€)
Mechanical screening	2,000 – 5,000 at a 20-stand range
Commercial bullet traps	13,000 – 44,000 per stand over a 20-year period
Berm construction	~90,000[1]
Netting at shotgun range with a berm	300,000+[2]
Berm covering	260,000[3]
Sand trap structure with liner	40,000 – 50,000 at a 20-stand range
Water collection and treatment	5,000+ (excluding granules)
Surfacing of a shotgun range with asphalt	150,000

[1] Berm 20m high at a shotgun range
[2] Costs of a berm and net combination 23 metres high
[3] Length 50 metres, width 21 metres, column interval 5 metres

The Bavarian Environment Ministry (Bavarian LFU, 2014) have outlined practical examples of environmental risk management measures at target shooting ranges, including some details of the costs involved. Table 5.6 details the management measures and associated costs at specific sites, but it should be noted that the costs likely include remedial works and only relate to the contribution made by the Bavarian LFU.

Table 5.6 Summary of environmental risk management measures at shooting ranges in Bavaria, Germany (Bavarian LFU 2014)

Site	Risk management measures	Cost and measures covered	Comments
Schützenverein St Sebastianus Aschaffenburg 1899	recovery of lead-shot (shot gutters) followed by their removal (in the future twice per year in spring and autumn)	€0.5M plus contribution by the shooting club. Total redesign of the facility, including soil remediation and modernisation of the shooting lanes	Shooting range in existence since 1974. Building works began in 2009 and the whole range was redeveloped. 25t of lead recovered during remedial works
Bayer. Jadgschutz- und Jägerverin e. V. Lichtenfels	Collecting/gathering up of waste annually. Reuse of **iron shot** via local scrap-dealers	Approx. €0.26M plus contribution by the shooting association. Total redesign of the clay target shooting range (including soil remediation)	Shooting range in use since 1935. Environment-focussed redevelopment took place between 2003 and 2005
'Friesenheimer Insel, Mannheim',	To catch the shot the 'shotnet' system was selected. The net catches the shot so that it falls into a tub.	Approx. €1.35M (50% funding from the State) plus the	In use since 1910 and modified in 1934. Environment-focussed redevelopment took

Baden-Würtremberg	The nets are erected approx. 67m from the shooting areas. Manual annual collection of lead shots with broom, reuse/recycling of lead shots through scrap dealers. Monitoring not considered necessary as the soil in the fall zone of the shot is completely covered with netting.	association's own contribution. Total redesign of the clay target shooting range (including soil remediation)	place between 2005 and 2010. Lead shot was found in depths of up to 0.25m depth. ~50,000 shots fired on a yearly basis. Collected lead shot sold to scrap dealers
Landesjägerschaft Niedersachsen e.V.	Old sea containers (placed on top of each other) were used to build the lead shot catching system, reaching 18 – 20m in height. The surface of the shooting side of the containers were covered in wood and textile fabric	Approx. €1.3M Total redesign of the clay target shooting range (incl. soil remediation)	In existence since 1969. During WWII the site was used as an ammunition depot. The alterations took place in 2004/5 ~90 t of lead-shot deposits were found in a depth of 0.1m
Schießstand Oberg e.V	Remediation (soil removal) followed by twice yearly monitoring of the soil	Approx. €0.75M plus the club's own contribution. Total redesign of the facility (target shooting range including soil remediation and modernization of the shooting range)	In existence since 1960 Site examined in 2005. Lead shot was found in layers of ground ranging up to 0.2 m and in places 0.4 m.

6 CONCLUSIONS

The soils at shooting ranges carry a very large load of lead compared with local ambient background concentrations and concentrations of up to 30-40% Pb have been measured in soil at some sites. At rifle and pistol ranges the highest concentrations of lead resulting from bullets and bullet fragmentation are found in the backstop berms behind the targets, whereas at clay target ranges the lead shot is deposited across the soil surface in the 'drop zone' beneath the target area.

The weathering of shot and bullets to more soluble forms of lead has been well studied and shown to be greater under acidic soil conditions and under vegetation, especially trees. The highest concentrations of lead are at the surface or in the upper layer of the soil profile and generally decrease rapidly with depth. There are some limited data that show elevated concentrations of lead in subsurface layers and in groundwaters. Significantly, soil water concentrations of lead in subsurface layers can show concentrations in the low mg L^{-1} range (up to 12.6 mg L^{-1} has been reported), which is several orders of magnitude above the drinking water standard for lead (10 µg L^{-1}). The concentration of lead in soil water has though been observed to rapidly decrease with depth and to date only a few studies have measured elevated lead in groundwater, with these being in near surface groundwaters under acidic soils (in areas that would generally be considered as wetlands).

Mobility of lead in soil occurs to a greater extent in conditions where processes of natural attenuation are reduced and loading of lead is relatively high. The factors that promote movement of lead from surface layers tend to be those that accelerate the weathering of the shot and bullets, such as acidic and organic rich soils with coarse soil texture and low iron, manganese and phosphate content.

Lead can be transported in soils, the vadose zone and in groundwater as either particles (e.g. small lead fragments from projectiles or lead sorbed to mobilised soil particles and organic colloids) or in solution. The connectivity between near surface soil, the vadose zone and underlying groundwaters is dependent on a combination of factors, such as soil texture (which will affect soil hydraulic properties and drainage characteristics), soil chemistry and organic matter content (which will affect lead mobility), soil structure (particularly the existence of preferential flow pathways), geology (particularly the existence of low permeability deposits such as glacial drift and the nature of dominant flow pathways in the vadose and saturated zones – specifically intergranular versus fracture flow), topography, climate (particularly the magnitude of average annual precipitation compared with average annual evapotranspiration) and the depth to the saturated zone. Shallow groundwater (e.g. on floodplains) is especially vulnerable to contamination but may also enhance lead mobility in the soil if water tables seasonally inundate near-surface horizons with high lead concentrations. This issue may be exacerbated in the future in parts of Europe which are predicted to have wetter winters under climate change

Groundwater vulnerability is defined by characteristics of the hydrogeological setting that affect the ability of contaminants at the soil surface to reach the water table. This vulnerability is usually separated into two components: (i) intrinsic vulnerability, which combines the principal physical factors influencing general contaminant transport to groundwater and (ii) specific vulnerability, which considers factors affecting the availability of the contaminant of interest, such as the size of the contaminant source and its mobility. An example of a setting with high intrinsic vulnerability would be an unconfined aquifer with a shallow water table, fractured calcareous geology (e.g. Karst) and thin, high permeability soils. In contrast, a low vulnerability setting might have deep groundwater overlain by thick, fine-textured soils and or low-permeability glacial drift deposits, such as boulder clay which can as a barrier to the migration of pollutants through the unsaturated zone.

Based on consideration of soil chemistry and groundwater vulnerability, the key factors influencing the mobilisation of lead and its potential to migrate through the vadose zone to groundwater are:

- Acidic soil (pH <6) with relatively high organic matter content and low iron, manganese and phosphate content;
- Coarse (usually sandy) soils that allow vertical migration of dissolved or fine particulate lead;
- Preferential flow pathways, including or 'fingered flow' in the soil matrix but more importantly macropore flow down soil cracks, plant root channels and animal burrows, along with fissured flow in the underlying vadose and saturated zone;
- Shallow depth to groundwater.

Specific groundwater vulnerability will also, clearly, be affected by the lead emission rate and historical loadings, driven by usage (e.g. number of rounds or amount of lead shot per day and the time over which the area has been used as a range).

Several of the conditions enhancing the mobilisation and downward migration of lead in shooting range soils (e.g. acidic pH and preferential pathways such as tree roots) are found in forested areas, which have been identified in a significant proportion number of the studies on shooting ranges reviewed for this report. Groundwater is commonly abstracted as drinking water across the EU and private wells in close proximity to (and at lower hydraulic potentials to) shooting ranges represent a particularly high risk because as they are often relatively shallow and are not regularly checked for water quality. The characteristics of shooting range sites susceptible to vertical migration of lead from soil to groundwater are detailed in the refined conceptual site model presented below.

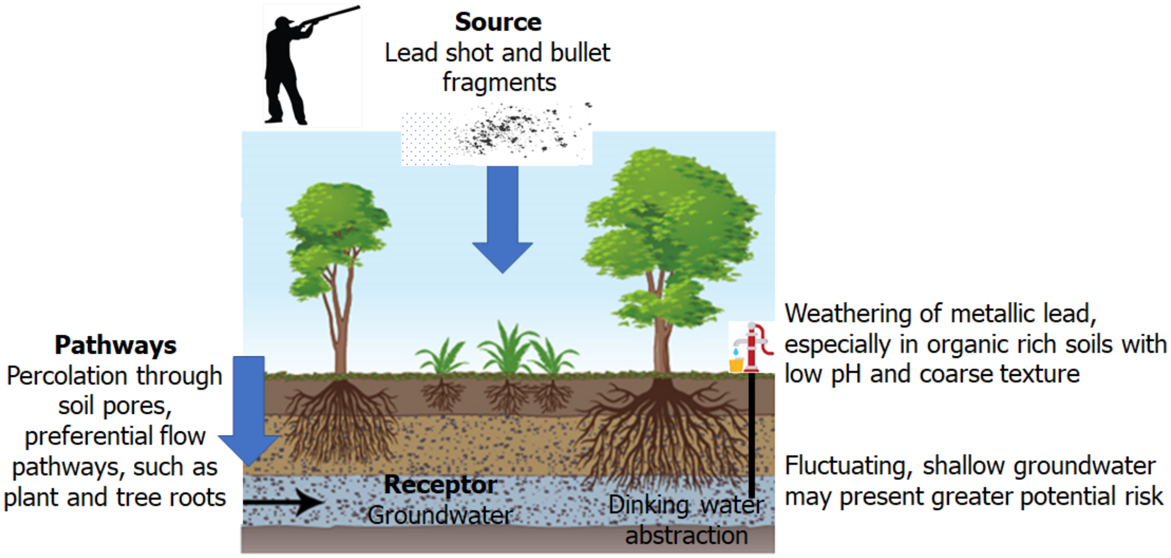

Figure 6.1 Refined conceptual site model based on findings of literature review

It is difficult to estimate the prevalence and extent of groundwater vulnerability to lead contamination at shooting ranges at European, national or even regional levels because many of the contributing factors are local and difficult to predict at wider geographical scales. Whilst there are zones of high intrinsic groundwater vulnerability in some regions, there may be localised situations elsewhere which also present high risk. Thus, although high specific groundwater vulnerability for lead from firing ranges is probably limited to a small fraction of total sites in Europe, this fraction may not be insignificant.

Local factors will always influence potential risks more than generic considerations but areas with high intrinsic vulnerability are likely to occur in all EU member states, although to differing extents. Detailed GIS analysis would be required to estimate the areas with high vulnerability which are also areas classed as aquifers. Information on the fraction of shooting ranges occurring in these high vulnerability areas would provide quantitative information on the extent of the risk and requirements for associated risk management.

Migration of lead to groundwater is probably more likely to occur at clay target shooting ranges where shotguns are used because shot discharges over a wider area resulting in more widespread areas of contamination with highly elevated concentrations (up to 30-40% Pb in surface soil and estimated site loadings of up to 40 t of lead over the operational lifetime of a shooting range). Furthermore, the lead shot can become entrained in the soil surface, ultimately becoming buried due to the accumulation of organic matter, particularly in forest areas. This is in contrast to rifle and pistol ranges where shooting activity is more focussed on fixed targets using bullets that have a smaller surface area:mass ratio, although this can increase due to fragmentation. In addition, bullets and associated fragments at small arms firing ranges are generally retained in bullet traps or berms, and specific sand traps may be in place that prevent lead from leaching to soil.

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Modelling of solute transport phenomena and the key geochemical processes which control Pb mobility (e.g. association with organic ligands) within complex and variable hydrogeologies is possible but remains challenging at a site-specific level. This means that field monitoring is still needed to provide firm underpinning evidence to support the identification of a substantial risk of lead contamination to groundwater. The difficulties of making accurate quantitative predictions for both weathering and subsurface transport of lead also frustrate accurate estimates of the timeframes over which the movement of lead from surface to groundwater will occur. Estimates in the literature range from 10's to 100,000's of years, with variations in rates depending on the type of ammunition used, soil conditions, intrinsic and specific groundwater vulnerability and the distance between the shooting range and the point of groundwater abstraction for drinking water.

Various risk management practices are employed at shooting ranges to reduce lead contamination of soil and prevent the migration of lead to groundwater; with preferred measures being those that prevent contact of the lead ammunition with soil. Containment strategies involve the inclusion of backstops (berms), bullets traps using shock absorbing materials and shot containment zones. It is now recommended that berms are constructed using sand rather than soil and to exclude organic matter as this can increase weathering and mobility of lead. Nets and barriers have also been successfully implemented at shotgun ranges to catch and retain pellets. Chemical stabilisation is used to prevent migration of lead in soil and several procedures (such as phosphate application and adjustment of soil pH by liming) were previously established practices although their effectiveness has been called into question and overuse of phosphate can present a contamination risk in itself. The removal and recycling of bullets and shot from shooting range soils is also employed to reduce the contaminant burden and associated risk to the environment; examples of removal processes include raking, sifting, screening, vacuuming and soil washing. Reclaimed lead can then be sold for scrap value to offset the costs of the removal process. Indicative prices for specific risk mitigation measures have been identified in the literature but the costs of employing these measures at individual shooting ranges (on either a one-off basis or annual basis) were not determined during this study due to a lack of response to the stakeholder consultation exercise.

REFERENCES

Aller L, Lehr JH, Petty R. 1985. DRASTIC: A standardized system for evaluating ground water pollution potential using hydrogeologic settings. National Water Well Association/EPA Series EPA-600/2-85/018. 57 pp.

Austrian UBA (Umweltbundesamt). 2002. “Tontaubenschiessplatz Kuchlmuhle”

Austrian UBA (Umweltbundesamt). 2018. Altlast S 15 “Schießplatz Glanegg“ Beurteilung der Sanierungsmaßnahmen

BAFU (Studie im Auftrag des Bundesamts fur Umwelt). 2020. Blei in der Schweiz: Verwendung, Entsorgung und Umwelteinträge. Accessed online at https://www.aramis.admin.ch/Default?DocumentID=67532&Load=true 8 July 2021

Barker AJ, Mayhew LE, Douglas TA, Ilgen AG, Trainor TP. 2020. Lead and antimony speciation associated with the weathering of bullets in a historic shooting range in Alaska. Chemical Geology. https://doi.org/10.1016/j.chemgeo.2020.119797

Bavarian LFU 2014. Technische Hinweise zum umwelt-verträglichen Bau und Betrieb von Wurfscheibenschießanlagen. Bayerisches Landesamt für Umwelt. Available at: https://www.bestellen.bayern.de/application/applstarter?APPL=ESHOP&DIR=eshop&ACTIONxSETVAL(index_portal.htm,USERxPORTAL:TRUE,ALLE:X)=X.

Bavarian WWA ASCHAFFENBURG 2019. Schießanlage Miltenberg OT Mainbullau; Anfrage auf Datenauskunft vom 16.06. und 28.07.2019. Wasserwirtschaftsamt Aschaffenburg. Available at: https://www.stadtwatch.de/app/download/9828581984/Me%C3%9Fwerte%20Schie%C3%9Fanlage%20Mainbullau%20Auskunft%20v.%2031.10.2019_geschw%C3%A4rzt.pdf?t=1573484834

BCCDC (British Columbia Centre for Disease Control) 2011. Lead from firing range and the potential to contaminate drinking water supply. Accessed online at https://ncceh.ca/sites/default/files/BCCDC-Lead_Shot_Drinking_Water_Nov_2011.pdf 8 July 2021

Bedekar, V., Morway, E.D., Langevin, C.D., and Tonkin, M., 2016, MT3D-USGS version 1: A U.S. Geological Survey release of MT3DMS updated with new and expanded transport capabilities for use with MODFLOW: U.S. Geological Survey Techniques and Methods 6-A53, 69 p., http://dx.doi.org/10.3133/tm6A53

Beven K, Germann P. 1982. Macropores and water flow in soils. Water Resources Research, 18: 1311-1325.

Butkus MA, Johnson MC. 2011. Reevaluation of Phosphate as a Means of Retarding Lead Transport from Sandy Firing Ranges. Soil and Sediment Contamination, 20:172–187, 2011

Carey M, Thursten N, Phillips N. 2017. New groundwater vulnerability mapping methodology in England and Wales. Environment Agency of England and Wales, Report – SC040016/R, Bristol, UK (ISBN: 978-1-84911-318-2)

Clausen J, Korte N. 2009. The distribution of metals in soils and pore water at three u.s. military training facilities. oil and Sediment Contamination: An International Journal. 18: 546-563.

Clausen JL, Kaste J, Ketterer M, Korte N. 2010. Sample preparation and digestion considerations for determining metal deposition at small arms ranges. International Journal of Environmental Analytical Chemistry, 90: 903–921.

Clausen JL, Bostick B, Korte N. 2011. Migration of lead in surface water, pore water, and groundwater with a focus on firing ranges. Critical Reviews in Environmental Science and Technology. 41: 1397-1448.

Daly D, Dassargues A, Drew D, Dunne S, Goldscheider N, Neale S, Popescu C, Zwhalen F. 2002. Main concepts of the “European Approach” for (karst) groundwater vulnerability assessment and mapping. Hydrogeol. Journal, 10: 340–345.

Dewey C, Bargar JR, Fendorf S. 2021. Porewater lead concentrations limited by particulate organic matter coupled with ephemeral iron(III) and sulfide phases during redox cycles within contaminated floodplain soils. Environmental Science and Technology. 55, 5878−5886.

Dinake, P., Kelebemang, R. and Sehube, N. (2019) ‘A Comprehensive Approach to Speciation of Lead and Its Contamination of Firing Range Soils: A Review’, Soil and Sediment Contamination: An International Journal, pp. 431–459

Dortch MS, Johnson BE, Jeffrey GA. 2013. Modeling fate and transport of munitions constituents on firing ranges. Soil and sediment contamination, 22: 667-688.

EC (European Commission). 2014. Framework for action for the management of small drinking water supplies.

https://ec.europa.eu/environment/water/water-drink/pdf/Small%20drinking%20water%20supplies.pdf Accessed 28/07/2021

EC (European Commission). 2016. *WFD Reporting Guidance 2016*. Final Draft 6.0.6. European Commission. 402 pp. http://cdr.eionet.europa.eu/help/WFD/WFD_521_2016/Guidance/WFD_ReportingGuidance.pdf Accessed 7/7/2021

EC (European Commission). 2020. Revised Drinking Water Directive. European Commission https://ec.europa.eu/environment/water/water-drink/legislation_en.html Accessed 12/07/2021

EC (European Commission). 2021a. *Groundwater as a resource.* European Commission. https://ec.europa.eu/environment/water/water-framework/groundwater/resource.htm Accessed 7/7/2021

EC (European Commission). 2021b. Groundwater quality and vulnerability: Assessment tools to prevent and control groundwater pollution by nitrates. https://water.jrc.ec.europa.eu/groundwater. Accessed 27/6/2021

ECHA (European Chemicals Agency). 2021. Annex XV Restriction Report, Proposal for a Restriction on Lead. Version 2.0 24 March 2021. ECHA, Helsinki, Finland

EEA (European Environment Agency). 2019. *Use of freshwater resources in Europe.* European Environment Agency. https://www.eea.europa.eu/data-and-maps/indicators/use-of-freshwater-resources-3/assessment-4 Accessed 7/7/2021

EFSA (European Food Safety Authority). 2010. Scientific Opinion on lead in food. EFSA Panel on Contaminants in Food Chain (CONTAM). EFSA Journal 2010; 8 (4): 1570, 151 pp.

ETH (Swiss Federal Institute of Technology Zurich). 2002. Blei- und Antimonbelastung bei Schiessanglen. Fallbeispiel Luzerner Allmend. Zurich, February 2002. Accessed online at https://www.research-collection.ethz.ch/bitstream/handle/20.500.11850/146364/eth-25320-01.pdf 8 July 2021.

Fayiga AO, Saha U, Ma LQ. 2011. Chemical and physical characterization of lead in three shooting range soils in Florida. Chemical Speciation & Bioavailability. https://doi.org/10.3184/095422911X13103191328195

Fayiga AO, Saha U. 2016. The effect of bullet removal and vegetation on mobility of Pb in shooting range soils. Chemosphere. 160: 252-257.

FOEN (Ed.) 2020. VASA payments for shooting ranges. Notification from the FOEN as the enforcement authority. 4. Updated Edition 2020; First edition 2006. Federal Office for the Environment, Bern. Environmental enforcement No. 0634: 36 p

Garrido F, Helmhart M. 2012. Lead and soil properties distributions in a roadside soil: Effect of preferential flow paths. Geoderma, 170: 305-313.

Hockmann K, Tandya S, Studer B, Evangelou MWH, Schulin R. 2018. Plant uptake and availability of antimony, lead, copper and zinc in oxic and reduced shooting range soil. Environ Pollut., 238: 255-262.

Hui N, Jumpponen A, Niskanen T, Liimatainen K, Jones KL, Koivula T, Romantschuk M, Strommer R. 2011. EcM fungal community structure, but not diversity, altered in a Pb contaminated shooting range in a boreal coniferous forest site in Southern Finland. FEMS Microbiol Ecol 76: 121–132.

Hulsmann A, Hulsmann E. 2005. Small systems large problems: A European inventory of small water systems and associated problems. Nieuwegein, Web-based European Knowledge Network on Water (WEKNOW).

ICMM. 2007. MERAG: Metals Environmental Risk Assessment Guidance. ICMM, London.

ITRC (Interstate Technology and Regulatory Council) 2005. Environmental management at operating outdoor small arms firing ranges. Washington, DC: ITRC, Small Arms Firing Range Team. Available from: https://connect.itrcweb.org/HigherLogic/System/DownloadDocumentFile.ashx?DocumentFileKey=83c01740-bc23-4c71-9c81-7e650be98058 (Accessed 8 July 2021)

Islam MN, Nguyen XP, Jung HY, Park JH. 2016. Chemical speciation and quantitative evaluation of heavy metal pollution hazards in two army shooting range backstop soils. Bull Environ Contam Toxicol., 96:179–185.

Janik LJ, Forrester ST, Soriano-Disla JM, Kirby JK, McLaughlin MJ, Reimann C, GEMAS Project Team. 2015. GEMAS: prediction of solid-solution partitioning coefficients (Kd) for cationic metals in soils using mid-infrared diffuse reflectance spectroscopy. Environ Toxicol Chem., 34: 224-34.

Jarsjö J, Andersson-Sköld Y, Fröberg M, Pietroń J, Borgström R, Löv A, Kleja DB. 2020. Projecting impacts of climate change on metal mobilization at contaminated sites: Controls by the groundwater level. Sci. Tot. Env. 712, 135560.
Jurgens BC, Parkhurst DL, Belitz K. 2019. Assessing the lead solubility potential of untreated groundwater of the United States. Environ Sci Technol., 53: 3095-3103.
Kajander S, Parri A. 2014. Best Available Techniques (BAT); Management of the Environmental Impact of Shooting Ranges. The Finnish Environment 4 | 2014. Ministry of the environment. ISBN 978-952-11-4352-6 (PDF)
Kelebemang R, Dinake P, Sehube N, Daniel B, Totolo O, Laetsang M. 2017. Speciation and mobility of lead in shooting range soils. Chemical Speciation & Bioavailability. DOI: 10.1080/09542299.2017.1349552
Knechtenhofer L, Xifra I, Scheinost A, Fluhler H, Kretzschmar R. 2003. Fate of heavy metals in a strongly acidic shooting range soil: small scale metal distribution and its relation to preferential water flow. J. Plant Nutrition Soil Sci., 166: 84–92.
Lafond S, Blais J-F, Mercier G, Martel R. 2014. A Counter-Current Acid Leaching Process for the Remediation of Contaminated Soils from a Small-Arms Shooting Range, Soil and Sediment Contamination: An International Journal, 23:2, 194-210, DOI: 10.1080/15320383.2014.808171
Lanno RP, Oorts K, Smolders E, Albanese KM. Chowdhury J. 2019. Effects of soil properties on the toxicity and bioaccumulation of lead in soil invertebrates. Environmental Toxicology and Chemistry. https://doi.org/10.1002/etc.4433
Laporte-Saumure M, Martel R, Mercier G. 2012. Pore water quality in the upper part of the vadose zone under an operating Canadian small arms firing range backstop berm. Soil and Sediment Contamination. 21: 739–755.
Larson SL, Martin WA, Griggs CS, Thompson M, Nestler CC. 2011. Comparison of Lead Dissolution from Antique and Modern Ammunition. Environmental Forensics, 12:149–155
Lessoff SC, Indelman P. 2004. Analytical model of solute transport by unsteady unsaturated gravitational infiltration. Journal of Contaminant Hydrology 72: 85 – 107.
Li Y, Zhu Y, Zhaob S, Liu X. 2015. The weathering and transformation process of lead in China's shooting ranges. Environmental Science: Processes and Impacts, DOI:10.1039/c5em00022j
Lin Z, Comet B, Qvarfort U, Herbert R. 1995. The chemical and mineralogical behaviour of Pb in shooting range soils from central Sweden. Environ Pollut., 89: 303–309.
Lodygin ED, Alekseev II, Vasilevich RS, Abakumov, EV. 2020. Complexation of lead and cadmium ions with humic acids from arctic peat soils. Environmental Research. 110058.
McLaughlin MJ, Hamon RE, McLaren RG, Speir TW, Rogers SL. 2000. Review: A bioavailability-based rationale for controlling metal and metalloid contamination of agricultural land in Australia and New Zealand. Australian Journal of Soil Research, 38: 1037 – 1086.
Mariussen E, Ljønes M, Strømseng AE. 2012. Use of sorbents for purification of lead, copper and antimony in runoff water from small arms shooting ranges. Journal of Hazardous Materials 243 (2012) 95– 104
Martin WA, Lee LS, Schwab P. 2013. Antimony migration trends from a small arms firing range compared to lead, copper, and zinc. Science of the Total Environment 463–464: 222–228.
Nativ R, Adar E, Dahan O, Geyh M. 1995. Water Recharge and Solute Transport Through the Vadose Zone of Fractured Chalk Under Desert Conditions. Water Resources Research, 31: 253 – 261.
Negley T., Newcome A., Sweeney P., Fish L., Hendley P. 2013. Development and Verification of a GIS Layer to Identify Shallow Groundwater Regions for Monitoring in the EU. Poster presented at York Pesticides Conference 2013
Ogawa S, Katoh M, Sato T. 2014. Contribution of hydroxyapatite and ferrihydrite in combined applications for the removal of lead and antimony from aqueous solutions. Water Air Soil Pollut., 225: 2023-2029.
Okkenhaug G, Smebye AB, Pabst T, Amundsen CE, Sævarsson H, Breedveld GD. 2018. Shooting range contamination: mobility and transport of lead (Pb), copper (Cu) and antimony (Sb) in contaminated peatland. J Soils Sediments. 18: 3310–3323.
Reigosa-Alonso A, Dacunha RL, Arenas-Lago D, Vega FA, Rodrıguez-Seijo A. 2021. Soils from abandoned shooting range facilities as contamination source of potentially toxic elements: distribution among soil geochemical fractions. Environ Geochem Health. https://doi.org/10.1007/s10653-021-00900-7.

Rooney CP, McLaren RG, Condron LM. 2007. Control of lead solubility in soil contaminated with lead shot: Effect of soil pH. Environmental Pollution 149: 149-157.

Sanderson P, Naidu R, Bolan N, Bowman M, Mclure S. 2012. Effect of soil type on distribution and bioaccessibility of metal contaminants in shooting range soils. Science of the Total Environment, 438: 452-462.

Sanderson P, Qi F, Seshadri B, Wijayawaredena A, Naidu R. 2018. Contamination, Fate and Management of Metals in Shooting Range Soils—a Review. Current Pollution Reports (2018) 4:175–187 https://doi.org/10.1007/s40726-018-0089-5

Scheckel KG, Diamond G, Maddaloni M, Partridge C, Serda S, Miller BW, Klotzbach J, Burgess M. 2013. Amending soils with phosphate as means to mitigate soil lead hazard: A critical review of the state of the science. J. Toxicol. Environ. Health B 16(6): 337–380.

Sehube N, Kelebemang R, Totolo O, Laetsang M, Kamwi O, Dinake P. 2017. Lead pollution of shooting range soils. S. Afr. J. Chem., 70: 21–28.

Selonen S, Liiri M, Strömmer R, Setälä H. 2012. The fate of lead at abandoned and active shooting ranges in a boreal pine forest. Environmental Toxicology and Chemistry, 31: 2771-2779.

Šimůnek J, van Genuchten MT, Šejna M. 2008. Development and applications of the HYDRUS and STANMOD Software Packages and Related Codes. Vadose Zone J. 7: 587-600

Smolders E, Oorts K, Van Sprang P, Schoeters I, Janssen CR, McGrath S, McLaughlin MJ. 2009. The toxicity of trace metals in soil as affected by soil type and ageing after contamination: using calibrated bioavailability models to set ecological soil standards. Environmental Toxicology and Chemistry, 28: 1633–1642.

SNIFFER (2004) Development of a groundwater vulnerability screening methodology for the Water Framework Directive. Final Report Project WFD28, (British Geological Survey and Macaulay Land Use Research Institute).

Soeder D, Miller C. 2003. Ground-Water Contamination from Lead Shot at Prime Hook National Wildlife Refuge, Sussex County, Delaware US Department of the Interior and US Geological Survey. Water-Resources Investigation, Baltimore, Maryland.

Townsend T, Dubey B, Tolaymat T. 2006. Interpretation of Synthetic precipitation leaching procedure (SPLP) results for assessing risk to groundwater from land-applied granular waste. Environmental Engineering Science. https://doi.org/10.1089/ees.2006.23.239

Trakal L, Kodesova R, Komarek M. 2013. Modelling of Cd, Cu, Pb and Zn transport in metal contaminated soil and their uptake by willow (*Salix × smithiana*) using HYDRUS-2D program. Plant and Soil, 366: 433 – 451.

US EPA (United States Environmental Protection Agency). 2005. Best Management Practices for Lead at Outdoor Shooting Ranges. EPA-902-B-01-001, Revised June 2005, Region 2

Van Bon J, Boersema J. 1988. Sources, Effects and Management of Metallic Lead Pollution. The Contribution of Hunting, Shooting and Angling. Contaminated Soil'88. Springer

Vanderborght J, Gahwiller P, Fluhler H. 2002. Identification of transport processes in soil cores using fluorescent tracers. Soil Sci Soc Am J., 66: 774-787.

Vías JM, Andreo B, Perles MJ, Carrasco F, Vadillo I, Jiménez P. 2006. Proposed method for groundwater vulnerability mapping in carbonate (karstic) aquifers: The COP method. Hydrogeology Journal, 4: 912-925.

Vrba J, Zaporozec A. 1994. Guidebook on Mapping Groundwater Vulnerability, Vol. 16. International Contribution to Hydrogeology, Hannover, Germany 131 pp.

Watmough SA, Koseva I, Landre A. 2013. A comparison of tension and zero-tension lysimeter and PRS™ probes for measuring soil water chemistry in sandy boreal soils in the Athabasca oil sands region, Canada. Water Air Soil Pollut., https://doi.org/10.1007/s11270-013-1663-5.

WHO. 2011. Small-scale water supplies in the pan-European region. WHO Regional Office for Europe, Copenhagen, Denmark 54pp

Yi X, Saha UK, Ma LQ. 2010. Effectiveness of best management practices in reducing Pb-bullet weathering in a shooting range in Florida. Journal of Hazardous Materials 179 (2010) 895–900

Zhang L, Verweij RA, Cornelis, Van Gestel CAM. 2019. Effect of soil properties on Pb bioavailability and toxicity to the soil invertebrate *Enchytraeus crypticus*. Chemosphere, 217: 9-17.

APPENDIX 1 SUMMARY OF CONTACTED STAKEHOLDERS

Country	Organisation name	Organisation website	Survey sent (version)	Response received?
Belgium	International Shooting Center Bauffe	https://www.iscb.be/fr/index.asp	28/06/2021 (FR)	No
Belgium	Klein Brabantse Shooting Club	https://www.shootingclub.be/	28/06/2021 (FR)	No
Belgium	Vlaamse Schietsportkoepel	https://www.sportschieten.be/nl	28/06/2021 (FR)	No
Denmark	Coldbore Range	https://www.coldborerange.dk/home/information/shooting-weekends/	28/06/2021 (EN)	No
Denmark	Skytteklubben DSB/ASF	https://www.dsbasf.dk/	28/06/2021 (EN)	No
Denmark	Københavns Flugtskytte Klub	https://claytarget.dk/	28/06/2021 (EN)	No
Finland	Salon Seudun Ampujat	https://www.sasa.fi/ampumaradat/	28/06/2021 (EN)	No
Finland	Ruutikangas Shooting Sports Center	https://ruutikangas.fi/	28/06/2021 (EN)	No
Finland	Hälvälän Ampumaurheilukeskus	https://www.haukry.fi/	28/06/2021 (EN)	No
Finland	Kokkovuori Shotgun Shooting Center	https://kokkovuoren.fi/	28/06/2021 (EN)	No
France	Centre National De Tir Sportif	https://www.cntir.com/	28/06/2021 (FR)	No
France	Ecole de Tir Sportif de Dijon-Norges	https://www.balltrapdijon.fr/	28/06/2021 (FR)	No
France	Centre régional de tir - Bretteville sur Odon	https://crt-bretteville.jimdofree.com/	28/06/2021 (FR)	No
France	Nimes Shooting Club	http://www.nimes-shooting-club.com/index.asp	28/06/2021 (FR)	No
France	Fitasc Federation	https://www.fitasc.com/fr	28/06/2021 (FR)	Yes
Germany	Waffen Obermeier	https://www.waffenobermeier.de/	28/06/2021 (EN)	
Germany	Schiessstand Warder	http://www.schiessstand-warder.de/	28/06/2021 (DE)	No
Germany	Shooting Sports Center Suhl-Friedberg	https://sszsuhl.de/en-2/	28/06/2021 (DE)	No
Germany	International Hunting and Sport Shooting Club eV Bad Neuenahr	https://www.ijssc.com/	28/06/2021 (DE)	No
Germany	Shooting range Garlstorf gGmbH	https://ssl.forumedia.eu/schiessplan-garlstorf.de/	28/06/2021 (DE)	No
Ireland	Courtlough Shooting Grounds	https://courtlough.ie/shooting-grounds/	28/06/2021 (EN)	No
Ireland	Balheary Shooting Grounds	https://www.balhearyshootinggrounds.com/	28/06/2021 (EN)	No
Ireland	Connemara Shooting School	http://connemarashootingschool.com/	28/06/2021 (EN)	No
Ireland	Harbour House Sports Club	https://harbourhouse.ie/	28/06/2021 (EN)	No
Ireland	The Midlands National Shooting Centre of Ireland	http://nationalshootingcentre.ie/	28/06/2021 (EN)	No
Italy	ASD Tiro A Volo Roma	https://www.tiroavoloroma.com/	28/06/2021 (EN)	No

Italy	TAV Lombard Academy	https://www.accademialombarda.it/	28/06/2021 (EN)	No
Netherlands	International Shooting Range De Wildenberg	https://www.schietbaandewildenberg.nl/	28/06/2021 (EN)	No
Netherlands	Wapenhandel Colenbrander	http://www.wapenhandelcolenbrander.nl/Kleiduiven-Schieten/Schietvereniging-De-Heide/	28/06/2021 (EN)	No
Netherlands	JST Walloon Village	https://www.jst-waalsdorp.nl/index.php?page=Home&sid=1	28/06/2021 (EN)	No
Poland	Pasternik shooting range	http://www.strzelnicapasternik.pl/	28/06/2021 (PL)	No
Poland	Sports shooting Paruszowiec LOK MKS Rybnik	https://www.mks-lok.rybnik.pl/home	28/06/2021 (PL)	No
Spain	Club de Tiro Pinto	http://www.clubdetiropinto.com/	28/06/2021 (EN)	No
Spain	Intensiu Vilaregut Sporting Clay	https://www.campodetirovilaregut.com/	28/06/2021 (EN)	No
Sweden	Skepplanda Sportskyttar	https://www.skepplandasportskyttar.se/	28/06/2021 (EN)	No
Sweden	Lund Pistol Club	https://www.lundspk.se/news.php	28/06/2021 (EN)	No
Sweden	Karlskrona JSK	https://www.karlskronajsk.se/	28/06/2021 (EN)	No

APPENDIX 2 STAKEHOLDER QUESTIONNAIRE (ENGLISH VERSION)

Background

This project is looking to gather information on the mitigation of lead contamination at shooting ranges to prevent migration of lead from ammunition into soil and eventually to groundwater and drinking water. Our primary focus is on the types of risk management measures employed to prevent this and their associated costs.

Questionnaire

1. Please fill in your contact details.

Contact name	
Name of shooting range and/or company	
Preferred contact phone number	
Email address	
Location of shooting range(s)	

2. Within your organisation is there awareness of the potential for lead ammunition in soil to contaminate groundwater beneath?

3. Lead is classified as hazardous for the environment and human health so potential risks should be minimised by the use of risk management measures (RMMs). Please can you describe any such RMMs you have (or have previously had) in place at your shooting range plus any estimates of their effectiveness?

4. Where did you obtain the information to make a decision on the appropriate RMMs?

APPENDIX 3 SITE INVESTIGATIONS

Ref	Site/details	Lead conc (mg kg^{-1})	Leachable lead conc/% [Test + criteria]	Soilwater or Groundwater conc	Soil properties	Underlying geology	Comments and Conclusions
IFUA 2013	Cologne, Germany	Max 52,000 mg kg^{-1}	7.1 mg L^{-1} Pb in eluate [exceeding 25 µg L^{-1} criteria]		Loam + clay	Sand + gravel; water table at 7-14m	Modelling indicated v.low Pb mobility and absence of risk
Austrian UBA, 2018	Glanegg, Austria Operated by the Austrian military since the 19th century still in use by the army as a shooting range	Max 16,000 mg kg^{-1} (North of site) 28,000 mg kg^{-1} (South)	1->6 mg/kg water soluble lead; >0.1%, 'indicating high mobility'	Pb elevated in percolation water <LoD in gw (Sb detectable at one borehole) – no values given		Gravel + sand deposits, underlain by calcareous hard rock; groundwater in gravel at 4-5m. Aquifer supplies the local city of Salzburg. Also, within a radius of 500 m there are four further groundwater withdrawals, including two house wells and two utility water wells	Soil removed from contaminated areas to max depth of 0.9m. No risk to groundwater after removal of contaminated soil
Austrian UBA, 2002	Kuchlmuhle [clay pigeon shooting site since 1955; 1,000 shots per day for 30 days per year, Pb input estimated to be	Max 39,000 mg kg^{-1} (85-110m from firing position)	Pb in Eluate: 2.56 mg L^{-1} (0-5cm) 0.25 mg L^{-1} (5-10cm) 0.07 mg L^{-1} (10-20cm)	Pb in soil water: 12.6 mg L^{-1} (5cm) 1.46 mg L^{-1} (15cm) 0.004 mg L^{-1} (40cm)	Lime-free brown soil with humus layer 15cm Low pH, 4.3 in humus layer and pH 3.8 at depth. Minimal CEC	Granite 15m thick, fissure/frack flow but with no continuous body of groundwater	Primary consideration is soil contamination; risk to deep groundwater is considered to be of secondary concern

Ref	Site/details	Lead conc (mg kg^{-1})	Leachable lead conc/% [Test + criteria]	Soilwater or Groundwater conc	Soil properties	Underlying geology	Comments and Conclusions
	1 tpa Pb so 40 t Pb over site operation]			100 µg L^{-1} Pb in slope water outlet from firing range			
ECHA 2021 Bavaria WWA	Mainbullau, Germany (Miltenberg shooting club)			Max 1.5 mg L^{-1} Pb in soil water from lysimeters at 70cm depth			Detailed investigation enforced by local authorities invoking Bavarian soil protection laws
Islam et al 2016	Cho-do, S.Korea (30m pistol range)	18,609 mg kg^{-1}	0.06% extracted with water; 3.14% TCLP		Sandy loam, pH 6.8. 12.4% OM, 17 Cmol kg^{-1} CEC		Both sites assessed to have 'high risk' to environment (not specifically assessed with regard to groundwater)
	We-rye, S.Korea (400m classification range)	3,918 mg kg^{-1}	0.01% extracted with water; 2.47% TCLP		Sandy loam, pH 6.0. 7.8% OM, 12.9 Cmol kg^{-1} CEC		
Fayiga et al 2011 Mid berm soils	Range G, Florida, USA (100 yd rifle, 9 years operation)	12, 689 mg kg^{-1}	4.34 mg kg^{-1} water soluble – 0.034%; 56.4% TCLP; 3.62 mg L^{-1} SPLP		Sandy (>95%), pH 6.11, 1.01% OM, 24.8 Cmol kg^{-1} CEC		Bioavailability of Pb in shooting range soils found to depend mostly on the geochemical and mineralogical constitution of the berm soils.
	Range O, Florida, USA (100 yd rifle, 23 years)	70,350 mg kg^{-1}	0.69 mg kg^{-1} water soluble – 0.001%; 68.8% TCLP; 3.80 mg L^{-1} SPLP		Sandy (>95%), pH 6.72, 0.21% OM, 11.1 Cmol kg^{-1} CEC		
	Range L, Florida, USA (200 yd rifle, 38 years)	10,068 mg kg^{-1}	12.1 mg kg^{-1} water soluble – 0.12%; 44.9% TCLP; 1.19 mg L^{-1} SPLP		Sandy (>95%), pH 6.68, 0.67% OM, 8.34		

Ref	Site/details	Lead conc (mg kg^{-1})	Leachable lead conc/ % [Test + criteria]	Soilwater or Groundwater conc	Soil properties	Underlying geology	Comments and Conclusions
					Cmol kg^{-1} CEC		
Dortch et al 2013	Fort AP Hill, Vancouver, USA Military firing range - Estimated loading of 7.13 tpa Pb over 98 firing ranges and 3 impact sites				Sandy loam, pH 5.5, 1.2% OM	Sand + gravel, water table at ~6.1m	Groundwater not impacted over last 60 years
Sehube et al 2017 Military shooting ranges in Botswana	MOG Since 1977	~6,000 mg kg^{-1}	~100 mg kg^{-1} SPLP - ~1.6%		Ferric Luvisols, pH 7.07, 0.62% OM, 5.01 Cmol kg^{-1} CEC		SPLP concentrations in all soils exceeded the USEPA 0.015 mg kg^{-1} critical level of hazardous waste indicating possible contamination of surface and groundwater
	TSH Since 1998	~13,000 mg kg^{-1}	787 mg kg^{-1} SPLP - ~0.06%		Ferric Luvisols, pH 6.80, 0.69% OM, 5.34 Cmol kg^{-1} CEC		
	MAK Since 1983	25,193 mg kg^{-1}	~200 mg kg^{-1} SPLP - ~0.8%		Chromic Luvisols, pH 6.64, 1.62% OM, 8.00 Cmol kg^{-1} CEC		
	TAB Since 1995	38,386 mg kg^{-1}	448 mg kg^{-1} SPLP - ~1.2%		Chromic Luvisols, pH 8.25, 0.35% OM, 10.8 Cmol kg^{-1} CEC		

Ref	Site/details	Lead conc (mg kg^{-1})	Leachable lead conc/ % [Test + criteria]	Soilwater or Groundwater conc	Soil properties	Underlying geology	Comments and Conclusions
	SHO R1 Since 1995	~3,000 mg kg^{-1}	~200 mg kg^{-1} SPLP – 6.6%		Chromic Luvisols, pH 7.15, 0.66% OM, 7.07 Cmol kg^{-1} CEC		
	SHO R2 Rifle range established in 1998 but now obsolete	728 mg kg^{-1}	No value discernible for SPLP (very low)		Chromic Luvisols, pH 7.20, 0.58% OM, 3.50 Cmol kg^{-1} CEC		
	PAJ R1 Since 1996	~5,000 mg kg^{-1}	448 mg kg^{-1} SPLP - ~8.9%		Luvic Arenosols, pH 6.95, 0.81% OM, 5.95 Cmol kg^{-1} CEC		
	PAJ R2 Pistol range since 1996	85 mg kg^{-1}	0.1 mg kg^{-1} SPLP – 0.12%		Luvic Arenosols, pH 8.70, 0.77% OM, 12.0 Cmol kg^{-1} CEC		
Kelebemang et al 2017	S/P Pistol	685 mg kg^{-1}	~50 mg kg^{-1} – ~7.3%		Haplic luvisols; pH 6.81, 0.52% OM, 8.20 Cmol kg^{-1} CEC		SPLP concentrations in all soils exceeded the USEPA 0.015 mg kg^{-1} critical level of hazardous waste indicating possible contamination of surface and groundwater
	S/P R2	10,386 mg kg^{-1}	~20 mg kg^{-1} - ~0.19%		Haplic luvisols; pH 8.20, 0.88% OM, 10.24		

Ref	Site/details	Lead conc (mg kg^{-1})	Leachable lead conc/ % [Test + criteria]	Soilwater or Groundwater conc	Soil properties	Underlying geology	Comments and Conclusions
					Cmol kg^{-1} CEC		
	S/P R1	2,741 mg kg^{-1}	~130 mg kg^{-1} – 4.7%		Haplic luvisols; pH 7.30, 0.65% OM, 11.93 Cmol kg^{-1} CEC		
	MAT R2 (active since 1985)	20,882 mg kg^{-1}	0.89 mg kg^{-1} – 0.0043%		Chromic Luvisols, pH 8.36, 2.13% OM, 21.87 Cmol kg^{-1} CEC		
	MAT R1	13,449 mg kg^{-1}	No value discernible for SPLP (very low)		Chromic Luvisols, pH 8.69, 1.32% OM, 20.6 Cmol kg^{-1} CEC		
	LEBO	6,413 mg kg^{-1}	No value discernible for SPLP (very low)		Pellic Luvisols, pH 7.50, 1.10% OM, 27.1 Cmol kg^{-1} CEC		
	TSHU (active since 1984)	14,731 mg kg^{-1}	267 mg kg^{-1}– 1.8%		Pellic Luvisols, pH 7.55, 0.31% OM, 9.26 Cmol kg^{-1} CEC		

Ref	Site/details	Lead conc (mg kg^{-1})	Leachable lead conc/ % [Test + criteria]	Soilwater or Groundwater conc	Soil properties	Underlying geology	Comments and Conclusions
Okkenhaug et al 2018	Elverum, Norway [Military shooting range]	1,400 mg kg^{-1}		~1 µg L^{-1} Pb in porewater 22 ± 5 µg L-1 Pb in groundwater	Peatland, pH 5-2-5.4,50% OM, 78-81 Cmol kg^{-1} CEC	Shallow groundwater in peat	Transport of Pb primarily occurs in the upper peat layer, as a result of a higher hydraulic conductivity close to the surface and a high groundwater table
Reigosa-Alonso et al 2021	NW Spain Clay target site closed for 20 yr	725 mg kg^{-1}	'Available Pb' (extracted with CaCl2) up to 88.98%	180-440 µg L^{-1} in surface water, 50 m from shooting range	Sandy loam, pH 4.81-6.59, 6.35-12.75% OM, <10 Cmol kg^{-1} CEC		With moderate-high contents of organic matter (6–12%), the studied soils have acidic values and low levels of Al, Fe and Mn oxides that favour the migration of Pb through the soil profile and potential transformation to more mobile forms
Selonen et al 2012 Ha¨lva¨la¨ shooting range, Finland	OC: Part of clay target shooting range abandoned 20 years ago	19,034-23,175 mg kg^{-1}	17-42 mg kg^{-1} water soluble – max 0.18%		Moraine soil in the area is characterized by a coarse and stony structure with weak podzolization		In the organic soil layer, weathering of pellets enhanced Pb availability and leaching, indicating an increased risk of groundwater contamination over time at shooting sites located above aquifers
	NC: Active part of clay target shooting range	12,239-28,328 mg kg^{-1}	38-57 mg kg^{-1} water soluble – max 0.47%				
Laporte-Saumure et al 20212	Canadian military range	66,972 mg kg^{-1}	SPLP – 3.4 mg L^{-1}	Average readings of 6.6-15 from lysimeters in berm (max of 30 µg L^{-1}); 11.5 µg L^{-1} from lysimeter in front of berm. Groundwater <LoD	Medium-grained sand composed mainly of quartz, feldspar (albite, orthoclase and	Water table at 6.5m depth	Groundwater established to be unpolluted

Ref	Site/details	Lead conc (mg kg^{-1})	Leachable lead conc/% [Test + criteria]	Soilwater or Groundwater conc	Soil properties	Underlying geology	Comments and Conclusions
					microcline), and mica (illite, a clay-mica). pH 7.6-8.0, 0.2-1.3% OC		
Rodriguez-Seijo et al 2016 50m long SAFR	Monforte de Lemos, Spain	55-6,309 mg kg^{-1}			Humic Cambisols, sandy loam - pH 5.55-6.75, 4.6-8.2% OM, 4.5-9.4 Cmol kg^{-1} CEC	Quartzite, schist and granodiorite alluvium and allochthonous fine alluvial sediments.	The moderate acidity and organic matter content favoured the availability of Pb
Clausen & Korte 2009 US Military sites (active SAFRs)	North-west USA (NW)	5,229-92,400 mg kg^{-1}		Max 670 µg L^{-1} in porewater			Continued soil and water monitoring is needed at these sites.
	South-east USA (SE)	326-20,800 mg kg^{-1}		Max 576 µg L^{-1} in porewater			
	North-east USA (NE) – previous remediation[15] & treated with proprietary reagent containing phosphorus	79-1,207 mg kg^{-1}		Max 50 µg L^{-1} in porewater			
ETH 2002 Rifle range in Luzern, Switzerland	Stand Zihlmatt In use since 1935 - approx 168,000 shots fired each year so a total of	225-247,797 mg kg^{-1}	1.2-29.6 mg kg^{-1} water soluble lead (0.002-1.7%)		pH 3.7-7 (lowest soil pH between bullet trap and wood)	The substratum is made up primarily of tertiary granite sandstone and grey and mixed	Requirement for groundwater to be monitored in order to judge whether contaminated water will reach the groundwater approx. 150m

15 excavation and removal of particles >2 mm considered to have removed a large fraction of the lead.

Ref	Site/details	Lead conc (mg kg^{-1})	Leachable lead conc/ % [Test + criteria]	Soilwater or Groundwater conc	Soil properties	Underlying geology	Comments and Conclusions
Allmend, Switzerland	5.548,500 over the years. Total of 35.3 t lead (since site in use)		Highest proportion of soluble lead found in soil with low pH			marl, which are covered in recent alluvial clay	away (particularly after heavy rainfall), further investigations on the hydrology and the underground flowpaths of the site are necessary. The acidic forest area should be examined further as these conditions enable the lead to leach deeper into the soil
	Stand B In use since 1966 - approx 205,500 shots fired each year so a total of 5.548,500 over the years. Estimated total of 27.7 t lead since site in use.	233,240 mg kg^{-1}	2.5 mg kg^{-1} water soluble lead (0.001%)		Neutral pH		