# Environmental Heath and Risk Assessment: Lead Poisoning, Remediation, and Project Sustainability in Zamfara, Nigeria and Lead-Arsenate Contamination in Washington, USA

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Casey Bartrem

Major Professor: Gregory Moller, Ph.D.
Committee Members: Kodjotse Afatchao, Ph.D.; Daniel Strawn, Ph.D.;
Margrit von Braun, Ph.D.; Ian von Lindern, Ph.D.
Department Administrator: Alistair Smith, Ph.D.

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# AUTHORIZATION TO SUBMIT DISSERTATION

This dissertation of Casey Bartrem, submitted for the degree of Doctorate of Philosophy with a Major in Environmental Science and titled "Environmental Heath and Risk Assessment: Lead Poisoning, Remediation, and Project Sustainability in Zamfara, Nigeria and Lead-Arsenate Contamination in Washington, USA", has been reviewed in final form. Permission, as indicated by the signatures and dates below, is now granted to submit final copies to the College of Graduate Studies for approval.

Major Professor: ______________________ Date: __________
Gregory Moller, Ph.D.

Committee Members: ______________________ Date: __________
Kodjotse Afatchao, Ph.D.

______________________ Date: __________
Daniel Strawn, Ph.D.

______________________ Date: __________
Margrit von Braun, Ph.D.

______________________ Date: __________
Ian von Lindern, Ph.D.

Department Administrator: ______________________ Date: __________
Alistair Smith, Ph.D.

# ABSTRACT

The global burden of environmental disease is increasingly and inequitably carried by low-income nations. In Zamfara State, Nigeria, spikes in gold prices driven by the world economic crisis caused an epidemic of severe lead poisoning, where 400 children died and thousands more suffered irreversible damage. The outbreak is an unprecedented crisis, and the long-term prognosis for future generations is unclear. Risk assessment identified incidental ingestion of contaminated soil and dust as the primary route of exposure. Post-harvest contamination of staple foods also resulted in significant dietary exposures. While lead was the risk driver for environmental and medical intervention, arsenic, cadmium, manganese, and mercury are also of concern as co-exposures to multiple toxicants often have synergistic effects. International groups and Nigerian governments undertook a collaborative, interdisciplinary response, including emergency removal of contaminated soils and medical treatment. The team worked in ten villages over seven years as the project transitioned from an emergency life-saving intervention to a comprehensive, long-term environmental health response. During this period, four remedial effectiveness evaluations were initiated to review project sustainability. These evaluations identified intentional and unintentional project impacts and the influence of social factors on the long-term resilience of the intervention.

The strides in environmental health and safety achieved in high-income countries have come at the expense of the health of low-income nations such as Nigeria. Within the United States, environmental burdens are also inequitably distributed. In central Washington, apple orchards were heavily treated with lead-arsenate insecticide from 1900-1950, resulting in significant soil contamination. Former orchard lands were later converted to residential, public, and commercial properties. The widespread lead and arsenic contamination has largely been ignored by the state government in favor of addressing contamination in wealthier regions of the state, putting marginalized residents – especially children – at risk of exposure to contaminated soils.

In central Washington and Zamfara, understanding exposure routes, dose/response relationships, and social influences on contaminant distribution and risk can aid the scientific and public health communities to better understand, prevent, and respond to the increasing incidence of environmental health crises.

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# DEDICATION

The experience of working in Zamfara changed my life. I am indebted to the people in the communities of Anka and Bukkuyum for sharing their lives with us. We had the honor to work with people from all over the world – from our neighbors in Anka and Gusau to other parts of the continent, Europe, and beyond – and every one of them is an inspiration and shining example of the goodness in humanity.

Each trip to Nigeria was both wonderful and exhausting. The people who supported me deserve more than thanks. I'm grateful to you all for listening, understanding, and supporting me even when you didn't understand.

Being a part of the project in Yakima was an unexpected privilege. The dedication of a few people to bring attention to the issue is inspiring and I count myself lucky to have worked with them.

For every number presented in this dissertation, there is a person with a story. The people behind the data should not be relegated to a statistic.

This dissertation is dedicated to the people of Zamfara.

# TABLE OF CONTENTS

# LIST OF TABLES

# LIST OF FIGURES

# INTRODUCTION

The disciplines of global health and humanitarian response are evolving to include environmental risk and disasters. Whereas public health once focused on communicable diseases, the emerging field of environmental health encompasses more disciplines and a broader scope of challenges. High-income nations such as the United States (US) have made great strides in environmental cleanup and protection in recent decades, resulting in wealthy communities benefiting from more stringent regulations. But industry and pollution have followed the path of least resistance to middle- and low-income nations where environmental and public health laws are less restrictive or not adequately enforced. While wealthy communities enjoy healthier environments, these benefits are at the cost of deplorable working and living conditions in low-income communities where mining, recycling, manufacturing, and industrial operations have relocated. International humanitarian organizations have begun to recognize this trend, widening their scope to include occupational and non-communicable diseases. The World Health Organization (WHO) attributes 23% of the global burden of disease and 26% of deaths in children to environmental factors (Pruss-Ustun and Corvalan, 2006; WHO, 2017). The emergency humanitarian medical organization Médecins Sans Frontières (Doctors Without Borders, MSF) has spent the past seven years responding to an economically driven lead poisoning crisis that previously would have been outside of their typical scope of practice (Calain, 2012; Pringle, 2012).

In Zamfara State, Nigeria, spikes in gold prices driven by the global economic crisis caused a sharp increase in artisanal gold mining. Families that typically relied on subsistence agriculture could make $20-$30 per day by crude methods of mining, breaking, crushing, and washing raw ores (Beaubien, 2012). A new vein of ore discovered late in 2009 yielded higher gold recovery, but it also contained up to 10% lead, which was inadvertently spread through residential areas where ore processing had occurred. The contamination was found in homes where women, confined inside walled compounds by *purdah* under Sharia Law, participated in ore processing while caring for their children and preparing food (Bartrem et al., 2014; Tirima et al., 2016; von Lindern et al., 2011).

When the outbreak was discovered by MSF in March of 2010, more than 400 children had died (Tirima et al., 2016). Subsequent emergency medical and environmental response activities began in June 2010 and continue into 2017 (Tirima et al., 2016). Data generated during the response are the basis for much of this dissertation. A risk assessment at a US site, where a politically neglected agricultural community has been developed on historically contaminated land, is also included.

Examining the exposure routes, dose/response relationships, and cultural influences on contaminant distribution can help the scientific community better understand and respond to environmental health crises. Further, we have a responsibility to acknowledge the social and environmental justice issues that fuel these epidemics to prevent such incidents from happening again.

**Zamfara Nigeria – Lead poisoning outbreak, emergency response, and risk assessment activities**

The most severe outbreak of lead poisoning in modern history (in terms of mortality) occurred in remote villages of northern Nigeria. Discovered in April 2010, response activities have been ongoing since May of that year (Tirima et al., 2016). Four hundred (400) children died of severe lead poisoning, with mortality rates of 25% in children <5 years old (Bartrem et al., 2014; Dooyema et al., 2011; Tirima et al., 2016; von Lindern et al., 2011). The cause of the outbreak was artisanal processing of gold ores later found to contain up to 10% lead. By the end of 2010, 8 villages were known to be affected by the epidemic and numerous international and domestic entities were scrambling to respond (Bartrem et al., 2014; Tirima et al., 2016; von Lindern et al., 2011). In high-income countries, a blood lead level (BLL) that reaches double digits is call for alarm (CDC, 2012). In Nigeria, the highest BLL in a surviving child was found to be over 700 µg/dl, a level previously thought to be lethal (CDC, 1992; MSF, personal communication). The pre-treatment geometric mean BLL was 149 µg/dl; thousands of children have been irrevocably impacted by the effects of lead exposure (Tirima et al., 2016).

Lead is potent toxicant effecting kidneys, liver, reproductive system, cardiovascular system, and nervous system. The neurotoxic effects are most severe, causing permanent damage by interfering with neurotransmitter function. These impacts are more pronounced in the developing brains of children and result in latent, irreversible impacts on behavior, motor skills, and Intelligence Quotient (IQ) scores (ATSDR, 2007a). Children are more susceptible to lead due to higher intake rates, higher absorption, greater neurological vulnerability, and lower detoxification capacities (Landrigan et al., 1999). At high exposures, lead poisoning presents with clinical symptoms: impaired motor skills, speech and hearing deficits, convulsions, behavioral changes, and eventually, paralysis, coma, and death (ATSDR, 2007a). At lower exposures, however, there are no overt neurological symptoms and children may appear healthy. Lead exposure is measured by testing blood, where a fraction of the toxicant is partitioned. Lead is molecularly similar to calcium and, as a result, the majority is stored in bones and teeth (ATSDR, 2007a).

**Lead in the US**

Since the second century, when the toxic effects of lead were first recognized, scientists have gradually added to the list of its pathologies, starting with the overt effects and eventually broadening the scope to include a suite of subtler subclinical neurological and target organ toxicities. Today, lead is among the most well researched toxicants. Despite acute scientific awareness of its hazards, regulations to protect human – and especially children's – health have historically been painfully slow. This pattern is observed in US domestic policies on lead in paint and gasoline as well as health policies on what can be considered a "safe" level of lead in blood.

The "safe" level of lead in blood has steadily reduced in the past century. Into the 1960s, medical doctors considered BLLs under 60 µg/dL to be safe. This reference dose decreased to 40 µg/dL in 1971, and continued to be lowered in the ensuing decades (US Department of Health and Human Services, 1991). Early in the 21$^{st}$ century scientists began to understand that at low exposures, impairments in IQ and behavioral interactions can be observed on a population level (Gilbert and Weiss, 2006; Lanphear et al., 2005). Today, the US Centers for Disease Control and Prevention (CDC) recognizes that there is no safe level of lead in blood and has adopted a reference dose of 5 µg/dl (CDC, 2012).

In the US, children have historically been exposed to lead additives in paint, canned food, lead pipes, gasoline, and various consumer goods (Bellinger, 2006; Jones et al., 2009; Warren, 2000), and from living within a certain distance of lead smelters (Landrigan et al., 1975; Landrigan and Baker, 1981). Screening of urban children for lead poisoning (resulting from lead-based paint exposure) didn't begin until the 1970s when African American activist groups called for government action. This was more than a decade after physicians first noted the correlation between elevated BLLs and substandard housing. 'White lead' continued to be used in paint until a ban in the 1970s, largely due to a strong political lobby on the part of industry (Warren, 2000). Similarly, the use of tetra-ethyl lead (TEL) as a gasoline additive continued long after its dangers were recognized in the scientific community. In both cases, industry employed its own set of medical experts and scientists to debunk independent research (Rosner and Markowitz, 2005; Needleman, 2000; Warren, 2000). The call to remove lead from gasoline began in 1959, but was not realized until 1972. Even then, the adverse health impacts of lead exposure were not the incentive; TEL was found to damage catalytic converters in cars (Needleman, 2000). The frustrating translation of knowledge to action has been attributed to intense industry lobbying and the race and socio-economic status of the people most impacted by lead exposure (Bellinger, 2006; Needleman, 2000). Historically, industry has not shown inclination towards responsible self-regulation, regardless of the prevailing science, and risk has not been equally distributed between members of the US population (Bellinger, 2006). Similarly, environmental health risks are not equally distributed across international borders.

**Inequalities and impacts of US lead policy**

Lead naturally occurs at low concentrations in soils globally. It is a stable heavy metal with numerous modern-day applications, including electronics, batteries, ammunition, and aviation fuel. Lead ore deposits are found globally (ATSDR, 2007b). Lead ores must be processed to remove impurities such as other metals, a process commonly called smelting (Ally et al., 2001). Lead obtained from mining is considered primary production; lead is also obtained via recycling, or secondary production, usually of spent or used lead-acid batteries (SLAB or ULAB). Data from the United States Geological Survey (USGS) indicate that world production of lead has risen substantially over the past century (Kelly et al., 2014). This includes primary and secondary lead during 1900-1954 and secondary lead during 1955-2014 (Kelly et al., 2014). In 1978, the US Environmental Protection Agency (USEPA) set a National Ambient Air Quality Standard (NAAQS) of 1.5 $\mu g/m^3$, which applied to facilities processing lead for both primary and secondary recovery (USEPA, 1978). Around this time, US primary lead production began to decline (Kelly et al., 2014). In 2008, the NAAQS was modified to 0.15 $\mu g/m^3$ (USEPA, 2011). In 2013, the last US lead smelter closed after a long-running legal battle with the community of Herculaneum, Missouri over severe contamination and adverse health effects. Doe Run Company opted to cease operating rather than install technology that would enable the smelter to meet emissions criteria, which it had been violating for several years (Jones, 2013). The fall in US production directly correlates with increasing environmental regulations, yet in spite of this decrease, US (and global) consumption has steadily risen, as has global primary production (Kelly et al., 2014).

Part of the growing disparity between US production versus demand can be explained by an increase in secondary production – or recycling – of lead. US secondary lead production increased steadily over the last century, though industry experts believe that trend is changing (CEC, 2013; von Lindern, 2016). Lead is an excellent candidate for recycling. It is a finite resource and recovery reduces the demand for – and environmental impact of – mining. Unlike the recycling of plastics or paper, there is no loss of quality in recycled lead (International Lead Association, n.d.). Recycling lead products (largely SLABs) prevents them from being landfilled and it is more cost-effective to recycle batteries than to mine and smelt raw ore. SLABs have the highest recovery rate of any recyclable material in the US (International Lead Association, n.d.). Ninety-four percent (94%) of secondary production lead comes from recycled batteries (Guberman, 2012).

Secondary lead recovery rose steadily in the US since the early 1900s. In 2010, SLAB recovery accounted for 91% of lead production in the US (Guberman, 2012). In 2011, the US strengthened lead emission standards at battery recycling facilities and, concurrently, US secondary lead production began to decline (CEC, 2013; Guberman, 2013). Current air quality regulations for primary and secondary smelting facilities are 0.15 $\mu g/m^3$ (USEPA, 2008). The increasingly stringent regulations have resulted in increasing exports to Mexico for secondary smelting (CEC, 2013). US exports of SLABs to Mexico increased between 450% and 525% between 2004 and 2011 (CEC, 2013). In addition to the 25 official smelters in Mexico, experts believe there are many small, unregulated secondary smelters, where emissions, occupational, and public health regulations are less stringent than those in the US and Canada (CEC, 2013).

There are limited data on BLLs outside western countries, though the body of knowledge is slowly growing. In 2009, Reuters reported on lead poisoning in China, which has grown tremendously over the past 15 years as more stringent laws in western nations incentivize the movement of industry (Gao et al., 2001; Wang and Zhang, 2006). China's battery industry is now the world's largest and exports to nations around the world (Hornby, 2009). In Haina, Dominican Republic, 9% of children living near an abandoned battery recycling operation had BLLs below 10 µg/dL. In total, 28% of children tested required immediate medical attention and 5% had BLLs over 70 µg/dL (Kaul et al., 1999; TIFO 2015a). Eighteen (18) children living near an informal battery recycling area in Dakar, Senegal died as a direct result of lead poisoning. Screening of 50 children from the area found BLLs ranging from 50 µg/dL to 600 µg/dL (Carol Potera, 2009; Haefliger et al., 2009; TIFO, 2015b; WHO, 2009).

In La Orya, Peru, severe soil and water contamination has been sporadically documented at a smelter owned by Doe Run Company, the same corporation that recently closed its facility in Missouri (Fairbanks, 2009). High levels of soil contamination pose a serious threat to children’s health (Reuer et al., 2012). Children’s BLLs in the nearby communities are known to exceed 50 µg/dL (Fraser, 2009). A 1999 study found that less than 1% of the 346 children tested had a BLL under 10 µg/dL and 67% of children needed immediate medical intervention (Cerderstav and Barandiaran, 2002). Data on the smelter located in Kabwe, Zambia are more limited despite confirmed soil lead contamination

(Tembo et al., 2006) and more than 30,000 children suspected to be lead poisoned (Branan, 2009; World Bank, 2011; Yabe, 2015). In Torreon, Mexico, soil and dust lead concentrations surrounding a lead smelter exceed US standards by an order of magnitude, resulting in BLLs 3-14 times the current average found in US children (Soto-Jiménez and Flegal, 2011). Other sites have been found to have serious soil contamination, though no blood lead testing has been done (Rosenthal, 2011).

Similarities have been drawn between domestic environmental injustice and the export of environmental risk to low-income nations (Adeola, 2000; Castleman, 1979; Marbury, 1995). The incentives for industry to move to countries with less stringent regulations has been called 'environmental racism on an international scale' (Marbury, 1995). This applies across industries and continents. The US has greatly reduced environmental hazards for most of its citizens at the cost of the health of low-income nations, where regulatory and/or enforcement frameworks are ineffectual (Coughlin, 1996). This trend has been established within the garment industry (Gaba, 2011), electronic waste recycling (Chen et al., 2010; Schmidt, 2006; Tsydenova and Bengtsson, 2011), and toxic waste export (Coughlin, 1996; Gaba, 2011). The injustice is enabled, in part, by the lack of data on environmental health impacts in these countries.

Investigations on the financial costs of lead poisoning in low- and middle-income countries indicate that as much as one trillion dollars may be lost due to the neurological and development impacts of lead exposure. In terms of gross domestic product (GDP) losses, low- and middle-income countries lose a greater percentage of their GDP due to childhood lead exposure than high-income countries (Attina and Trasande, 2013). As US health researchers investigate the subclinical impacts of low lead exposures, lead poisoning outbreaks resulting in encephalopathy and death are occurring in impoverished communities around the world.

**Neglected communities within the US**

Domestic regulations of lead in gasoline, paint, and air resulted in a steady improvement in BLLs of US children under the age of five (Bellinger, 2006; Jones et al., 2009). Overall, the US has achieved great success in lowering BLLs and reducing exposures to environmental contaminants. Unfortunately, clear socio-economic disparities in BLLS persist (Hoover et al., 2012), especially with lead-based paint sources (Levin et al., 2008). The trend of pollution following the path of least resistance occurs between international borders, but also within the US.

In 2015, Flint, Michigan entered the national spotlight when results from water and blood lead screening revealed that the state government had failed to protect its citizens' water source.

Elevated BLLs were significantly correlated with economically disadvantaged neighborhoods (Hanna-Attisha et al., 2015). Government agencies, including the office of the state Governor, are accused of intentionally disregarding warnings over the corrosive nature of the water source and unnecessarily putting thousands of residents at risk (Kennedy, 2016; Wang, 2015). This recent case of environmental injustice highlights the inequitable way that environmental health burdens are placed on socially or economically disadvantaged communities. It also highlights the disparity between what elicits alarm in the developed world – 4.0% of children have elevated BLLs in Flint – versus in low-income nations, where US corporations run mining and smelting operations that would not be tolerated in the US, severely poisoning hundreds of thousands of people. Flint was declared a state of emergency and has been the center of a media frenzy for more than a year. Few people have ever heard of the lead poisoning crisis requiring medical intervention in 67% of children in La Orya, Peru (Cerderstav and Barandiaran, 2002).

**This Dissertation**

The lead poisoning outbreak in Nigeria was fueled by endemic poverty and the global metals market. The 2008 economic crisis resulted in a spike in gold prices that made mining more lucrative than ever before. Occupational and environmental risks that would be unacceptable in high-income countries are the norm in the mining camps in northern Nigeria because mining is the only economic option beyond subsistence agriculture. The immediate result was a children's environmental health crisis on a scale not seen previously. The long-term prognosis for future generations, for miners, and for the communities is unclear.

While lead was the risk driver for environmental and medical intervention, the gold ores also contained elevated levels of arsenic, cadmium, and manganese. In the Nigerian villages, gold ores are pulverized and milled to a fine powder, sluiced, and dried. Mercury is widely used during the ore processing to create a gold-mercury amalgam that is later burned, releasing mercury vapors. Prior to intervention and remediation, all stages of ore processing were undertaken in residential areas, exposing children and adults to hazardous levels of multiple heavy metals via contaminated soils and dusts. ***CHAPTER 1*** *reviews the risks associated with co-exposure to multiple toxicants with a focus on those metals found in residential areas in the Nigerian villages*. Beginning in June 2010, international groups collaborated with various levels of Nigerian governments to develop an emergency environmental and medical response. MSF mobilized in-patient treatment clinics near Dareta and Yargalma villages and began to administer chelation treatment to children under five years of age.

TerraGraphics International Foundation worked with the Zamfara State Government to design and implement emergency cleanup procedures in residences and public areas throughout the two villages. *Details of the cleanup of the 8 Zamfara villages can be found in **CHAPTER 2**.*

In addition to lead exposure via soils and dusts, other pathways were identified as routes of exposure for children and adults. The dietary contribution of lead to the total exposures experienced by children in the lead poisoning outbreak was investigated in three villages in 2011 and 2012. Samples of staple crops and other foodstuffs were collected from un-remediated villages and analyzed for Pb. Parents were interviewed about children's typical daily food consumption to estimate total pre-remediation dietary lead exposure from major staple foods. ***CHAPTER 3** explores dietary contribution of lead to total pre-remediation lead exposures.*

The remediation team worked in ten villages in two states over seven years, enabling more than 2,500 children to receive life-saving chelation treatment from MSF. As both the number of affected villages and the capacity of local and state agencies to implement remediation increased, the project transitioned from an emergency intervention to a comprehensive, long term environmental health response. After each phase of remediation, a remedial effectiveness evaluation (REE) was initiated to review project challenges and lessons learned. Retrospective Social Impact Assessment (SIA) was a valuable tool for analyzing qualitative data from the REEs. SIA also highlighted the importance of solutions that come from locals, rather than solutions that are imposed on the effected villages. Socio-economic conditions are a major cause of the lead poisoning outbreak in Nigeria. More importantly, the social, cultural, and local contexts have proven to be the most effective drivers of addressing the lead poisoning issue in any sustainable manner. ***CHAPTER 4** summarizes data from the four REE efforts and evaluates project sustainability and resilience*.

We have seen that from e-waste to mining to textiles to recycling, pollution follows the path of least resistance from the wealthy who use these products to the most vulnerable populations. Many of the improvements in environmental health and safety seen in the past decades in the US have come at the cost of the health of low-income nations. Within the US a similar pattern of inequality has been highlighted by the environmental justice movement. Hazardous waste facilities and polluting industries are more likely to be placed in or near a low-income neighborhood than an affluent one (Mohai and Saha, 2015). Central and eastern regions of Washington State are major producers of agricultural products, including apples. During the first half of the $20^{th}$ century, a lead arsenate insecticide was applied heavily to orchard trees. Over decades of use, heavy metals accumulated in orchard soils. In some areas, such as the Yakima Valley, former orchard lands have been converted to residential and commercial properties as cities have grown. In 2002 investigations into lead and arsenic levels in residential and public properties revealed widespread contamination and significant risk, especially for children (Ecology, 2003). Yet to date, statewide efforts to address contamination have focused in wealthier regions of the state where soils are contaminated from smelter emissions, neglecting the agricultural regions where legacy contamination put residents – especially children – at risk. ***CHAPTER 5*** *details an investigation into legacy lead arsenate soil contamination at childcare centers of Yakima Valley*.

In both central Washington and Zamfara, investigating the exposure routes, dose/response relationships, and cultural influences on contaminant distribution, risk, and project sustainability can help the scientific community better understand and respond to future environmental health crises.

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# CHAPTER 1 – UNKNOWN RISK: CO-EXPOSURE TO LEAD AND OTHER HEAVY METALS AMONG CHILDREN LIVING IN SMALL-SCALE MINING COMMUNITIES IN ZAMFARA STATE, NIGERIA.

Bartrem, C., Tirima, S., von Lindern, I., von Braun, M., Worrell, M.C., Mohammad Anka, S., Abdullahi, A., Moller, G. "Unknown Risk: Co-exposure to lead and other heavy metals among children living in small-scale mining communities in Zamfara State, Nigeria." *International Journal of Environmental Health Research,* vol. 24, No. 4, 2014, pp. 304-319.

**Abstract**

The lead poisoning crisis in Zamfara State, Northern Nigeria has been called the worst such case in modern history and it presents unique challenges for risk assessment and management of co-exposure to multiple heavy metals. More than 400 children have died in Zamfara as a result of severe lead intoxication since early in 2010. A review of the common toxic endpoints of the major heavy metals advances analysis of co-exposures and their pathologies. Environmental contamination in Bagega village, examined by X-ray fluorescence of soils, includes lead, mercury, cadmium, arsenic, and manganese. Co-exposure risk is explored by scoring common toxic endpoints and hazard indices to calculate a Common Pathology Hazard Risk Ranking of Pb > As > Hg >> Cd > Mn. Zamfara presents an extreme picture of both lead and multiple heavy metal mortality and morbidity, but similar situations have become increasingly prevalent on a global scale.

## 1.1 Introduction

### 1.1.1 Background

Since May, 2010, numerous national and international organizations have been involved in responding to one of the largest incidences of childhood lead poisoning in recent history in Zamfara State, Nigeria. The source of the exposure, which has resulted in an estimated 400 deaths in children, is small-scale artisanal processing of gold from lead contaminated ores in remote communities (MSF). An estimated 16,000 people including more than 3000 children under age 5 years are exposed in this on-going incident (TerraGraphics, 2011). Large increases in global precious metals prices created a gold-rush in the region; villagers moved ore processing operations into communities and residential areas to enable families to participate in the lucrative work and unknowingly introduced highly contaminated ore dust into the villages and homes. Gold ore processing methods are crude; crushing, washing, and mercury amalgamation are done by hand and by using flour milling machines to reduce the ore to a fine powder. All stages of processing took place within the villages and within residential compounds (TerraGraphics, 2011).

Children, especially those 5 years and younger, are known to be most vulnerable to the effects of lead (ATSDR, 2007b). Risk assessment, contaminant characterization, and remedial assessment studies done from May 2010 to July 2011 confirmed that the primary exposure route is the incidental ingestion of lead contaminated soils and household dusts (TerraGraphics, 2011). Soil and dust ingestion rates are likely higher in this region of the world as compared to western settings because homes are constructed with mud and children have no relief from soil exposure. Soil lead levels regularly exceeded 5000 mg/kg, in excess of the United States Environmental Protection Agency (USEPA) limit of 400 mg/kg for residential soils (Dooyema et al., 2011; USEPA, 2001). Conservatively, this results in an estimated 300 µg/day lead uptake, assuming 200 mg of soil ingested per day and 30% absorption based on the Integrated Exposure and Uptake Biokinetic model (IEUBK) for lead. Contamination of food sources during food preparation and processing, inhalation of contaminated dusts, and consumption of contaminated water are secondary exposure pathways that magnify lead intake for the exposed populations (TerraGraphics, 2011).

Since May 2010, two phases of emergency environmental remediation – implemented by the Zamfara Ministry of Environment (ZMOE) – and subsequent medical intervention – administered by Médecins Sans Frontières (MSF) – have addressed soil contamination in 7 villages, home to more than 1500 children (TerraGraphics, 2011). Data available for 5 of the 7 remediated villages indicate that 88% of family compounds had maximum soil lead levels over 400 mg/kg, 40% were between 1000-4999 mg/kg, 15% were between 5,000-24,999, 13% were between 25,000-100,000 mg/kg, and 6% of residences had maximum soil lead levels over 100,000 mg/kg (TerraGraphics, 2011).

Mean blood lead levels (BLLs) for children 5 years and younger in May-July 2010 were greater than 140 µg/dL, and individual cases exceeded 700 µg/dL (Biya et al., 2010; Dooyema, et al., 2011; TerraGraphics, 2011). An estimated 26% of children less than age 5 years died between May 2009 and 2010, with 82% of deaths occurring in the latter 6 months. Two hundred and five (205) blood samples collected in May-June 2010 revealed that 97% of children had levels over the 10 µg/dL threshold (Biya et al., 2010; Dooyema, et al., 2011).

In addition to lead, other heavy metals are present in the ores, including but not limited to arsenic, manganese, and cadmium. Mercury is used during the final stages of processing to amalgamate gold particles and has been detected in both soil and air sampling (TerraGraphics, 2011). Lead is the primary toxicant of concern and both remediation and medical chelation treatment remove secondary heavy metals while targeting lead. However, the overall toxic risk to the populations, especially children, is not limited to lead poisoning.

While the effects of lead exposure to children are well researched and understood, the combined effects of co-exposure to a mixture of heavy metals is largely unknown, especially for intense, chronic exposures. This information is critically relevant to the current situation in Zamfara as not all affected villages have been remediated. More than 40 additional villages have been identified with confirmed cases of lead poisoning (Lo et al., 2012). Bagega village has been reported as a priority for remediation as it is home to more than 1500 children under age five years, and has the greatest extent of contamination of any village identified to date (TerraGraphics, 2011; World Health Organization, 2011). Children tend toward greater exposure to chemicals in contaminated environments due to behavioral and physiological factors and are exceptionally susceptible lead poisoning. In comparison to adults, children (i) have increased incidental hand-to-mouth activity resulting in higher ingestion rates, (ii) have greater absorption rates , and (iii) are more biologically sensitive to toxicities (ATSDR, 2007b; Doyle, Blais, & White, 2010; US Department of Health and Human Services; Public Health Service, 1991). During infancy, pathways necessary to metabolize and detoxify xenobiotics are not well developed, the blood brain barrier is not fully developed until age six, and children are in a stage of development critically sensitive to toxicants (Landrigan, Suk, & Ameir, 1999).

We focus on children's exposure to five metals in the Zamfara situation: lead, mercury, arsenic, cadmium, and manganese. All five of these metals are neurotoxic and several elicit overlapping toxicodynamics and toxic endpoints (see Table 3).

**1.1.2 Five Metals of Concern**

Lead is one among the most researched environmental toxicants. Children exposed to lead in infancy or early childhood perform lower on IQ tests and have a higher likelihood of developing residual social and behavioral problems later in life (Lanphear et al., 2005; Needleman, Schell, Bellinger, Leviton, & Allred, 1990). An estimated decline of one to five IQ points is expected with an increase of 10 µg/dL in blood lead (Hubbs-Tait, Nation, Krebs, & Bellinger, 2005). At high BLLs (over 70 µg/dL in children) lead produces encephalopathy, vomiting, convulsions, coma, and death (ATSDR, 2007b). CDC recently established a reference value of 5 µg/dL and recommends that children's blood lead levels be below this level (Advisory Committee on Childhood Lead Poisoning Prevention, 2012), though no safe level of lead in blood has been established (Lanphear, et al., 2005; Needleman, et al., 1990).

Mercury toxicity in children results in abnormal speech, cerebellar ataxia, motor dysfunction, auditory and visual processing impairment, and cognitive damages. Prenatal methylmercury exposure is inversely associated with children's scores on neuropsychological tests (ATSDR, 1999; Hubbs-Tait, et al., 2005). Of the three commonly used categories of mercuric forms, organic (methyl) mercury and metallic (elemental) mercury are more relevant to the situation in Zamfara than is inorganic mercury. Metallic mercury is unique to the other heavy metals discussed here in terms of the active exposure pathways in Zamfara. Incidental ingestion of soils and dusts is the primary route by which children are exposed to heavy metals. However, 80% of inhaled elemental mercury enters the blood stream from the lungs, as compared to less than 0.01% from the GI tract, though methylmercury is readily absorbed after ingestion (ATSDR, 1999).

Correlations between pre- and postnatal cadmium exposure and IQ deficits have been demonstrated, implicating cadmium as a potential neurotoxicant. Negative correlations between cadmium exposure and infant scores on the Apgar test support the observations. Developmental exposure in laboratory animals indicates that operant performance and conditioned avoidance are negatively impacted as well. Cadmium appears to cross the placental barrier and accumulate in the foetus. The developing foetus is negatively impacted by exposure *in utero*, resulting in neurodevelopmental toxicity (ATSDR, 2008a). Cadmium absorption is relatively low for both inhalation (25%) and ingestion (1-10%) routes (ATSDR, 2008a).

Manganese is a neurotoxic metal that adversely impacts cognition, memory, motor skills, and social behaviour in the developing brains of children. The toxicodynamics of manganese and lead are similar in that they involve calcium channels, damage to cells of the central nervous system (CNS), and impair dopamine transmission (Claus Henn et al., 2011). Manganese is an essential trace element, unlike the other heavy metals discussed here. It occurs in both inorganic and organio-metalic forms, though toxic profiles are similar for both. Neurotoxicity is observed in both adults and children. Though most information on toxicity is derived from occupational exposure in adults, effects are more severe in children (ATSDR, 2008a). Manganese uptake is not well quantified, but in general is low in adults (5%) and higher in infants (ATSDR, 2008b; Hubbs-Tait, et al., 2005). Adults in chronic exposure occupational settings exhibit memory loss, behavioural/social impairments, and even motor dysfunction as a result of toxicity to the CNS (Wright, Amarasiriwardena, Woolf, Jim, & Bellinger, 2006).

Exposure to arsenic results in similar toxicities in adults and children, although as with lead, chronic exposure in children may result in lower IQ scores. High doses result in encephalopathy, presenting as confusion, hallucination, memory dysfunction, and peripheral neuropathy (ATSDR, 2007a). Arsenic occurs in both organic and inorganic forms, though most of the arsenic that naturally occurs in rock and soil is inorganic. Inorganic arsenic is considered to pose a greater risk to human health and it is classified as a known human carcinogen (ATSDR, 2007a).

Thus, we see that co-exposure, common in many hazardous site, remains a challenge for risk assessment and risk mitigation. In this work, we report and interpret data collected during an emergency response project under challenging field conditions to support public health and remedial action triage in the context of co-exposure to lead and other heavy metals with high rates of mortality and morbidity. In particular, we apply this analysis to Bagega village in Zamfara, with an exposed population of 7000-9000 people (Anka Local Government Area, 2006). We use these results to explore the complex challenges of hazard analysis of mixtures of heavy metals, and comorbidity with other disease, as a driver for risk management and remedial actions. As of early 2013, Bagega is yet to be remediated; exposures cited are on-going and no medical treatment is possible until remediation is completed.

### 1.2 Methods

#### 1.2.1 Sample collection

Prior to *in situ* testing with handheld X-ray fluorescence (XRF), areas were mapped by hand. Residential compounds are open-air, walled in areas with smaller single rooms, measuring three by three meters on average. Homes are built with mud, though concrete floors are found in some areas. The dimensions and buildings of a family compound were drawn to approximate scale, with rooms labelled for primary use (bedroom, storage room, etc.) and other areas such as open wells and food preparation areas were identified. Ore processing and storage locations were also noted. Of more than 380 homes, 139 were chosen for characterization. Homes were chosen at random based on geographical distribution.

*In-situ* data were obtained using XRF spectrometers (InnovX model Alpha-2000). XRFs were standardized at start-up and every 4 hours thereafter and were calibrated twice a day using NIST

2702, NIST 2781, and $SiO_2$ blank standards. The XRF reports a different instantaneous limit of detection (LOD) for 35 metals for each analysis based on background signal. The goal of sampling was to determine the horizontal and vertical extent of contamination to define areas where remediation was required. XRF results were recorded on sample maps and field data were entered into Microsoft Excel®. The *in situ* XRF sampling approach included probabilistic and judgmental sampling. Probabilistic sampling involved a systematic grid; judgmental sampling relied on the samplers' experience with contamination patterns present within the affected communities. This includes tree-shaded areas where ore processing work was performed, areas near water sources, and areas where ore processing or storage was reported. For risk assessment purposes, bias sampling also targeted areas where children spend a majority of their time. Because sampling was performed to determine remedial action, samplers stopped testing areas when sufficient contamination was identified to support removal. Conversely, clean areas were extensively sampled to justify no action.

*Ex-situ* data were obtained from samples collected during risk assessment, characterization, and remediation activities from May 2010-March 2011. *Ex-situ* ore samples were obtained at processing areas with permission from the owner of the ore, and were placed into Whirl-Packs®, labelled with reported origin information, processing area, and stage of processing. Samples were later analysed with the XRF through the plastic bag after homogenization. Ore sample results reported here are of finely ground samples that ranged from the texture of sand to that of a fine powder.

**1.2.2 Data Analysis**

Data were imported, managed and analysed using Microsoft Excel® and Sigmaplot® version 12.3 for statistical parameters. ArcGIS® was used to spatially plot compound and public area maximum XRF results. *In-situ* XRF data were categorized by village, compound or public area unique identification number, and area usage (common area, bed room, well area, etc.). Homes (compounds) are classified as "interior; public areas are classified as "exterior" residential locations. Seven villages have been assessed and remediated by the ZMOE. Bagega village has not been remediated but preliminary assessment work was carried out from January to March, 2011 when 139 of an estimated 380 home compounds were characterized. Only data from Bagega village are presented here as it is representative of on-going exposures.

*Ex-situ* XRF data of 42 ore samples were collected from 5 different area villages. Ore sample XRF data were analysed by individual ore sample minimum, maximum, average, and geometric mean heavy metal concentration in ppm (mg/kg). The nature of ore processing and lack of certainty regarding "purity" of ores makes it impossible to ascertain the exact origin of ore or ensure that no mixing of ores had occurred during processing. As a result, ore concentrations presented are an average of the 42 samples analysed. This ore composition is representative of the metal mixture to which children are exposed. Each sample was tested with the XRF a minimum of two times. Work on site samples comparing field XRF lead and laboratory analysis shows XRF analysis underestimates actual lead values by 30-50% (Plumlee, personal communication, 2012). This underestimation may be the result of compositing/sieving of samples, analysis method bias, or the lack of an appropriate calibration standard matching the extremely high lead concentrations found in the villages.

**1.3 Results**

The ores used for gold extraction in Zamfara vary considerably in their mineralization. An example of heavy metal composition of various ores using XRF data collected by the remediation team is presented in Table 1. A Hazard Index (HI) was calculated using available US residential standards for each of the five metals.

Remediation and medical treatment have focused exclusively on lead as blood testing and risk assessment indicated lead to be the primary toxicant of concern (MSF, personal communication, 2010). An overview of soil heavy metal levels in Bagega village is presented in Figure 1 along with available regulatory residential soils criteria for heavy metals. Notably, 88% of residential exteriors and 63% of residential interiors had average Pb in soil results exceeding the USEPA standard (maximum Pb results exceeded the standard in 93% and 100% of interiors and exteriors, respectively). Maximum results exceeded referenced standards for Mn and Cd in 7% and 23% of homes and 12% and 24% of exterior areas, respectively. Field staff used an "outside-in" sampling strategy and ceased testing areas once sufficient contamination levels were identified. As a result, overall contaminant measurements are biased low toward "cleaner" areas, as these regions were tested more thoroughly to confirm that no contamination was present.

Ninety three percent (93%) of residences characterized had soil lead levels over 400 mg/kg and 47% of homes tested had soil lead levels in excess of 5000 mg/kg, as indicated in Table 2. Figure 2 shows an aerial photo GIS overlay of Bagega village with compound and exterior contamination categories for characterized areas. Bagega is a regional centre for health, commerce, education and the gold trade; a 50,000 $m^2$ abandoned industrial-scale ore processing site sits directly adjacent to the village and a large water reservoir. Characterization of this area revealed large areas of ore wastes highly contaminated with lead and other heavy metals, some in excess of 10%, or 100,000 mg/kg lead (TerraGraphics, 2011).

An abbreviated description of overlapping toxic endpoints of the five metals discussed is presented in Table 3. This is not intended to be an exhaustive list; rather it covers a review of the potential simultaneous effects of co-exposure to the toxicants discussed here. An overlap of toxicities resulting from metal exposure is evident, especially in terms of neurotoxic action. Clinical observations of permanent alterations to the CNS in terms of IQ and memory are observed and dopaminergic effects are elicited by all but one metal. Overlapping nephrological and haematological toxicities are also noted.

Modelling is often used as a substitute for quantified data on the interaction mechanisms of multiple chemical exposures. For chemical mixtures in which the components are known to cause the same deleterious effect and/or cause harm to the same target organ, a Hazard Index (HI) approach is often implemented, where dose addition is calculated based on reference doses of the individual chemical components. This approach may oversimplify the reality of the mixture's effects (Hertzberg & Teuschler, 2002), but the HI approach is commonly applied to metal mixtures when determining the priority of mixture components within the substance (Nordberg, 2007).

The HI presented in Table 1 rank as > Pb >> Hg >>Cd > Mn. In the present analysis, the information in Table 3 summarizing common pathologies associated with the observed heavy metals can be used to advance consideration of co-exposure risk in site response actions when used to weigh the individual HI associated with each toxicant. Only coinciding non-cancer pathologies are considered in this research. In what we call a Common Pathology Hazard (CPH), we calculate $CPH_i = CPS_i \times HI_i$ , where for toxicant *i*, *CPS* is the common pathology score of and *HI* is the Hazard Index. Summing the common toxic endpoint pathologies scored in Table 3 for each metal produces a relative ranking of: Pb (19) > Hg (18) > Mn (10) > Cd (9) > As (6). When the pathology score is weighted by multiplying by the HI (Table 1), we find a Common Pathology Hazard Risk Ranking (CPHRR) as Pb (4300) > As (2000) > Hg (520) >> Cd (20) > Mn (1).

**1.4 Discussion**

**1.4.1 Metal Mixtures**

Overall, there is limited qualitative and quantitative risk characterization available on metal mixtures that occur at hazardous sites. Manganese seems to effectively lengthen the amount of time that lead resides in blood plasma, prolonging the haematological toxic effects of lead (Roney, Abadin, Fowler, & Pohl, 2011). Co-exposure to arsenic, cadmium, chromium and lead were highly dose dependent, with antagonistic effects presenting at the higher range of dosage and synergistic at the lower exposures, for a range of effects in human keratinocytes (Bae, Gennings, Carter, Yang, & Campain, 2001).

Lead, mercury, manganese and cadmium have all been shown to effectively cross the placental barrier (Hubbs-Tait, et al., 2005). Exposure to lead-arsenic mixture resulted in decreases in norepinephrine and increases in serotonin expression in the midbrain and frontal cortex of mice when exposure to either metal alone did not elicit these effects (Mejia, Diaz-Barriga, Calderon, Rios, & Jimenez-Capdeville, 1997). Synergistic effects of lead and arsenic on mercury exposure have been demonstrated in female mice (Bells, 2002). Cadmium exposure has been demonstrated to increase arsenic toxicity in rats (Diaz-Barriga et al., 1990). Combinations of lead and manganese elicited synergistic effects on nerve action potentials in rats orally exposed over the course of a 12 week trial (Papp, Pecze, Szabo, & Vezer, 2006).

Children with elevated manganese and arsenic levels living the area of the Tar Creek Superfund Site in Oklahoma performed significantly lower on IQ, learning, and memory tests. This interaction was greater-than-additive for children with manganese and arsenic hair levels above the median value (Wright, et al., 2006). In a study of children aged 12 to 36 months in Mexico City, a synergistic interaction between manganese and lead levels was observed in motor skill and intellectual development at moderate to high blood lead and manganese levels. This association was more pronounced at 12 months than 24 months of age and on the psychomotor tests as compared to IQ tests (Claus Henn, et al., 2011). In a cross-sectional study of adults exposed to cobalt, lead and cadmium in occupational settings, co-exposures resulted in synergistic mutagenic effects, potentially due to both the damage to DNA strands and the inhibition of certain DNA repair mechanisms (Hengstler, 2003).

Conversely, less-than-additive (antagonistic) effects have been observed in *in vivo* and *in vitro* studies of heavy metal co-exposures. Fortual et al. (2005) found in mice that simultaneous inhalation of Cd and Pb demonstrated lower Pb lung tissue accumulation than when exposed to either metal alone. Yet the same animals showed a more severe decrease in the production of non-ciliated bronchiolar cells when co-exposed as compared to either metal alone (Fortoul et al., 2005).

Haematological effects of arsenic and cadmium have been shown to decrease erythrocyte count in a less-than-additive manner and in some instances, the presence of two heavy metals has resulted in an antagonistic effect, whereby one metal ameliorates the toxic effects of the second (Roney et al., 2011). An *in vitro* study of lead, cadmium, arsenic and chromium as potential gene-expression interrupters found that individually, each metal produced significant effects on the various assays but no synergistic effects were detected with any combinations of these metals (Mumtaz, Tully, El-Masri, & Rosa, 2002).

The CPHRR approach is a simple and straightforward second-order approximation, using a Common Pathology Score, $CPS_i$, and the Hazard Index that can be useful in providing guidance to drive emergency remediation efforts in co-exposure scenarios by coupling site contamination data, common toxic endpoint pathologies and individual toxicant HI analysis. With the added knowledge of the diverse environmental chemistry and exposure pathways of these heavy metals, field teams can be better equipped to mitigate risk. In Zamfara, the combination of the CPHRR and environmental concentrations results in soil lead as the dominant risk driver for the clean-up. One notable exception, however, occurs in homes where ores low in lead were processed; in those cases, mercury used in amalgamation is the risk driver.

The situation in Zamfara is somewhat unique; reported soil lead and BLLs are unprecedented and the severe morbidity and mortality necessitated coordinated emergency medical, environmental, and public health awareness responses. Yet, with respect to the multiple toxicants involved, it is not a unique exposure scenario. It is common for heavy metals to co-exist at hazardous waste sites and for those metals to involve simultaneous exposure pathways. Agency for Toxic Substances and Disease Registry (ATSDR) identified chemical mixtures as one of the top priority research areas in environmental public health (De Rosa, El-Masri, Pohl, Cibulas, & Mumtaz, 2004). In the US, a majority of uncontrolled waste sites have more than one contaminant (Landrigan, et al., 1999). On a global scale, hazardous waste sites with chemical mixtures are a growing issue as local economies develop and rich countries export their wastes to be recovered and recycled in uncontrolled environments.

### 1.4.2 Qualitative Influences on Toxicity

Qualitative influences must also be evaluated. The overall toxicity experienced by these populations is not limited to metal mixtures. Some of these factors can be quantified by the IEUBK for lead in addition to the limited information on co-exposure toxicity reviewed above. Yet multiple qualitative uncertainty factors influence the dose (intake), the absorption (uptake), and the resulting physiological toxicities in individual children. Most of these factors also influence exposure to the metal mixture of various ores.

Dose may be influenced by behavioural factors, such as the individual child's hand-to-mouth activity, hygiene, age, mobility, and play areas. Contamination levels and patterns in the household also play a significant role – maximum soil lead levels presented here are for the compound as a whole, but the areas where younger, less mobile children spend time are of greater concern from a risk assessment perspective. Non-ore processing related sources of lead exposure in Zamfara are also important, such as the use of galena as a cosmetic and the recycling of lead-acid batteries for the production of lead bullets.

Absorption is a function of several variables including metal speciation. The presence of certain genes may increase uptake of lead (Schmidt, 2008). Nutritional status of the individual, including iron, calcium, and zinc status as well as fat stores, ascorbic acid, and protein levels, play a modifying role in lead uptake (Goyer, 1997; Mahaffey, 1983). Specifically, while chronic lead poisoning can lead to anaemia (Counter, Buchanan, Ortega, & Rifai, 2000; Hutchinson & Stark, 1961; Schwartz, Landrigan, Baker, Orenstein, & von Lindern, 1990), predisposition to low haemoglobin levels may facilitate higher lead absorption (Bradman, Eskenazi, Sutton, Athanasoulis, & Goldman, 2001; Goyer, 1997; Morrison, 1987). The particle size of the lead contaminated soils and dusts influences absorption through both ingestion and inhalation (Barltrop & Meek, 1979; Rendall, Baily, & Soskolne, 1975).

Finally, toxicity—especially in the case of immunotoxic lead—may be affected by comorbidities with other diseases such as malaria and malnutrition, both of which are prevalent across the region and elicit haematological crisis (Miller et al., 1998; Nriagu et al., 2008; Torre et al., 2002). At the conclusion of the 2010 rainy season, 90% of patients at the MSF outreach clinic had malaria and were provided nutritional supplements. More than 200 children were simultaneously treated for lead poisoning and cholera (TerraGraphics, 2011). Both pesticides and lead are known to have negative neurodevelopmental effects in children (Gulson, 2008). As a result, the prevalence of pesticide use in Zamfara, a region known for agricultural production, could also play a role. These factors in addition to co-exposure to heavy metals have the potential to exacerbate physiological response to lead poisoning and resulting toxicities.

Few components of children's exposure in Zamfara can be viewed as reducing overall toxicity, though evidence of antagonistic effects of iron, calcium, copper and zinc on lead toxicity at low doses

(ATSDR, 2004). Preliminary mineralogical sample analyses indicate that copper and zinc are present in the ores. In addition, soil sample XRF results reveal high levels of iron in Zamfara soils (TerraGraphics, unpublished data).

**1.5 Conclusions**

Zamfara presents an extreme picture of both lead and co-exposure heavy metal mortality and morbidity. Lead was identified as the priority toxicant posing the most serious risk to human health and remediation of lead contaminated soils results in exposure reduction to other metals. Common toxic endpoint pathologies from exposure to the observed heavy metals are expected with Common Pathology Hazard Risk Ranking (CPHRR) of Pb > As > Hg >> Cd > Mn. Coupled with comorbidity with common diseases such as malaria, low nutritional status, and the chronic nature of the high level exposures, villages such as Bagega in Zamfara are a case of significant environmental health crisis, especially for children.

Internationally, concurrent exposure to multiple metals near mining sites is not uncommon and environmental health issues in developing nations are often amplified by socio-economic and political instabilities. The scenario of increasing chemical disease combined with morbidity due to communicable disease further disables developing nations (Landrigan et al., 1999). As the incidence of hazardous waste sites, pesticide and pharmaceutical production, manufacturing enterprises and mining become more prevalent in these countries, environmental health crises will increasingly contribute to public health burden. Small-scale mining operations grew by 20% in a five year period in 35 countries across Africa, Asia and Latin America (Yanez et al., 2002). Increasingly, developed nations are relying on poor countries to “import” undesirable environmental risks, and such scenarios can increase the incidence of exposure to multiple toxicants.

Studies of toxicological interactions of chemical mixtures predominantly focus on pairs of xenobiotics (Hertzberg & Teuschler, 2002). Yet the relevance of multiple toxicant exposures is critical in the arena of global public health; in the US alone, approximately 11 million people, including 3 to 4 million children, live within 1 mile of a Superfund site. Heavy metals are one of the top toxicants of concern at uncontrolled sites and the majority of hazardous sites involve more than one contaminant (Landrigan et al., 1999).

Globally, the World Health Organization estimates that children under 5 years of age are at a five-fold increased risk of losing “healthy life years” to environmental risk factors. Children in developing countries lose 8 times more “healthy life years” than those in developed countries, and these statistics do not include chronic effects of exposures (Pruss-Ustun & Corvalan, 2006). The developing approach of quantifying cumulative environmental vulnerability may be a useful tool in advancing environmental health and mitigating risk (Huang & London, 2012). It is manifest that Bagega village has both environmental hazards and social vulnerability, and the co-exposure to lead and other heavy metals is a clear and present danger. Developing our scientific ability to better understand severe environmental public health crises similar to the heavy metal poisoning in Zamfara is becoming more relevant on a global scale, especially in understanding impacts on children's health and development.

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Table 1.1. *Ex situ* XRF results of Zamfara ore samples. Hazard Index values are calculated using United States regulatory standards for residential areas (see Figure 1 for standards) and average concentration for each metal (EPA 1992; Louisiana Department of Environmental Quality 2003; Colorado Department of Public Health and Environment; Massachusetts Department of Environmental Protection 1996; EPA 2001). Statistics calculated with ½ instantaneous XRF LOD for data ≤ LOD.

| mg/kg (ppm) | **Mn** | **As** | **Cd** | **Hg** | **Pb** |
|---|---|---|---|---|---|
| Count | 42 | 42 | 42 | 42 | 42 |
| Max | 7420 | 49200 | 1150 | 2002 | 703000 |
| Min | 25 | 4 | 19 | 5 | 12 |
| Avg | 515 | 3910 | 160 | 286 | 89500 |
| Geomean | 255 | 346 | 72 | 147 | 15000 |
| Hazard Index | 0.1 | 326 | 2.2 | 28.6 | 224 |
| No. above reg std | 2 | 40 | 13 | 39 | 38 |
| % above reg std | 5% | 95% | 31% | 93% | 90% |

Table 1.2. Maximum *in situ* XRF Pb level categories for Bagega village. Categories are based on a "triage" response level used to determine remediation priority.

| mg/kg (ppm) | **<400** | **400-999** | **1000-4999** | **>5000** |
|---|---|---|---|---|
| Residential<br>Interiors | n=10<br>7% | n=12<br>9% | n=52<br>37% | n=66<br>47% |
| Residential<br>Exteriors | n=0<br>0% | n=3<br>9% | n=3<br>9% | n=27<br>82% |

Table 1.3. Select pathologies associated with heavy metals found in Zamfara ore samples. Toxic endpoints with three or more contributing metals in the contaminated villages are highlighted.

| | **Neurological, clinical** | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | convulsions/ encephalopathy | muscle weakness | decreased IQ | alterations in memory/ attention spans | inability to follow instructions | Neurobehav. effects (depression, anxiety, social disorders, etc.) | abnormal speech | motor dysfunction |
| Manganese[a] | | + | + | + | | + | | + |
| Arsenic[b] | + | | + | + | | + | | |
| Cadmium[c] | | | + | + | | | | + |
| Mercury[d] | + | + | + | + | + | + | + | + |
| Lead[e] | + | + | + | + | + | + | + | + |

| | **Neurological, physiological** | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | altered neuronal transmission via NMDA inhibition | cellular dysfunction | enzymatic inhibition | glutathione induction | neuronal degradation | reduced dopamine concentrations/ transmission | altered ATP production | |
| Manganese[f] | | + | + | | + | + | + | |
| Arsenic | | | | | | | | |
| Cadmium[g] | + | | | | | + | | |
| Mercury[h] | | + | + | + | + | + | + | |
| Lead[i] | + | + | + | + | | + | | |

| | **Nephrological, physiological** | | | **Hematological, clinical** | **Hematological, physiological** | | | |
|---|---|---|---|---|---|---|---|---|
| | reduced glomerular filtration | proteinuria | necrosis of tubules | Anemia | interference with heme synthesis | increased protoporphyrin levels | inhibition of ALAD | shortened erythrocyte life span |
| Manganese | | | | | | | | |
| Arsenic[j] | | | | | + | + | | |
| Cadmium[k] | + | + | | + | + | | | |
| Mercury[l] | + | + | + | | | | + | |
| Lead[m] | + | | | + | + | + | + | + |

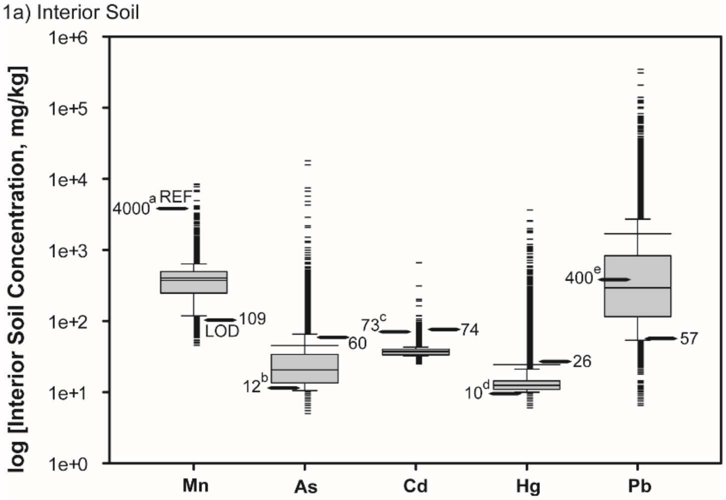
References associated with Table 1.3:

[a](ATSDR, 2008b; Claus Henn et al., 2011)
[b](ATSDR, 2007a)
[c](Hubbs-Tait, Nation, Krebs, & Bellinger, 2005)
[d](ATSDR, 1999; Hubbs-Tait, et al., 2005)
[e](ATSDR, 2007b; Hubbs-Tait, et al., 2005; Needleman, Schell, Bellinger, Leviton, & Allred, 1990)
[f](ATSDR, 2008b; Claus Henn, et al., 2011)
[g](Hubbs-Tait, et al., 2005)
[h](ATSDR, 1999; Hubbs-Tait, et al., 2005)
[i](Hubbs-Tait, et al., 2005)
[j](Roney, Abadin, Fowler, & Pohl, 2011)
[k](ATSDR, 2008a; Roney, et al., 2011)
[l](ATSDR, 1999; Roney, et al., 2011)
[m](ATSDR, 2007b; Roney, et al., 2011)

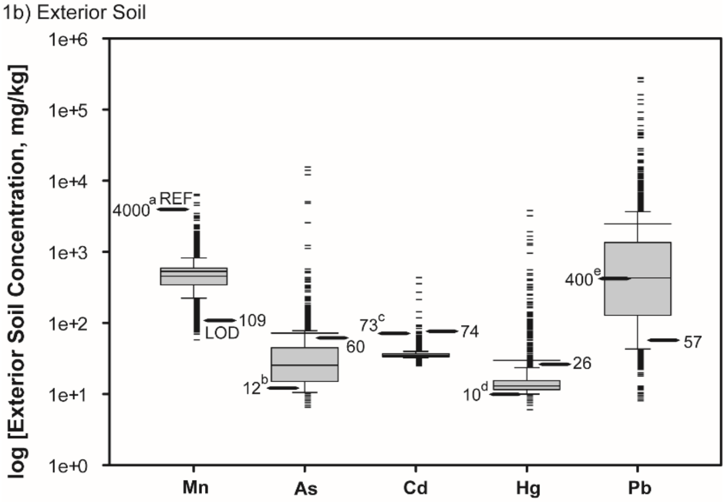
Figure 1.1(a). Log Y-axis plot of the Rank Sum Test box plot graphs of the percentiles and the median of Bagega interior soil data (mg/kg). N=7360 XRF results in 139 homes.

Figure 1.1(b). Log Y-axis plot of the Rank Sum Test box plot graphs of the percentiles and the median of Bagega exterior soil data (mg/kg). N=1705 XRF results. The ends of the boxes define the 25th and 75th percentiles, with a line at the median and error bars defining the 10th and 90th percentiles. Mean is shown. LOD = average LOD and REF = regulatory reference standard (Colorado Department of Public Health and Environment, 1993; Louisiana Department of Environmental Quality, 2003; Massachusetts Department of Environmental Protection, 1996; USEPA, 2001, 2007). Summary statistics calculated with ½ instantaneous XRF LOD for data ≤ LOD.

1a) Interior Soil




1b) Exterior Soil



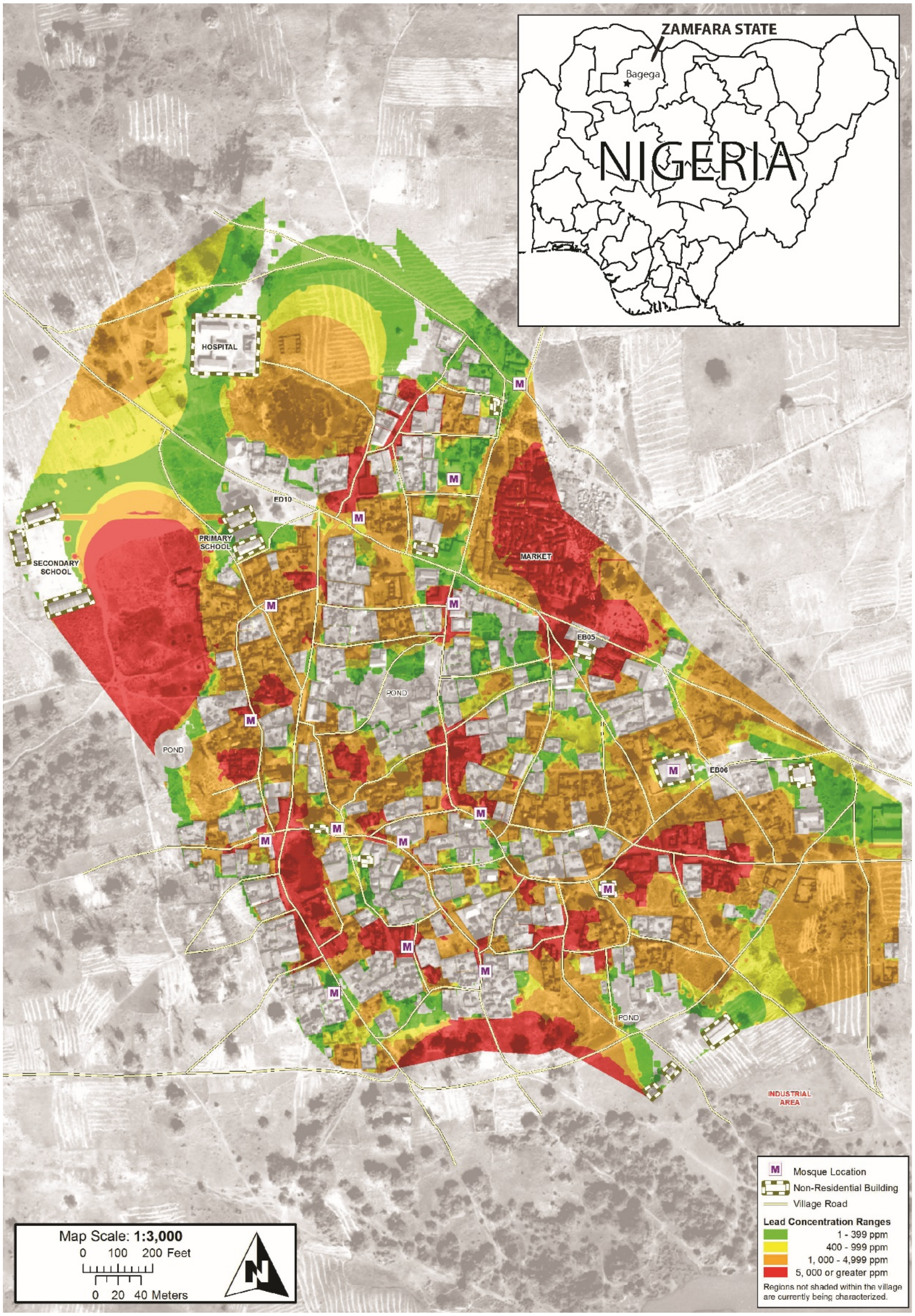

References associated with Figures 1(a) and 1(b):

[a](USEPA, 2007)

[b](Louisiana Department of Environmental Quality, 2003)

[c](Colorado Department of Public Health and Environment, 1993)
[d](Massachusetts Department of Environmental Protection, 1996)
[e](USEPA, 2001)

Figure 1.2. Bagega village aerial map. Residential and public area *in situ* XRF results are geospatially mapped by color-coded zones of maximum soil Pb concentration ranges.



# CHAPTER 2 – ENVIRONMENTAL REMEDIATION TO ADDRESS CHILDHOD LEAD POISONING EPIDEMIC DUE TO ARTISANAL GOLD MINING IN ZAMFARA, NIGERIA

Tirima, S., Bartrem, C., von Lindern, I., von Braun, M., Lind, D., Mohammed Anka, S., Abdullahi, A. "Remediation of childhood lead poisoning epidemic due to artisanal gold mining in Zamfara, Nigeria." *Environmental Health Perspectives,* vol. 124, 2016, pp. 1471-1478.

**Abstract**

**Background:** From 2010-2013, integrated health and environmental responses addressed unprecedented epidemic lead poisoning in Zamfara State, northern Nigeria. Artisanal gold mining caused widespread contamination resulting in the deaths of > 400 children. Socioeconomic, logistic, and security challenges required remediation and medical protocols within the context of local resources, labor practices, and cultural traditions. **Objectives:** Our aim was to implement emergency environmental remediation to abate exposures to 17,000 lead poisoned villagers, to facilitate chelation treatment of children ≤ 5 years old, and to establish local technical capacity and lead health advocacy programs to prevent future disasters. **Methods:** U.S. hazardous waste removal protocols were modified to accommodate local agricultural practices. Remediation was conducted over 4 years in three phases, progressing from an emergency response by international personnel to comprehensive cleanup funded and accomplished by the Nigerian government. **Results:** More than 27,000 $m^3$ of contaminated soils and mining waste were removed from 820 residences and ore processing areas in eight villages, largely by hand labor, and disposed in constructed landfills. Excavated areas were capped with clean soils (≤ 25 mg/kg lead), decreasing soil lead concentrations by 89%, and 2,349 children received chelation treatment. Pre-chelation geometric mean blood lead levels for children ≤ 5 years old decreased from 149 µg/dL to 15 µg/dL over the 4-year remedial program. **Conclusions:** The unprecedented outbreak and response demonstrate that, given sufficient political will and modest investment, the world's most challenging environmental health crises can be addressed by adapting proven response protocols to the capabilities of host countries.

## 2.1 Background

In March 2010, the humanitarian organization Médecins Sans Frontières (MSF) discovered an unprecedented epidemic of lead poisoning in remote villages of Zamfara State, Nigeria (MSF 2010b, 2010c). The first children brought to MSF clinics with convulsions and high fevers were treated for severe malaria and meningitis. As patients failed to recover, blood samples sent to a German laboratory (Labor Lademannbogen MVZ GmbH, Hamburg, Germany) confirmed lead poisoning (Greig et al. 2014). In May 2010, the U.S. Centers for Disease Control and Prevention (CDC) and the World

Health Organization (WHO) dispatched medical and environmental investigators to collaborate with the Zamfara State and the Nigerian Federal Ministries of Health (ZMOH, FMOH) in assessing the epidemic (CDC 2010; WHO 2011). At CDC's request, a U.S. firm, TerraGraphics Environmental Engineering (TG), accompanied the mission to investigate the potential for remediation (Brown MJ, Chief CDC Lead Poisoning and Prevention Program, personal communication, 11 May 2010). Extensive health and environmental assessments in two villages, Dareta and Yargalma, documented 163 deaths, including up to one-third of children < 5 years of age (Dooyema et al. 2012). Subsequent surveys of six additional villages showed that > 17,000 people were severely exposed and an estimated 400–500 children had died of acute lead poisoning (Dooyema et al. 2012; Greig et al. 2014; MSF 2010c; Thurtle et al. 2014; von Lindern et al. 2011; WHO 2011).

The source of lead contamination was prolific artisanal gold mining in response to high gold prices in 2009–2010. For several months, ore processing was conducted at sites within Dareta and Yargalma. Gold ore, sourced from mines throughout Zamfara State, is crushed by hand to gravel consistency using scrap hammers or mortars and pestles, ground to a fine powder in modified flour mills, sluiced by water to separate heavy particles, amalgamated with mercury, and burned over open flame to obtain a low-grade "sponge" gold that is sold to local traders for eventual refining for Dubai or Chinese markets. Grinding produces large quantities of dust, which settle on soils and surfaces. Sluicing results in water-source contamination and large piles of ore tailings. Because local religious and cultural practices include sequestration of married women, ore crushing, washing, and gold recovery were undertaken within the homes ("residential compounds") to use the women's labor. During the rapid increase in mining activities, a vein of ore exceeding 10% lead (Pb) was processed, resulting in severe residential exposures (Plumlee et al. 2013). By April 2010, local emirates suspected a link to children's mortality and ordered artisanal ore-processing operations moved approximately 0.5 km from the villages. Extremely hazardous processing wastes and contaminated soils (> 3% Pb) remained in the compounds and public areas (von Lindern et al. 2011).

Blood lead levels, mortality and morbidity, and environmental lead concentrations observed in this epidemic are unprecedented (Moszynski 2010; Thurtle et al. 2014). In May 2010, the mortality rate in the initial villages surveyed was 25% of children ≤ 5 years; 59% of the surviving children ≤ 5 years were tested and 85% exceeded 65 µg/dL blood lead levels (BLLs) (Dooyema et al. 2012; Greig et al. 2014; Thurtle et al. 2014). The ensuing cleanup was one of the largest and most comprehensive undertaken by an African government (von Lindern et al. 2011; World Bank 2011). Several international organizations, Nigerian health authorities, and local civil and traditional governments collaborated to provide emergency medical, environmental, technical, and public health response. Due to the continuing mortality, the Zamfara State Ministry of Environment (ZMOE) and TG focused on emergency response, as MSF, ZMOH, and FMOH developed village clinics. Chelation therapy was limited to children ≤ 5 years of age and, from July through October 2010, was administered at inpatient clinics in 3–4 week cycles (Greig et al. 2014; Thurtle et al. 2014). All entities agreed that returning treated children to contaminated homes would compromise medical treatment. Coupled with local resistance to relocation, this required ZMOE and TG to remediate Dareta and Yargalma villages before inpatient discharge.

Cleanup of the villages presented numerous resource, logistic, cultural, institutional, and technical challenges. The remote area has little transportation or medical infrastructure. Villages are governed by overlapping civil, tribal, and Sharia law; exhibit sex-segregated social structure; suffer numerous endemic diseases with limited health care; and are supported by a workforce dependent on primitive tools and labor practices. Remediation activities were adapted to local technical and institutional capabilities from protocols developed at the Bunker Hill Superfund Site (BHSS), Idaho, USA (NRC 2005; Sheldrake and Stifelman 2003; U.S. EPA 2002; von Lindern et al. 2003, 2016). The mud-walled residential compounds could not be accessed by mechanical equipment and excavation was accomplished by modifying local hand labor agricultural practices. Remediation personnel adapted to sex, family, and village conventions. For example, initially, Sharia traditions limited compound access to men of the immediate family. Female remediation team members conducted initial characterization and interviews within the homes.

The overall lead health response program followed the model of the BHSS (i.e., curtailment of active sources, BLL testing and appropriate medical and environmental follow-up, remediation of residual soil contamination, and institutional controls to ensure sustainability of the remedy). Here we address adaption of the BHSS remediation and institutional controls to the Zamfara emergency. Details regarding the medical response can be found in studies by Greig et al. (2014) and Thurtle et al. (2014). The remediation program had two primary objectives: *a*) to reduce ongoing lead exposures and *b*) to develop in-country capacity to sustain the remedies and prevent future disasters. The first objective was 2-fold, both to facilitate the MSF/ZMOH chelation program for young children and to simultaneously decrease soil metals exposures for village residents. To sustain the remedies, three subobjectives included developing effective cleanup protocols that local communities could implement, building in-country technical capacity, and developing community awareness of the dangers of artisanal mining.

### 2.2 Methods

The BHSS remediation strategy integrates contaminant removal and clean soil replacement, institutional controls, and lead health advocacy to reduce children's lead intake to acceptable levels (NRC 2005; Sheldrake and Stifelman 2003; U.S. EPA 2002; von Lindern et al. 2003, 2011, 2016). These protocols were adapted for rural Zamfara using existing institutions, employing local labor, and using familiar labor practices, technology, and equipment. For example, traditional farming tools were used to excavate contaminated soils from compounds, which were not accessible by heavy equipment. ZMOE and local government areas (LGA) staff were trained to supervise labor, administer payrolls, and procure materials, supplies, and equipment. The international contingent provided quality assurance/quality control services and was responsible for database management to verify complete contaminant identification and removal.

The project was carried out in three phases: In Phase I, emergency response took place in Dareta and Yargalma villages in June–July 2010; these villages had an estimated combined population of 2,166 and are about 80 km apart, requiring separate operations bases. Phase II included remediation of Abare, Duza, Sunke, Tungar Daji, and Tungar Guru (October 2010–March 2011), with an estimated total population of 6,385 in a 1,400 km2 area requiring three operations bases. Phase III involved the remediation of Bagega (February–July 2013), with an estimated population of 7,323 and one base of operations (von Lindern et al. 2011). Remediation within each village was carried out in four steps: *a*) characterization, *b*) excavation of contaminated media, *c*) replacement with clean soils or concrete, and *d*) waste disposal (von Lindern et al. 2011). The social and technical context of the cleanup required adaptability, and remedial protocols were reevaluated and modified based on experience during all three phases. Examples include developing health messaging for both males and females as well as community engagement efforts; employing village tailors in the production, repair, and laundering of work uniforms; and having excavation hoes manufactured from scrap metal by local blacksmiths.

#### 2.2.1 Risk Assessment

Initial village surveys estimated that > 90% of ongoing lead exposure was attributable to ingestion of lead-contaminated soils and food (Dooyema et al. 2012; von Lindern et al. 2011). Primary sources were contaminated surface soils (generally < 5 cm depth) within the compounds, dusts on surfaces and soft materials (e.g., sleeping mats), and food preparation utensils used in ore processing. Several cubic meters of contaminated waste were identified in shady locations and near water sources throughout the villages where crushing, grinding, drying, sluicing, and storage of ores took place. Also, dangerous concentrations of arsenic, cadmium, mercury and manganese were observed, and these metals would be removed with the lead during remediation (Bartrem et al. 2014).

### 2.2.2 Characterization

Common areas and residential compounds were assessed for lead and other metals *in situ* by modifying BHSS protocols for hand-held X-ray fluorescence (XRF) and by laboratory XRF of bulk and sieved samples of surface soils, dusts, and mining wastes (Bartrem et al. 2014; TerraGraphics 2009; von Lindern et al. 2011). Innov-X Alpha 4000 (Olympus), Innov-X (Olympus) Delta 4000, Innov-X (Olympus) DS 4000, and Niton XL3 (Thermo Fisher Scientific Inc.) models were used. A subset of soil samples was sent to four U.S. laboratories (U.S. EPA Region X, U.S. Geological Survey, University of Idaho Analytical Laboratory Services and Anatek Laboratories, Moscow, ID) for confirmatory analyses by inductively coupled mass spectrometry. Sharia traditions were amended by the local emirate, granting special permission to male staff and village workers to enter the compounds. Hand-drawn maps of homes and common areas included key features and XRF readings. Maps were modified to delineate areas requiring excavation, clean soil and concrete placement, and materials to be cleaned or replaced. Every residence and common area in all villages was sampled. Sample locations focused on the areas identified by the residents, with the objective of identifying locations requiring removal. Typically, 30–150 XRF readings were obtained within a single compound or common area. Sample-site selection was biased to avoiding false negatives (i.e., falsely identifying areas as not requiring removal).

### 2.2.3 Excavation and Replacement

Before compound remediation, residents were required to remove, clean, and store utensils, bedding, and clothing. During Phase I, sleeping mats and carpets were collected, destroyed, and replaced. Areas with surface soil lead concentrations > 1,000 mg/kg were excavated to 5 cm depth. The exposed surface was retested by XRF and excavation continued until criteria (≤ 400 mg/kg Pb) were met (U.S. EPA 2008). Unexcavated areas with soil concentrations > 400 mg/kg and < 1,000 mg/kg were capped with ≥ 8 cm of clean soil obtained from landfill or borrow area excavations. Depending on drainage considerations and clean soil availability, unexcavated soils with ≤ 400 mg/kg Pb were covered with clean soil at the discretion of the project manager. All clean soils were confirmed ≤ 25 mg/kg lead by XRF. No delineation of the clean/contaminated soil interface was provided because foot and animal traffic quickly compacted the surface. Local agricultural hoes were used to scrape the surface backwards (to prevent recontamination) from the walls inward, starting at the rear of each compound and ending at the main entrance. If needed, interior compound walls were brushed to remove contaminated dust before excavation. Contaminated cement floors were capped with new concrete. Excavated soils were shoveled into grain sacks, removed by wheelbarrow, and trucked to landfills by village laborers. Heavy equipment was leased from foreign mining companies operating in the region to construct the waste disposal facilities and clean soil sources, and to excavate public areas. Landfill and health and safety protocols are described below.

In Phases I and II, most exterior removals were done by hand labor due to stability concerns for the mud walls surrounding the compounds and a lack of suitable mechanized equipment. In Phase III, the government obtained small skid-steer loaders that could safely maneuver the narrow streets, facilitating exterior area excavations and waste disposal. Both heavy equipment and hand labor were used to clean several contaminated human-made village ponds. By long-standing practice, village residents had constructed ponds to provide water for livestock and for clay to make bricks for residential compounds in the dry season. These ponds had been exploited for ore sluicing/amalgamation and were severely contaminated with lead and mercury. Bricks produced from the contaminated muck often had lead concentrations exceeding 1%. All bricks, mortar, and plaster were tested by XRF. Most contaminated bricks were identified before being used and were purchased by the ZMOE from the home owner and disposed of in landfills. A small number of contaminated bricks and clay plaster that had been incorporated in homes were capped or removed. The ponds were excavated when completely dry in late February 2011 and April 2013, and were closed or lined with clean soil. Clean soil was delivered to the villages for brickmaking and other construction purposes. Phase III included remediation of a large mining encampment, the Industrial Area, adjacent to the Bagega reservoir. More than 1,000 migrant miners had used several dozen crushing/grinding operations that sluiced ore in the regional water supply reservoir. Several thousand tons of highly contaminated tailings were removed from the Industrial Area. Much of this waste material, which regularly exceeded 10% lead, was relocated to area *dabas* (mining sites located remote from the villages) for reprocessing to recover additional gold. The reservoir, which was also highly contaminated, was drained and dredged.

### 2.2.4 Disposal

Contaminated soil and mine wastes not designated for reprocessing were disposed of in a series of constructed landfills outside each village. Excavated soil from the landfills was used for clean cover material, fill for ponds, and repair of village roads. Landfills were sited in consultation with ZMOE and village elders familiar with seasonal groundwater levels, water holding capacity, and the structural characteristics of local soils. Landfills were typically 10 m wide and 5 m deep, and extended 30 to > 50 m in length. Those landfills containing contaminated soil < 1% lead were bottom-lined with compacted clay and permanently closed with a 1-m compacted clay cap. Phase III landfills accepting highly contaminated wastes from the Industrial Area were additionally bottom-lined with a polyvinyl chloride (PVC) liner of undetermined thickness purchased and provided by the FMOE. The landfills were delineated by global positioning system (GPS) and closed with permanent monuments dedicated to the children who died in the epidemic.

### 2.2.5 Health and safety

Health and safety protocols for all personnel, workers, and the public were implemented in Phase I and progressively enhanced throughout the cleanup. Training conducted in each village included reviewing remediation and health and safety protocols emphasizing hygiene, construction safety, and decontamination practices. Village health and safety managers were appointed to ensure that best practices were implemented. In Phases II and III, facilities were constructed for workers to shower, change clothes, and clean footwear prior to leaving the cleanup sites. All laborers participating in excavation or disposal of contaminated materials were provided clean work clothes and dust control masks daily. Lunch was prepared and served to all project participants in specially constructed areas with ample potable water for drinking and personal hygiene. All workers were required to wash prior to being served lunch. Worker blood lead levels were monitored by the ZMOH. In addition to protecting the workers and communities during remediation, health and safety protocols provided model practices for miners and their families, as many of the cleanup laborers were also engaged in mining.

### 2.2.6 Implementation and Advocacy Campaign

Phase I remediation commenced as an emergency response in Dareta and Yargalma, in June 2010. TG and ZMOE developed and implemented emergency remediation plans while MSF and ZMOH established village clinics. The cleanup was conducted by ZMOE, with TG providing technical guidance. Funding and equipment came from Zamfara State, TG, MSF, and Blacksmith Institute.

Security and logistical support for the international contingent was provided by the Zamfara State government and MSF (von Lindern et al. 2011). Phase I was completed in mid-July. Phase II remediation commenced in October 2010 with funding from the United Nations Central Emergency Response Fund, United Nations Children's Fund, Zamfara State, TG, and BI. Phase II, also conducted by ZMOE with TG oversight, addressed five villages (Abare, Duza, Sunke, Tungar Daji, and Tungar Guru). Remediation activities were completed in March 2011. Preliminary characterization in Bagega was suspended from March 2011 until February 2013 due to lack of funding and security concerns related to the Nigerian presidential election.

MSF initiated blood lead screening and chelation therapy as the remediation program progressed (Greig et al. 2014: Thurtle et al. 2014). However, in May 2011, MSF noted increasing BLLs in a small number of individual chelation patients and suspected lead poisoning in the deaths of two children in remediated villages. Follow-up environmental investigations by TG and ZMOE revealed resumption of mineral processing and recontamination in isolated locations in two villages. Out of concern for recontamination issues, MSF and TIFO initiated an advocacy campaign to persuade the Nigerian federal, state, and local governments to undertake Phase III remediation of Bagega and establish programs to sustain the remedy and promote safer mining. A three-part proposal advocating a) MSF establish an outpatient chelation clinic for the children of Bagega, predicated on b) remediation of Bagega by the Nigerian federal government under TIFO guidance and certification, and c) development of a safer mining program in Zamfara. The proposal was presented to representatives of both Zamfara State and Federal Ministries of Health, Environment, Mining and Solid Minerals, and LGA officials at a conference sponsored by MSF and TIFO, originally scheduled for January 2012 in Abuja, but delayed until May 2012 due to civil unrest in the country (MSF 2012). Numerous delays in the release of cleanup funds postponed the start of Phase III remediation. Significant media attention and pressure came from international and Nigerian nongovernmental organizations (NGOs) (Follow the Money 2012; Human Rights Watch 2012; Murdock 2012). MSF and TIFO maintained permanent staff in Nigeria to conduct negotiations regarding project roles and responsibilities, and to secure the release of funds. In late January 2013, Nigerian President Goodluck Jonathan released $3.2 million to FMOE to undertake the Bagega cleanup, with the bulk of the work to be accomplished by the newly established Zamfara Environmental Sanitation Agency (ZESA), LGA, and local village labor. TIFO provided technical guidance and assistance. Upon certification of completion by TIFO, MSF opened clinics and began BLL testing and treatment. In addition, the Nigerian government allocated $1.1 million to initiate a safer mining program (Abatu 2014).

### 2.2.7 Human Subjects, Data Sources, and Analyses

Environmental and demographic data and analyses were developed from characterization and construction reports obtained during investigation and remediation activities. Permission to test homes for heavy metal contamination was obtained from residents by Nigerian government representatives by informed consent (TerraGraphics, unpublished data). All environmental and health data presented are de-identified. Numbers of children tested and BLLs were provided by MSF. MSF blood lead data were collected during life-saving medical intervention, and met the standards set by the independent MSF Ethics Review Board for retrospective analyses of routinely collected programmatic data. Review of anonymous, routinely collected programmatic data does not constitute research under the Nigerian National Health Research Ethics Committee guidelines (MSF 2013). CDC blood lead data were collected during medical intervention in collaboration with the FMOH in accordance with the Declaration of Helsinki (Dooyema et al. 2012).

Environmental exposure estimates were developed consistent with the BHSS cleanup model. Every compound in each village was tested by XRF, and residential soil exposure is defined as the average of all surface soil lead concentrations obtained in a compound. The aggregate community soil lead exposure is calculated by averaging mean soil concentrations from all compounds in a village. Percentage reductions and analysis of variance in residential soil exposure were calculated by comparing the pre- and postremediation community mean concentrations (SAS Institute Inc. 2008). Because remediation and soil exposure reductions were prerequisite to chelation treatment, temporal mean first-draw, pretreatment BLL results for children ≤ 5 years of age are presented for context. Temporal means reflect project phases, which were determined by medical urgency, climatic conditions, funding, and logistic factors. MSF BLL screening targeted all children ≤ 5 years in all villages regardless of exposure (Greig et al. 2014; Thurtle et al. 2014).

## 2.3 Results

Overall, project remediation achieved 77–98% reductions in mean residential soil lead concentrations by village to maximum levels < 400 mg/kg U.S. criteria (U.S. EPA 2008). Soil lead exposures were reduced for an estimated 17,000 residents in eight villages. Testing of all 944 compounds in the eight villages showed pre- and postremediation lead concentrations for individual compounds ranged from 19 mg/kg to 35,380 mg/kg, and 13 mg/kg to 400 mg/kg, respectively. Mean soil concentrations by village ranged from 300 mg/kg to 4,143 mg/kg preremediation and 70 mg/kg to 179 mg/kg, postremediation, with overall means of 1,311 mg/kg and 94 mg/kg, respectively. A total of 820 compounds, 181 common areas, 31 ponds, and the reservoir and Industrial Area at Bagega were remediated (Table 1). Figure 1 shows geometric mean first-draw BLLs for all 4,399 children ≤ 5 years old MSF screened against the chelation threshold (≥ 45 µg/dL) (Greig et al. 2014; Thurtle et al. 2014). Of these 2,349 were provided chelation (Table 2) and MSF observed geometric mean preremediation BLLs decrease from 149 µg/dL to 15 µg/dL over the 4-year cleanup (Figure 1). Phases I, II, and III accomplished 18%, 39%, and 43%, respectively, of the total number of residential compounds remediated, costing an estimated $400,000, $1.9 million, and $3.2 million U.S. dollars, respectively, according to budget estimates presented to the Nigerian government (MSF 2012; von Lindern et al. 2011; TerraGraphics, unpublished data). A total of 27,390 $m^3$ of soil and wastes were excavated and disposed of in 14 landfills. Collectively, an estimated 187,000 kg of lead were removed from eight villages and the processing areas (Table 3). More than 43,000 $m^3$ of clean soil was imported as replacement fill and cover.

Phase I remediation included 85 residential compounds and 13 common areas in Dareta, and 63 compounds and 11 common areas in Yargalma. Mean soil lead concentrations were reduced by 98% and 96%, to 83 mg/kg and 179 mg/kg, respectively (Table 1). A total of 3,919 m3 of contaminated soils and waste, including 300 m3 of 3.2% lead process waste, were excavated and disposed in two landfills (Table 3). MSF initiated blood lead testing concurrent with Phase I remediation. The 74 children screened before and during remediation in May–June 2010 had a mean BLL of 149 µg/dL; 230 children screened in July–August 2010 after Phase I remediation had a mean BLL of 76 µg/dL (Figure 1). MSF provided inpatient chelation to 282 children ≤ 5 years of age from Dareta and Yargalma during Phase I, who were able to return to remediated homes between June and September 2010 (Table 2).

Phase II remediation addressed 320 residential compounds, 103 common areas, and 23 processing ponds in five villages reducing soil lead exposures for an estimated 6,385 residents (von Lindern et al. 2011). Additionally, removal of highly contaminated materials from seven ponds in Yargalma and Dareta, and closure of landfills left open from Phase I were accomplished during Phase II (Table 1). Mean preremediation residential soil lead concentrations in Phase II villages ranged from 300 mg/kg in Duza to 1,343 mg/kg in Abare, and were reduced by 77% and 93%, respectively, postremediation. Five landfills, accommodating 8,981 m3 of excavated waste and contaminated soil, were constructed and permanently closed (Table 3). MSF introduced outpatient clinics during Phase II, screening 3,326 children as remediation was completed (Figure 1), and provided chelation to 1,920 children (Table 2).

During Phase II, local government and community activities were initiated to develop technical skills and environmental response capacity in Zamfara. Separate male and female advocacy/environmental health promotion teams were established to facilitate remediation and prevent recontamination. Technology transfer, technical training, and certifications were provided to > 200 ZMOE, LGA, and village personnel. Additionally, nearly one-third of the compounds, common areas, and mineral processing locations in Bagega were characterized, and preliminary design and cost estimates were prepared during Phase II. A 2,000 m3 landfill was constructed for Phase III remediation. Supplemental investigations by the CDC and Nigerian authorities during Phase II suggested that artisanal gold mining was occurring in an additional 114 villages in three LGAs. Significant soil contamination (> 400 mg/kg Pb) was found in approximately half of 74 of those 114 villages sampled (Bashir et al. 2014; Lo et al. 2012). Another study revealed metals contamination of surface water and open wells where ore processing had occurred (UNEP/OCHA 2010).

In Bagega during Phase III, 352 compounds, 54 common areas and one pond were remediated (Table 1). A total of 5,090 m3 of contaminated soils from compounds and village common areas was disposed of in three landfills. Excavation of the Industrial Area and reservoir produced 8,700 m3 and 700 m3 of contaminated waste, respectively. These wastes ranged from 2.5% to > 10% lead *in situ* (data not shown). However, the disposed materials averaged 1.0% and 0.8% lead, respectively, because higher concentration materials were removed for reprocessing and the remaining materials were co-disposed with underlying contaminated soils (Table 3). Five kilometers of village roads were graded and capped with laterite soil generated from borrow areas. Approximately 10,000 m3 of clean soil used to cap excavated compounds, common areas, the Industrial Area, and newly graded village roads; for construction and brickmaking; and to backfill a large pond that was a drowning hazard for children.

In Phase III, mean soil lead concentrations for all compounds were reduced by 87% from 670 mg/kg to 90 mg/kg. Bagega, with an estimated 7,323 people during 2011, was remediated sequentially in four geographic subareas or quadrants, allowing MSF to commence blood lead screening and outpatient chelation for children in the first quadrant in March 2013. The last quadrant was remediated in July. The mean BLL of 564 children screened in April–August was 25 µg/dL, and for 205 children screened in September–December was 15 µg/dL (Figure 1). Chelation was provided to 236 children with BLLs ≥ 45 µg/dL (Table 2) (Greig et al. 2014). Phase III remediation was accomplished largely by Nigerian personnel trained during Phases I and II. A total of 78 environmental professionals from FMOE, ZMOE, and LGAs directed and supervised all remediation activities including procurement and logistics. More than 300 local community members were trained. Local businesses provided supplies and equipment. Two on-site expatriates participated in Phase III activities, compared with 16 and 28 during Phases I and II, respectively.

Analyses of variance revealed that preremediation soil exposures differed significantly by phase and village ($p < 0.0001$). Mean preremediation soil lead concentrations in Phase II villages (951 mg/kg) tested from October 2010 to March 2011 were 74% lower than May–June 2010 Phase I village levels (3,728 mg/kg) (Table 1). Mean preremediation soil lead levels tested in about one-third of Bagega compounds during Phase II in February 2011 showed concentrations (1,059 mg/kg not shown) (TerraGraphics, unpublished data) similar to those in Phase II villages, but were 670 mg/kg, or 37% lower, by 2013.

## 2.4 Discussion

Zamfara is a severe example of an evolving trend of the world's poorest, most remote and vulnerable populations becoming unwitting hosts to environmental and occupational disease. BLLs and mortality rates were unprecedented, exacerbated by risk cofactors that complicated health and environmental response actions (Moszynski 2010; MSF 2010c). Malnutrition and childhood pestilent diseases, including measles, mumps, meningitis, polio, malaria, and cholera, are endemic among these populations (MSF 2010a; Ogwumike et al. 2012). Health care services are largely nonexistent in the villages. During Phase II, characterization and remediation were conducted in the midst of a cholera epidemic, with > 200 children simultaneously treated for cholera and lead poisoning (Shaffi M, Medical Coordinator, MSF, personal communication, 1 October 2011). Intervention and remediation efforts were hampered by poor infrastructure, as the villages lack electricity and running water, are 1–5 hr travel from the nearest paved roads, and are largely inaccessible in the rainy season. Supplies and equipment had to be procured and serviced from hundreds of kilometers away. Religious and cultural practices required strict separation of adults along sex lines, parallel male and female response teams, and in some situations, suspension of Sharia law. The cash-only economy made the transport and distribution of the large sums of money necessary for payroll, local supplies, and services a logistically complicated and dangerous undertaking. Corruption in the civil government is endemic (Transparency International 2014). Crime and terrorism are constant concerns (U.S. Department of State 2015).

Despite these challenges, cleanup objectives were achieved through adaptation of established U.S. protocols. The soil lead concentrations in Table 1 include every compound in all eight villages and are directly comparable with the 400 mg/kg U.S. standard and are equivalent to the residential soil exposures defined in the BHSS cleanup model. The BLLs in Figure 1 are from nearly the entire population of children ≤ 5 years old. CDC surveyed the Phase I population that MSF screened in May 2010, testing 59% of 345 total surviving children ≤ 5 years old identified in the villages. CDC reported mean BLLs of 130 µg/dL for 86 children in the same two villages (Dooyema et al. 2012), similar to the 149 µg/dL observed by MSF. Population surveys conducted by MSF and TG estimated that 88% of all children ≤ 5 years age in both Phase I and II villages had been screened by January 2011 (von Lindern 2011). As a result, residential soil lead concentrations are an effective exposure metric for these populations, and mean BLLs in Figure 1 are an indicator of the severity of the epidemic at the time.

The emergency measures taken to reduce blood lead levels and environmental exposures (i.e., relocation of mineral processing, remediation, clean drinking water and food sources, chelation) markedly decreased mortality, from 25% in the 6 months preceding May 2010 to < 2.5% by September 2010, significantly reducing the risk of adverse health effects to the resident populations (Dooyema et al. 2012; Greig et al. 2014; Thurtle et al. 2014). Since remediation, new village residents and children born to mothers with low body burden are not experiencing high residential soil lead exposures (i.e., > 400 mg/kg). Similar reductions in other toxic metal concentrations (arsenic, cadmium, and mercury) were achieved simultaneously (Bartrem et al. 2014).

The analysis of variance results suggests preremediation soil lead exposures decreased over time, likely due to both environmental and anthropogenic factors. Phase I and II soil sampling was conducted 1–2 months and 6–12 months following cessation of mineral processing, respectively, with an intervening rainy season; and in Bagega, 9 months and 3 years later. Limited blood lead testing, mortality reports and soil tests conducted in short-term visits to the Phase II villages during Phase I (before the rainy season) suggest that soil lead concentrations and exposures were similar to Dareta and Yargalma in three of the five Phase II villages and in Bagega (TG, unpublished data). In August/September 2010, CDC Nigeria tested 185 children ≤ 5 years old in Bagega; 59% had BLLs > 65 µg/dL (CDC Nigeria, unpublished data). Initially, soil lead concentrations following the relocation of mineral processing varied by village, likely reflecting the intensity of high-lead ore utilization. Environmental factors may have included natural dilution from soil accumulation, runoff during the highly erosive rainy seasons, and wind erosion from seasonal *harmattans* (dust storms). Socially, inadvertent remediation occurred through maintenance, repair, and construction of the mud-brick homes and regular sweeping of floors. Additionally, some families in Bagega remediated their own compounds by scraping the top layer of soil in areas identified as contaminated during the 2010/2011 Phase II characterization effort. These families placed the contaminated soil in sacks and disposed of it at the new ore processing site outside of the village.

These factors suggest that residential soil exposures and BLLs in these villages were much higher in May 2010 than was observed when the remediation and blood lead screening program reached the Phase II villages 6 months later and in Bagega nearly 3 years later. Unfortunately, during the epidemic, nearly every village resident tested showed dangerously high blood lead levels, resulting in deaths, significant adverse health effects among survivors, and continuing body burdens of lead that may require years to equilibrate (Greig et al. 2014; Thurtle et al. 2014). Because the fetal skeleton develops from the mother's bone store, and newborn BLLs approximate maternal burden, the current population of mothers and young women present an especial risk to future generations (Ettinger et al. 2007; Miranda et al. 2010; Riess and Halm 2007). Potential health effects associated with blood lead levels observed in this epidemic include adverse reproductive and child development outcomes, and irreversible neuropsychological effects ranging from severe brain damage resulting in permanent dysfunction to depressed mental capacity, impairment of nerve function, behavioral and learning problems, loss of quality of life, and inability to participate in or meet village social obligations (Thurtle et al. 2014; U.S. EPA 2006, 2013). There is also a range of possible damage to other organ systems (ATSDR 2007; Caravanos et al. 2013; Lanphear et al. 2005; U.S. EPA 2006, 2013). Because treatment was unavailable for children > 5 years or for adults, entire generations of village residents are potentially suffering lifelong debilitating effects (Needleman et al. 1990; Reyes 2007).

To manage and sustain the remedy and to undertake future cleanup activities, technical capacity was transferred to local entities. Throughout the cleanup, local communities became increasingly aware of the dangers of artisanal mining and measures necessary to protect their families. Several hundred community members were employed and acquired experience in implementing remedial protocols. The Nigerian federal government assumed responsibility to fund remediation and regulate artisanal mining throughout the country. ZESA was created to undertake cleanup activities and regulate pollution from artisanal mining (Sada I, Director General, ZESA, personal communication, 1 February 2011). Local government and emirate officials established committees to address artisanal mining, discourage resumption of dangerous activities, and prevent recontamination in the villages.

Nevertheless, the economic contribution of small-scale mining to village livelihood poses a continuing recontamination threat. Alleviating this requires all stakeholders to engage in sustainable region-wide safer mining practices. Institutional controls implemented in 2011–2013 included no longer employing women in ore processing, relocating *dabas* sufficient distances to prevent visits by children, and requiring self-remediation of recontaminated compounds (von Lindern et al. 2011). Wet milling was introduced in 2013–2014 with recommendations to wash and change clothes before going home. Longer-term efforts underway with Nigerian authorities include completing remediation in other villages; developing monitoring, maintenance, periodic reassessment, advocacy, and public health information programs; and establishing safer mining practices (Anka SM, Director, Pollution Control ZESA, personal communication, 3 June 2013; ELI 2014).

**2.5 Conclusions**

Despite complex challenges, substantial remediation and subsequent medical interventions were accomplished by adapting established health and environmental response protocols to local conditions and capabilities. These efforts, combined with cessation of processing in the villages, provision of clean drinking water, decreased contamination of the food supply, natural attenuation of soil lead concentrations, and behavioral changes, effected 77–98% decreases in soil lead exposures (Table 1) and > 100 µg/dL reductions in mean BLLs (Figure 1). The capacity, authority, funding, and responsibility for the cleanup were transferred to the Nigerian federal, state, and local governments. A cadre of Zamfara State and local government staff was trained to manage and supervise the remediation and undertake sustainable programs to prevent future epidemics. Engagement of villagers and community leaders developed awareness of the dangers of artisanal mining and protective measures families can employ. Sustaining the remedy will require the Nigerians refraining from mineral processing in the villages, and developing and enforcing safer mining practices. This tragic incident and subsequent response demonstrate that, with sufficient political will and modest investment, even the world's most challenging environmental health crises can be addressed and resolved within the capabilities of the host countries.

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Table 2.1. Pre- and postremediation soil Pb concentrations and range of residential compounds (cpds), common areas (CAs), and ponds.

| Village | Phase | N cpds tested | N cpds remediated | N CAs remediated | N ponds remediated | Pre-rem mean Pb (range) of cpds tested (mg/kg) | Post-rem mean Pb (range) of cpds tested (mg/kg) | % conc. reduction |
|---|---|---|---|---|---|---|---|---|
| Dareta | I | 94 | 85 | 13 | 4 | 3,436 (40–35380) | 83 (25–252) | 98 |
| Yargalma | I | 66 | 63 | 11 | 3 | 4143 (83–23296) | 179 (25–400) | 96 |
| *Phase I total* | | *160* | *148* | *24* | *7* | *3,728* | *123* | *97* |
| Abare | II | 96 | 74 | 20 | 0 | 1343 (43–18921) | 90 (25–400) | 93 |
| Tungar Guru | II | 38 | 31 | 6 | 1 | 874 (85–4,446) | 83 (23–321) | 91 |
| Sunke | II | 93 | 83 | 38 | 10 | 1119 (19–9688) | 106 (25–400) | 91 |
| Tungar Daji | II | 78 | 75 | 31 | 10 | 780 (59–4952) | 72 (25–235) | 91 |
| Duza | II | 57 | 57 | 8 | 2 | 300 (24–1779) | 70 (25–209) | 77 |
| *Phase II total* | | *362* | *320* | *103* | *23* | *951* | *86* | *91* |
| Bagega | III | 423 | 352 | 54 | 1 | 670 (18–20748) | 90 (13–400) | 87 |
| *Phase I, II, and III total* | | *944* | *820* | *181* | *31* | *1311* | *94* | *89* |

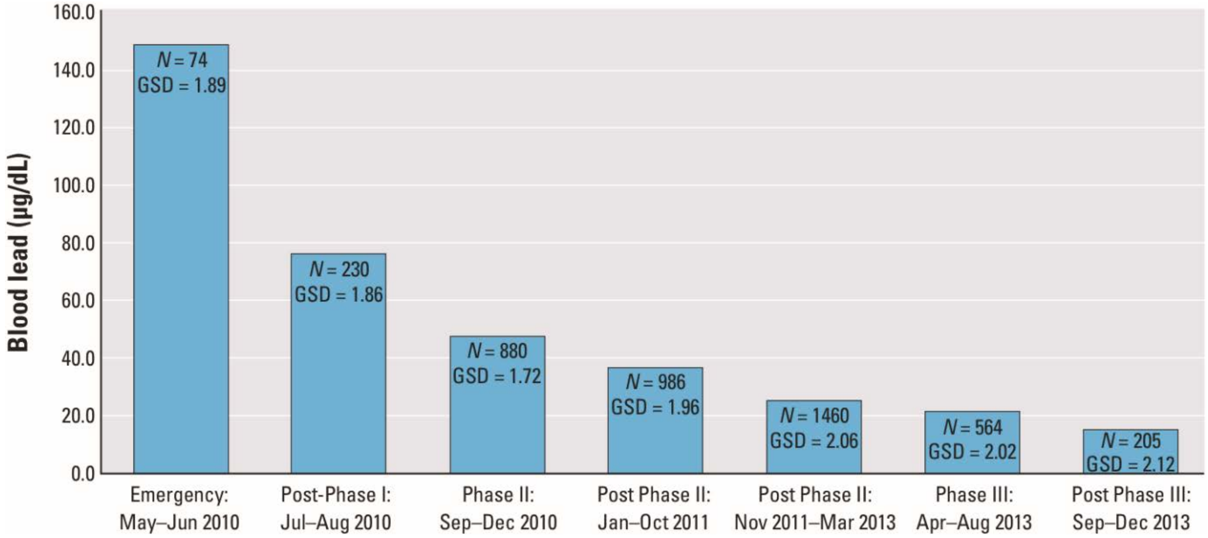
Table 2.2. Number of ≤ 5-year-old children provided oral chelation treatment following remediation phases. Data from Médecins Sans Frontières (Greig J, personal communication, Operational Epidemiologist, Médecins Sans Frontières).

| Village | Phase I June–Sept 2010 | Phase II Oct–Dec 2010 | Phase II 2011 | Post Phase II 2012 | Phase III 2013 | Total treated |
|---|---|---|---|---|---|---|
| Abare | 10 | 208 | 255 | 86 | 84 | 633 |
| Bagega | 5 | 6 | 0 | 1 | 236 | 243 |
| Dareta | 101 | 182 | 86 | 51 | 53 | 372 |
| Duza | 0 | 1 | 53 | 2 | 0 | 56 |
| Sunke | 23 | 81 | 161 | 17 | 25 | 284 |
| Tungar Daji | 0 | 5 | 196 | 23 | 4 | 228 |
| Tungar Guru | 22 | 107 | 24 | 7 | 5 | 143 |
| Yargalma | 181 | 268 | 70 | 30 | 22 | 390 |
| Total | 342 | 858 | 845 | 217 | 429 | 2,349 |

Table 2.3. Excavation/disposal volumes ($m^3$), Pb concentrations (mg/kg), and total Pb (kg) by phase. Abbreviations: conc, concentration; Pb, lead; vol, volume. Calculation uses soil bulk density of 1,600 kg/m3 with 30% bulking factor (http://www.engineeringtoolbox.com/soil-rock-bulking-factor-d_1557.html).

| Area | Phase I | | Phase II | | Phase III | | Total disposed | |
|---|---|---|---|---|---|---|---|---|
| | Vol ($m^3$) | Pb conc (mg/kg) | Vol ($m^3$) | Pb conc (mg/kg) | Vol ($m^3$) | Pb conc (mg/kg) | Vol ($m^3$) | Pb (kg) |
| Compounds | 2,602 | 3,863 | 5,183 | 1,029 | 3,343 | 670 | 11,128 | 21,502 |
| Common areas | 417 | 2,649 | 2,418 | 2,688 | 1,747 | 560 | 4,582 | 10,471 |
| Ponds/reservoirs | 600 | 11,280 | 1,380 | 13,100 | 700 | 8,000 | 2,680 | 37,144 |
| Process waste | 300 | 32,000 | N/A | N/A | 8,700 | 10,000 | 9,000 | 117,852 |
| *Total* | *3,919* | | *8,981* | | *14,490* | | *27,390* | *186,969* |

Figure 2.1. Geometric mean blood lead levels (µg/dL) for initial draw following remediation (prior to chelation) for 0-5-year-old children by cleanup phase (MSF 2014).



# CHAPTER 3 – FOOD CONTAMINATION AS A PATHWAY FOR LEAD EXPOSURE IN CHILDREN DURING THE 2010-2013 LEAD POISONING EPIDEMIC IN ZAMFARA, NIGERIA

Tirima, S., Bartrem, C., von Lindern, I., von Braun, M., Lind, D., Mohammed Anka, S., Abdullahi, A.

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**Abstract**

**Background:** In 2010, an estimated 400 to 500 children died of acute lead poisoning associated with artisanal gold mining in Zamfara, Nigeria. Processing of gold ores containing up to 10% lead within residential compounds put residents, especially children, at the highest risk. The principal routes of exposure were incidental ingestion and inhalation of contaminated soil and dusts. Several Nigerian and international health organizations collaborated to reduce lead exposures through environmental remediation and medical treatment. The contribution of contaminated food to total lead exposure was assessed during the environmental health response. **Objectives:** Assess the role of cultural/dietary habits on lead exposure pathways and estimate the contribution of contaminated food to children's blood lead levels (BLLs). **Methods:** A survey of village dietary practices and staple food lead content was conducted to determine dietary composition, caloric intakes, and lead intake. Potential blood lead increments were estimated using bio-kinetic modeling techniques. **Results:** Most dietary lead exposure was associated with contamination of staple cereal grains and legumes during post-harvest processing and preparation in contaminated homes. Average post-harvest and processed cereal grain lead levels were 0.32 mg/kg and 0.85 mg/kg dry weight, respectively. Lead ingestion and absorption were likely aggravated by the dusty environment, fasting between meals, and nutritional deficiencies. **Conclusions:** Contamination of staple cereal grains by highly bioavailable pulverized ores accounted for 11-34% of children's BLLs during the epidemic, and as a continuing source after residential soil remediation until stored grain inventories were exhausted.

## 3.1 Introduction

According to the World Health Organization (WHO), the 2010 Zamfara lead poisoning epidemic was an "unprecedented environmental emergency" (Moszynski, 2010) with soil lead levels exceeding 100,000 mg/kg (10%) and individual venous blood lead levels greater than 400 µg/dl (Bartrem et al., 2014; Dooyema et al., 2011; Lo et al., 2012; Plumlee et al., 2013; von Lindern et al., 2011). Within a few months of the outbreak, an estimated 400 children age five years and younger died of acute lead poisoning. Thousands more were severely poisoned and at risk of incurring irreversible neurocognitive damage with geometric mean blood lead levels (BLLs) exceeding 149 µg/dl (Burki, 2012; Greig et al., 2014; Thurtle et al., 2014; Tirima et al., 2016). Emergency response efforts, commencing in two villages in June 2010, included chelation therapy for children ≤ 5 years old conditioned upon remediation of their homes to preclude continuing exposure. By March 2011, the Zamfara Ministry of Environment (ZMOE) with guidance from the United States (US) based TerraGraphics Environmental Engineering (TG) had completed Phases I and II cleanup activities, remediating 468 residential compounds in seven villages and facilitating chelation therapy for 2146 children by Médecins Sans Frontières (MSF). In 2013, another 352 homes in Bagega village were remediated during Phase III. Overall, soil and dust exposures were reduced by 77-98% for more than 17,000 villagers. More than 2300 children received chelation therapy that reduced individual BLLs to <30 µg/dl (Tirima et al., 2016; Greig et al., 2014; Thurtle et al., 2014).

Investigations during February 2011 pre-remediation sampling in Bagega village and July 2011 remedial effectiveness evaluations (REEs) in remediated villages provided a more detailed understanding of secondary exposure pathways and risk co-factors (Bartrem et al., 2017; Tirima et al., 2016). These surveys revealed the food supply was compromised through interactions between artisanal mineral exploitation and indigenous agricultural labor practices. Additional efforts to quantify the dietary exposure pathway continued in 2012. These findings were important in modifying subsequent remedial sustainability, environmental health, medical, advocacy, and institutional responses to the epidemic (Bartrem et al., 2017; Tirima et al., 2016). This manuscript focuses on the assessment of pre-remediation lead exposures from food in Bagega village in 2011-2012.

### 3.2 Background

#### 3.2.1 Mining Practices

Gold deposits have been exploited in northern Nigeria since colonial times. A gold rush took place in the 1930s, mostly by European entrepreneurs using primitive mining methods (Ochonu, 2009). As gold prices increased in the late 2000s, the legacy colonial mines were revisited and developed into commercially viable sites. Artisanal mining practices in remote areas world-wide are largely driven by poverty (Mallo, 2012). Zamfara rural residents are extremely poor, subsisting on less than $2/day, with little formal education and limited employment alternatives (IFAD, 2010). Subsistence agriculture is the main economic activity in this semi-arid region and climate change is negatively impacting both crop and livestock production (Farauta et al., 2012; Odjugo et al., 2009). In 2011, Zamfara State had the highest unemployment rate in Nigeria at 42% (Royal Times Nigeria, 2016), and village rates are likely higher as agricultural sector jobs are seasonal (Nnaji, 2001). Consequently, the "gold rush" beginning in 2008-09 was a welcome economic relief, albeit with catastrophic environmental health consequences.

Despite large revenues from this gold trade, little investment in improved mining infrastructure or technology followed (WHO, 2011). Zamfara artisanal miners use low cost milling and gravity concentration during ore processing. Manual extraction of gold from rock involves "pounding" the ore to gravel-like consistency using hammers and locally designed mortars and pestles. The crushed material is then ground using modified steel flour mills. Generally, 50 to 200 mesh (75-300 µm) uniform powdered ores are produced, mixed with water and sluiced to obtain a gold concentrate, which is then amalgamated with mercury. The mercury is evaporated by torch, leaving an unrefined sponge gold nugget. Dry processing produces enormous quantities of dust, which were deposited throughout the villages and residential areas by wind, foot traffic, and direct disposal of tailings. The use of mercury also resulted in soil, dust, water, food, and vapor exposures.

### 3.2.2 Cultural and Dietary Considerations

Before traditional leaders banned the practice in the villages in May 2010, ore processing took place in residential compounds and village public areas. The situation was exacerbated by a religious/cultural practice called *purdah* (or *auren kulle* in Hausa), which involves the sequestration of married women in residential compounds. In order to employ women in mining operations, ores were brought into the compounds for processing, resulting in widespread contamination of living areas and throughout the villages. The Hausa household (*gida*) is a family farming unit, often containing multiple families of several generations and is the fundamental unit of residence, production, distribution, transmission of culture, and reproduction (Adamu, 2009). Walled on the outside, with a gradation of space from *public* to *private* on the inside, the *gidas* express the gendering of space and the importance of sequestering women (Pellow, 2002).

Most post-harvest food processing is accomplished by women and children within the *gida*. Grains are dried and hand-threshed by beating on the ground and wind winnowing, and then stored in specially built ovoid mud walled granaries. Most residential compounds are constructed from soil and thatch, though a few homes have cement floors and corrugated iron sheet roofing. In some homes, adobe bricks and plaster were made using contaminated ore tailings mixed with mud. Almost all food is processed within the compound by the women, often in the same contaminated areas where ores were processed. Women sometimes pounded ore with the same mortars and pestles used to prepare food. Flour mills were used for grinding both grain and ores. Box sluicing often took place inside compounds leading to the contamination of residential wells. As a result, the likelihood of contaminating food with ores and tailings was extremely high.

### 3.2.3 Food Production and Post-Harvest Processing in Zamfara

Most small-holder agriculture in Zamfara is carried out during the single rainy season from April/May through October. Staple crops such as millet (*Pennisetum*), guinea corn (sorghum, *Sorghum bicolor*) maize (corn, *Zea mays*) and rice (*Oryza sativa*) are intercropped with legumes including cowpeas (*Vigna unguiculata*), groundnuts (*Arachis hypogaea*) and soybeans (*glycine max*). Tomatoes, hot peppers, onions, and cabbage are grown both during the rainy season and the dry season under irrigation (Bush, 2013; Ene-Obong et al., 2013). Harvest begins in September and continues through December depending on crop type. Dry season harvest from irrigated crops usually ends in March. Some families exhaust home-grown supplies by late January and purchase additional foodstuffs from wealthier neighbors or weekly markets. There is limited dietary diversity during the food shortfall period. Families often supplement their diet with foraged foods such as baobab leaves and other wild plants (Bush, 2013).

The post-harvest system for grains and opportunities for contamination are shown in Figure 1. Most grains and pulses are left to dry in the field. Zamfara farmers sell surplus grain through the rural assemblers, wholesalers, retailers, and consumers as well as through local processors. Traditionally, un-threshed grains are stored in solid mud walled silos (*rumbus*), and may be held for up to three years before being consumed or sold. Primary processing of grains includes cleaning, hulling, pounding, milling, grinding, tempering, soaking, parboiling, drying, and sieving. Secondary processing involves baking, frying, cooking, extruding, blending, fermenting, and roasting. Each step introduces potential for contaminated soil/dust to enter the food supply. Conversely, cleaning and washing of larger seeds (maize, beans, local rice) prior to cooking can remove contaminated dusts before consumption if uncontaminated water is used. Children usually eat with their hands from common bowls placed on the floor where soil and dusts can readily contaminate the food.

### 3.3. Objectives

The objectives of this investigation were to: i) explore the role of cultural/dietary habits and ore processing activities on lead exposure pathways, and; ii) estimate the contribution of contaminated food to children's blood lead levels.

### 3.4. Materials and Methods

#### 3.4.1 Bagega Village Characterization 2011

In 2009-2010, Bagega village (population 8500) hosted an adjacent mining camp utilizing the regional water reservoir for sluicing gold ores, which contrasted to the artisanal practices in the other villages, operated on an industrial scale (Tirima et al., 2016; von Lindern et al., 2011). This site was inaccessible and unknown during the 2010 Phase I remediation and was not included in any Phase II cleanup funding requests. By November 2010, the camp was abandoned leaving behind thousands of cubic meters of highly contaminated waste, a polluted reservoir, and hundreds of homes and public areas poisoned by ore processing. Bagega village and the adjacent industrial area were extensively surveyed in February 2011 by portable x-ray fluorescence (PXRF). Emergency life-saving treatment protocols were implemented by MSF until remediation could be undertaken. Water and mercury exposures were assessed under a coordinated effort by the United Nations, the Dutch government, and Nigerian environmental authorities (UNEP/OCHA, 2010). More than two years were required to secure the necessary funds from the Nigerian government and Bagega was remediated in 2013 (Tirima et al., 2016).

#### 3.4.2 Food Samples and Lead Content

During the 2011 Bagega pre-remediation characterization, several commonly consumed food samples were opportunistically sampled from households and rural and regional markets by TG and Centers for Disease Control and Prevention (CDC) personnel. Thirteen (13) samples were obtained from the Anka regional market, 11 from the Bagega regional market, 2 from the Bagega bakery, and 9 from 5 randomly selected homes in Bagega village. Samples included raw food from storage bins, food processed at home or at market (e.g., by grinding with mortar/pestle or flourmill), and prepared food. The type and quantity of food samples shipped to the US for analysis were limited because of sample handling and export/import regulations (e.g., no dairy products could be analyzed). Lead concentration and *in vitro* bioaccessibility were analyzed by the US Geological Survey (USGS) by inductively coupled plasma-atomic emission spectroscopy (ICP-AES) (Plumlee et al., 2013). Soil, dust and ore lead concentration and *in vitro* bioavailability were estimated by US Environmental Protection Agency (USEPA) Methods 3050b and 9200B, respectively (USEPA, 2007a, 2007b).

### 3.4.3 Diet Composition Questionnaire and Caloric Intakes

In 2012, dietary intake estimates were developed for individual children ages 12-60 months from 29 different families. Data were obtained using a culturally sensitive food frequency questionnaire (FFQ) adapted to the local context in collaboration with area health and environment workers and community members (Dehghan et al., 2005; MacIntyre and Labadarios, 1999; Osowski et al., 2007; Scales et al., 2013; Teufel, 1997). A list of commonly prepared foods was generated by local health authorities and augmented after discussion with village residents. FFQs were administered to parents in the Hausa language by local health teams assisted by international staff. Respondents were shown common utensils and containers (825 ml, 500 ml, 100 ml, 60 ml) and asked for each food type to indicate the appropriate container size and number of times per day, week, and month that the specific food item was consumed. Raw and processed foods were later obtained from local families and markets to determine quantities captured in the FFQs. Women from the Local Government Area were hired to demonstrate the processing of various foods in a controlled setting. Replicate cooking trials using similar ingredients and recipes were conducted in the US to quantify raw, dry, and wet weight serving quantities.

Reported quantities and frequencies were combined to determine the volume, serving weight, and dry weight of daily intake for each food type by child. Estimated reported daily caloric intake was calculated by multiplying caloric content obtained from literature sources by the appropriate serving or dry weight. Total reported daily intakes were then calculated by summing the totals for all food types. For breastfeeding children (n=4), mothers were unable to estimate total quantity of milk consumed/day. WHO estimates for caloric intake from breast milk in low-income countries were substituted by age group for these children (WHO, 1998). The total caloric intake results are likely biased high, as frequently occurs in self-reporting (Chemaly et al., 2004; MacIntyre et al., 2001; Shahar et al., 2003; Wojtusiak et al., 2011). Examination of these results suggested that the over-estimates are largely due to redundant reporting of the frequency of servings over the year. For example, parents would report a child consumes guinea corn and yams 5 times/week, when frequencies were actually seasonal, resulting in an over-estimate. To adjust for this redundancy, these estimates were normalized by determining a representative single composite serving for each child reflecting the relative proportion (volume and content) of staple foods (i.e. maize, guinea corn, millet, rice, legumes, yams/sweet potato/potato/cassava, and ground nut) (Haard et al., 1999; Onofiok and Nnanyelugo, 1998). This composite serving was multiplied by 2.5 servings per day to calculate an estimated daily intake for these staple food types. The resultant geometric mean total caloric intake for all children exclusive of milk products was 804 kcal/day corresponding to a detailed caloric intake study conducted in nearby Zaria State, Nigeria which found 810 kcal/day for young children, exclusive of milk products (Oranusi et al., 2007). Adjusting to 5 servings per day resulted in a geometric mean of 1251 kcal/day, which corresponds to the Food and Agriculture Organization (FAO) recommended daily caloric intake of 1208 kcal/day (FAO, 2001). Normalized results preserved the relative proportion of staple food components in the child's diet reported by the parents and utilizing both adjustments (i.e. 2.5 and 5 servings/day) provided a range of total caloric intake.

### 3.4.4 Lead Intake and Uptake Estimates

Daily dietary lead intake was estimated by combining the typical diet with the average metals concentrations for each food item. Dry weights (g/day) (corresponding to the adjusted caloric intake, kcal/day) were multiplied by lead concentration (µg/g) to determine lead intake lead (µg/day). Lead uptake (µg/day) was estimated by multiplying lead intake by absolute bioavailability (%). As the lead contamination was predominantly adhered or comingled mineral ore dust particulate (Plumlee et al., 2013), ranges of potential lead uptake were determined. The low estimate assumed 30% absolute bioavailability reflecting soil and ore test results, and 50% was used as the recommended absorption rate for lead in food in the USEPA Integrated Exposure Uptake Biokinetic (IEUBK) model for lead (USEPA, 2007b). Possible contributions to BLLs (µg/dl) for the 5 age groups were obtained by multiplying the uptake estimate (µg/day) by the age-specific Harley-Kneip biokinetic coefficient (unitless) for conversion to blood lead concentrations used in the IEUBK (Harley and Kneip, 1985; USEPA, 2007b; von Lindern et al., 2003; 2016).

## 3.5. Results

### 3.5.1 Contemporaneous Environmental Media Contaminant Concentrations

Table 1 summarizes pre-remediation soil, water, and blood lead levels for Bagega village contemporaneous to food sampling. Soil lead contamination results for other villages and time periods can be found in Bartrem et al., 2017, Tirima et al., 2016, and von Lindern et al., 2011.

### 3.5.2 Food Contamination Levels

Table 2 shows dry-weight lead concentration results from ICP-AES analyses for all food samples, grouped by food type and stage of post-harvest processing. Extensive contamination was noted in both farm and market products, with only three of 34 samples showing levels below the 0.05 µg/g detection limit. Excluding an extremely contaminated sample of baobab leaves, lead concentrations ranged from 0.09 to 3.41 µg/g with a mean of 0.66 µg/g. Levels exceeding 1.0 mg/kg lead were observed in several dried/processed market goods. Foods that had been pulverized showed significantly higher lead content, two to four times greater than raw foods obtained post-harvest or from markets.

Some results from food sample analysis were not included in the total dietary lead estimate. A sample of wheat from the state capitol was excluded because most families purchase breads from village bakeries. Local seed-cakes (*dadawa*) were excluded due to difficulties in determining the quantities used in soups. Similarly, the intakes of baobab leaves, ginger, dried chilies, okra, tamarind, and traditional medicines were difficult to quantify and these foods did not make up a significant portion of reported dietary intake.

### 3.5.3 Dietary Composition

Supplemental Table S1 (Appendix E) summarizes all food types and preparations reported by villagers in the overall food survey. Children's diets show a variety of foodstuffs, largely consisting of cereal grains, soybean, peanuts, sweet potato and yams, and lesser amounts of milk products, bread, and various vegetables. Infrequent inclusion of meat, foraged foods, and pasta were identified. Table 3 summarizes i) self-reported dietary components and caloric intake from the FFQ; ii) rates adjusted to 2.5 and 5 servings/day; and iii) corresponding Zaria and FAO caloric intakes. Table 4 summarizes lead intake and associated blood lead increments for each age group based on the two adjusted intake scenarios. Total reported geometric mean caloric intakes ranged from 927 kcal/day (ages 12-23 months) to 5419 kcal/day (60-71 months), with a geometric mean of 2733 kcal/day for all age children. The low range geometric mean caloric estimates adjusted to the Zaria study showed 493 kcal/day (ages 12-23 months) to 2153 kcal/day (60-71 months) with a geometric mean of 1118 kcal/day for all ages, including milk. Similar results adjusted to FAO recommended intakes showed 612-3104 kcal/day with a geometric mean of 1582 kcal/day, including milk. Breast/cow/goat milk accounted for 18-23% of total adjusted calories at 2.5 and 5 servings/day, respectively. Cereal grains supplied 25-34%, or 359 kcal/day and 718 kcal/day for the 2.5 and 5 servings/day, respectively (data not shown). In comparison, the Zaria study showed 74% of caloric intake from carbohydrates (Oranusi et al., 2007). The youngest children's caloric intakes are dependent on milk products (252 total kcal/day, 57-49% milk at 2.5 and 5 servings/day, respectively). Total caloric intakes are below FAO recommendations for the youngest 2 age groups as children's diets are transitioning to solid foods. Intakes then increase with the eldest 3 age groups as solid foods (rice, yams/sweet potato/cassava, beans, and ground nuts) are regularly consumed.

### 3.5.4 Lead Intake and Absorption

Estimated daily lead intake was derived by multiplying dry intake (g) by lead concentration (µg/g) for each staple dietary component (FAO, 2001; Llobet et al., 2003). Adjusted total lead intake for all ages averaged 48.79 µg/day to 91.59 µg/day for the Zaria and FAO adjusted estimates, respectively (Table 4). Mean lead intake varied by age, with average intake for the Zaria-adjusted group increasing from 6.73 µg/day for 12-24-month-old children to 78.18 µg/day for the 60-71-month-old children. Comparable daily lead intake for the FAO adjusted estimates were about 51% higher for older children, but were similar for the youngest group dependent on milk. Estimated blood lead increments also increased by age, depending on the combination of intake, bioavailability, and the age-specific HK coefficient. The youngest group showed relatively low increments 1.36 µg/dl to 2.5 µg/dl for the 50% absorption rate for the Zaria and FAO adjusted caloric intakes, respectively. However, this assumes no lead contribution from breast milk; which is unlikely, as adults in the villages were observed to have high BLLs (Wurr and Cooney, 2014) and experienced similar pre-remediation exposures as children. Blood lead increments increase by 81% in 24-35-month-old children (12.7 µg/dl to 66.1 µg/dl) with the transition to cereal, and then peak at 60-71 months (13.49 µg/dl to 26.33 µg/dl, respectively) when the percentage of cereal in the diet and total caloric intakes are highest.

### 3.6. Discussion

The interrelationship between ore processing and food and dietary practices in the Zamfara villages is complex and affects environmental exposure pathways, lead intake and uptake rates, and subsequent toxic effects of lead and other metals. The influence of physiological pre-disposition and environmental, cultural, behavioral, and socio-economic risk factors on lead ingestion and absorption rates are relatively well understood in the US, Western Europe, and Australia (USEPA, 2013). However, neither the etiology nor the significance of these factors in settings such as northern Nigeria have been investigated.

### 3.6.1 Sources

Most food contamination occurred during post-harvest during threshing, processing, and preparation on contaminated soils within the compounds. Plant uptake of metals was likely minimal, as field observations noted that relatively small portions of cropland were contaminated and that little ore processing took place near the fields (Abdu and Yusuf, 2013; Plumlee et al., 2013). Several studies suggest that uptake of inorganic lead by commonly grown garden plants is relatively low, and that human ingestion of adhered contaminated dust is of greater importance (Intawongse and Dean, 2006; Lee et al., 2013; Roy and McDonald, 2015). Additionally, USGS chemical and electron microscopy analyses of dusts adhering to both raw and processed food samples from the Zamfara villages revealed the same metallic species composition as the ores, village soils, and interior home sweep sample. The same analyses suggest that processed foods, especially those pulverized in the residential compound kitchens, showed higher lead levels than raw or market foods (Plumlee et al., 2013).

**3.6.2 Intake**

Total caloric intake normalized to 2.5 servings/day of staple foods were consistent with observed levels in studies in nearby Zaria State. Normalizing to 5 servings/day agreed with FAO recommendations for adequate nutritional requirements. It is likely the former applies to the majority of Zamfara villagers, as malnutrition is common among children in the region (Alabi et al., 2016; UNICEF, 2016). Reported caloric intakes, ranging from moderate malnutrition to unlikely high levels among some families, likely reflect relative affluence, individual preferences, or over-reporting. Most of the suspected dietary lead intake was attributed to staple foods made from maize, guinea corn, millet, and local rice prepared in home compounds. Mineralized dust adhering to raw food or post-harvest processing contamination resulted in mean lead intakes of 48.79 µg/day (range 6.73 µg/day to 78.18 µg/day) in the Zaria adjusted diet. More affluent families could expect 53% higher lead intakes consistent with the FAO adjusted diet. For comparison, the WHO drinking water guidance for lead is 10 µg/l, based on 3.5 µg/day/kg body weight for a 5-kg infant or about 18 µg/day (WHO, 2008). In 2010, WHO acknowledged these criteria were insufficient to protect against adverse neurological effects (WHO, 2010). Overall lead intakes are likely higher than those reported here as these calculations exclude the contribution of breast milk and certain local foods that were difficult to quantify and contaminated water that averaged 40 µg/l in private wells (Table 1).

**3.6.3 Absorption**

Lead speciation results indicate the soil/dust particles in food samples were rich in lead carbonates and lead oxides (Mushak, 2011; Plumlee et al., 2013; Cornelis et al., 2005). *In vitro* bioaccessability tests on 12 ore, soil, and dust samples from the villages ranged from 6 % to 66%, averaging 54%

(equivalent to 27% absolute bioavailability) (Plumlee et al., 2013). Eleven additional soil and ore samples analyzed by USEPA Method 9200B showed *in vitro* absolute bioavailability ranging from 17% to 41%, averaging 33% (USEPA, 2008; unpublished data). The absorption rate for food-borne lead in the villages likely varies between 30% and 50%, the latter recommended for dietary lead in the IEUBK model (USEPA, 2007b). Lower rates might be considered, as the contaminant is mainly soil-borne particulate adhered to food. However, higher absorption rates may apply as village diets in this region are deficient in certain nutrients, including calcium, which results in increased rates of lead absorption (Mushak, 1991; Okonofua et al., 1991; Pettifor, 2004; USEPA, 2013). Deficiencies in other nutrients, fasting between meals, and low caloric intakes during seasonal declines in food availabilities also may contribute to increased gastrointestinal absorption rates (Ahamed et al., 2007; Gallicchio et al., 2002; James et al., 1985; Liu et al., 2011; Rabinowitz et al., 1980). As a result, a range of potential mean blood lead increments from adulterated food were calculated at 30% and 50% bioavailability. Resultant BLLs are age specific, dependent on both physiological, behavioral, and dietary factors. Lead intake is determined both by the quantity and composition of food in the diet. As the child transitions from nursing to cereal to solid food, the contribution of the most contaminated cereal grains increases with total intake as the child grows. This has the unfortunate effect of causing significant increases during the vulnerable developmental stage for adverse neuro-cognitive effects (ATSDR, 2007; Cecil, 2010; Miranda et al., 2007). Food samples were collected during the second phase of remediation in 2010-2011, when pre-remedial geometric mean BLLs were 47 μg/dl to 76 μg/dl across all villages and averaged 87 μg/dl for the eight Bagega children referred to clinics in other villages. At that time, adulterated food potentially contributed as much as 11% to 34% of absorbed blood lead, drinking water likely contributed similar amounts for individuals using private wells, with the largest contribution due to incidental ingestion of contaminated soils and dusts (Tirima et al., 2016; von Lindern et al., 2011).

**3.6.4 Remedies**

Banning ore processing in the villages and establishing outside mining camps (*dabas*) away from the settlements greatly reduced direct exposures, and provision of potable water from community boreholes reduced the drinking water exposures. Because the principal source of lead in food was contaminated soil and dust in the residential compounds and common processing areas, the soil remediation program ultimately addressed this exposure route by removing the contaminated soil and dust (Tirima et al., 2016; von Lindern et al., 2011). However, post-remediation follow-up surveys identified para-occupational lead exposures that remain a risk factor (Bartrem et al., 2017; Chan et al., 2000; Dolcourt et al., 1978; Knishkowy and Baker, 1986; Tirima et al., 2016;). Lead is brought home on worker's clothing and mining equipment. Processed ores are often stored in the compounds for security reasons. These sources likely contribute to post-remediation elevated BLLs observed by MSF via soil/dust ingestion and possibly renewed contamination of the food supply (Bartrem et al., 2017). Older children sent to the *dabas* to sell food items are exposed to contaminated dusts and can carry lead back to the home on their clothes and the food (which is often displayed in open containers). Unpurchased food is returned home for family consumption at the end of the day (Bartrem et al., 2017). Polluted water is another source of contamination, as sluicing continues in streams that feed open wells where food is washed. Some open wells tested were found to exceed WHO and Nigerian lead standards (10 µg/l), in at least one case by more than tenfold. These wells may have contributed to dietary lead prior to being rehabilitated in 2011 (UNEP/OCHA, 2010). However, most villagers used boreholes for drinking and cooking which showed lead levels below the health standard.

Food exposures likely continued until the contaminated food stores were depleted (up to 3 years). Other villages in Zamfara could have experienced lead exposures when purchasing grains from contaminated villages. The extent of these practices is unknown. The feasibility of abating the food exposure directly is doubtful in these largely subsistence farm communities; market forces undermine attempts to ban contaminated foods. Elimination of all sources of food contamination and the overall sustainability of the Zamfara remediation depends on civil and traditional governments working with local leaders to establish and maintain safer mining practices and to discourage children from participating in mineral processing (Bartrem et al., 2017).

### 3.6.5 Study Limitations

This study was not intended as an exhaustive analysis of children's diets in these communities. Food samples were obtained from a limited subset of homes in Bagega village, and from two regional markets. The sampling strategy could introduce bias into the results if these locations were not representative of typical pre-remediation households. Additionally, any samples that represent foods in the primary processing stage would likely be washed during further preparations before consumption, potentially reducing lead content if clean water was used. The exceptions are dried items such as peppers, tomatoes, and leaves, which are not washed before adding to foods and, except tomatoes, were not included in this analysis. Lead content analyses were not obtained for several food groups, i.e. roots and tubers, fresh vegetables, dairy, meat, etc. Although these crops are less likely to be contaminated, due to the differing harvest and processing procedures, these may have been adulterated during meal preparation due to the ubiquitous lead in the home. Tirima et al. (2016) noted a significant difference in mean village soil lead concentrations after one and two rainy seasons, compared to those remediated during the initial 2010 outbreak. These differences were attributed, in part, to natural attenuation of soil lead concentrations over time (Tirima et al., 2016). The food samples in this study were collected after one rainy season and may not reflect the initial exposures experienced from food contamination when soil lead levels were highest.

### 3.7 Conclusions

Rudimentary artisanal processing of ores with high lead content resulted in severe soil and dust contamination. The cultural practice of gender sequestration exacerbated exposures, resulting in geometric mean children's BLLs of 145 µg/dl and 400 deaths. Incidental hand-to-mouth ingestion of lead-contaminated soil and dust by children, dietary exposures due to food adulteration during processing and preparation, and contaminated water were the primary exposure pathways. Reductions in pre-chelation BLLs after soil remediation confirmed that removal of contaminated soils substantially decreased lead intake (Tirima et al., 2016). However, REEs showed recontamination from para-occupational sources and continuing dietary exposures. The food pathway was poorly understood at the beginning of the epidemic and specific studies were undertaken to characterize this exposure route and associated socio-economic, cultural, and demographic risk co-factors. Dietary exposure was found to be a significant source, contributing as much as 11 to 34% to BLLs prior to remediation.

These pathways were ultimately addressed by banning mining in residential areas; establishing remotely located processing areas (*dabas*), eliminating the residual sources through soil remediation, and encouraging locally developed and implemented safer mining practices to minimize occupational and para-occupational exposures. The long-term sustainability of efforts to keep exposures low depends on local civil, traditional, religious, and informal leadership's involvement and support.

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Table 3.1. Summary pre-remediation soil, water, and blood lead levels in Bagega village. These data are contemporaneous to food samples collected prior to remediation. ([1]Pre-remediation February 2011 data [Tirima et al., 2016]; [2]water summary statistics from September 2010 are for detects only [UNEP/OCHA, 2010]; [3]August 2010 screening [MSF 2010]).

| | | non-detects | detect values | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | n | n | min | max | mean | stdv | gmn | gstdv |
| ***in situ* residential soils** | (mg/kg)[1] | 0 | 143 | 48 | 12691 | 1452 | 2287 | 636 | 3.5 |
| **communal handpump** | (μg/l)[2] | 1 | 0 | . | . | . | . | . | . |
| **communal well** | | 8 | 4 | 13 | 56 | 35 | 16 | 30 | 1.7 |
| **private well** | | 17 | 8 | 10 | 130 | 40 | 47 | 22 | 2.7 |
| ***all water sources*** | | *26* | *12* | *10* | *130* | *38* | *40* | *24* | *2.4* |
| **blood lead levels** | (μg/dl)[3] | 0 | 8 | 49 | 135 | 87 | 34 | 80 | 1.5 |

Table 3.2. Matrix of lead contamination levels (μg/g) in local foodstuff (ICP-AES results – dry weight) by food type and processing stage. Cells in gray were not included in dietary Pb intake analyses. ([1]Excluding baobab leaves).

| Food Type | Source | Notes | Harvested, Thrashed, Stored | Processed (ready to cook or eat) | Dried vegetables, herbs, spices | Traditional medicines and herbs | Mean (stdev) |
|---|---|---|---|---|---|---|---|
| guinea corn (sorghum) | Bagega village | whole grain | 0.93 | | | | 0.86 (077) |
| | Bagega farm | whole grain | 0.41 | | | | |
| | Anka market | whole grain | <0.05 | | | | |
| | Bagega village | dried, milled | | 2.06 | | | |
| millet | Bagega farm | whole grain | <0.05 | | | | 0.41 (0.27) |
| | Anka market | whole grain | 0.53 | | | | |
| | Bagega village | ground, ready to cook | | 0.66 | | | |
| maize (corn) | Bagega farm | whole grain, dried kernels | 0.27 | | | | 0.20 (0.08) |
| | Anka market | whole grain, dried kernels | 0.12 | | | | |
| local rice | Bagega market | whole grain | 0.73 | | | | 0.30 (0.26) |
| | Bagega farm | hulled | <0.05 | | | | |
| | Bagega farm | whole grain, with hulls | 0.2 | | | | |
| | Anka market | whole grain, with hulls | 0.44 | | | | |
| white rice | Anka market | whole grain | 0.09 | | | | |
| bread (white) | Bagega bakery | dried 3 days, pulverized | | 0.31 | | | 0.52 (0.29) |
| | Bagega bakery | dried 3 days, pulverized | | 0.92 | | | |
| wheat flour | Gusau market | sifted wheat flour | | 0.32 | | | 0.32 |
| cowpea | Bagega village | whole | 0.24 | | | | 0.27 (0.12) |
| | Bagega farm | whole | 0.39 | | | | |
| | Anka market | whole | 0.36 | | | | |
| tapery bean | Anka market | whole | 0.081 | | | | |
| *dadawa* | Bagega market | boiled, pounded, dried | | 3.41 | | | 1.93 (1.49) |
| | Anka market | boiled, pounded, dried | | 0.44 | | | |
| peanuts | Bagega village | pounded, ready to eat | | 0.92 | | | 0.51 (0.41) |
| | Anka market | paste ready to eat | | 0.1 | | | |
| baobab leaves | Bagega market | dried, ready to cook | | | 146 | | 146 |
| tomatoes | Bagega market | dried, ready to cook | | | 0.3 | | 0.3 |
| ginger root | Anka market | dried, ready to cook | | | 1.86 | | 1.86 |
| chilies | Anka market | dried, pulverized | | | 0.62 | | 0.62 |
| okra | Anka market | dried, ready to cook | | | 0.18 | | 0.18 |
| tamarind | Anka market | pods | | | 0.48 | | 0.48 |
| medicine | Bagega market | local medicine | | | | 0.69 | 1.40 (0.93) |
| | Bagega market | local medicine | | | | 0.79 | |
| | Bagega market | local medicine | | | | 2.72 | |
| | | **overall means (stdev)** | 0.32 (0.29) | 0.85 (0.64) | 0.69 (0.68) | 0.74 (0.93) | 0.66 (0.77)[1] |

Table 3.3. Reported and adjusted food and caloric intakes, by age. Two different intake adjustment scenarios are presented, along with results from corresponding Zaria and FAO studies. Geometric mean values for all ages are bolded for comparison to Zaria and FAO results ([1]Oranusi et al., 2007; [2]FAO, 2001).

| **Age (months)** | | | **Reported Intakes (g/da)** | **Reported Calories (kcal/day)** | **Intakes Adjusted to 2.5 servings/day (g/day)** | **Intakes Adjusted to 5 servings/day (g/day)** | **Calories Adjusted to 2.5 servings/day (kcal/day)** | | **Calories Adjusted to 5 servings/day (kcal/day)** | | **Zaria, Nigeria Intakes (kcal/day)[1]** | **FAO Recommended Intakes (kcal/day)[2]** | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | | with milk | without milk | with milk | without milk | | males | females |
| 12-23 | n=3 | min | 571 | 419 | 1 | 2 | 434 | 72 | 482 | 120 | | | |
| | | max | 1248 | 4531 | 45 | 90 | 617 | 572 | 892 | 847 | | | |
| | | avg | 797 | 1799 | 21 | 41 | 500 | 247 | 636 | 384 | | 948 | 865 |
| | | std | 390 | 2366 | 22 | 45 | 102 | 282 | 223 | 403 | | | |
| | | gmn | 742 | 947 | 8 | 17 | 493 | 159 | 612 | 266 | | | |
| | | gsd | 8.0 | 3.9 | 8.0 | 8.1 | 1.2 | 3.1 | 1.4 | 2.8 | | | |
| 24-35 | n=6 | min | 239 | 612 | 19 | 38 | 362 | 237 | 534 | 358 | | | |
| | | max | 2122 | 4174 | 223 | 447 | 1210 | 1141 | 2049 | 1995 | | | |
| | | avg | 1175 | 2300 | 73 | 146 | 851 | 664 | 1205 | 1018 | | 1129 | 1047 |
| | | std | 725 | 1517 | 76 | 152 | 364 | 361 | 561 | 599 | | | |
| | | gmn | 938 | 1821 | 52 | 104 | 777 | 573 | 1091 | 870 | | | |
| | | gsd | 2.4 | 2.2 | 2.4 | 2.4 | 1.6 | 1.9 | 1.7 | 1.9 | | | |
| 36-47 | n=9 | min | 386 | 876 | 48 | 95 | 400 | 371 | 594 | 565 | | | |
| | | max | 3419 | 7145 | 241 | 481 | 2471 | 2073 | 3173 | 2775 | | | |
| | | avg | 1686 | 3772 | 111 | 223 | 1308 | 1135 | 1884 | 1711 | | 1252 | 1156 |
| | | std | 909 | 2016 | 61 | 122 | 608 | 545 | 817 | 754 | | | |
| | | gmn | 1445 | 3238 | 98 | 197 | 1169 | 1004 | 1698 | 1536 | | | |
| | | gsd | 1.7 | 1.9 | 1.7 | 1.7 | 1.7 | 1.7 | 1.7 | 1.7 | | | |
| 48-59 | n=5 | min | 438 | 943 | 41 | 82 | 462 | 424 | 719 | 681 | | | |
| | | max | 5348 | 7952 | 189 | 377 | 3089 | 1870 | 3710 | 2490 | | | |
| | | avg | 2306 | 3506 | 121 | 242 | 1467 | 1100 | 2013 | 1646 | | 1360 | 1241 |
| | | std | 2232 | 2772 | 73 | 146 | 1032 | 574 | 1191 | 769 | | | |
| | | gmn | 1469 | 2724 | 99 | 198 | 1189 | 967 | 1711 | 1476 | | | |
| | | gsd | 2.1 | 2.2 | 2.1 | 2.1 | 2.1 | 1.8 | 1.9 | 1.7 | | | |
| 60-71 | n=6 | min | 1454 | 2617 | 70 | 140 | 1286 | 631 | 1755 | 976 | | | |
| | | max | 5982 | 11192 | 485 | 971 | 4706 | 4191 | 6078 | 5563 | | | |
| | | avg | 3414 | 6378 | 231 | 462 | 2432 | 1857 | 3466 | 2891 | | 1467 | 1330 |
| | | std | 1857 | 3738 | 161 | 322 | 1381 | 1263 | 1802 | 1651 | | | |
| | | gmn | 2981 | 5419 | 184 | 368 | 2153 | 1557 | 3104 | 2496 | | | |

| Age (months) | | | Reported Intakes (g/da) | Reported Calories (kcal/day) | Intakes Adjusted to 2.5 servings/day (g/day) | Intakes Adjusted to 5 servings/day (g/day) | Calories Adjusted to 2.5 servings/day (kcal/day) | | Calories Adjusted to 5 servings/day (kcal/day) | | Zaria, Nigeria Intakes (kcal/day)[1] | FAO Recommended Intakes (kcal/day)[2] | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | | with milk | without milk | with milk | without milk | | males | females |
| | | gsd | 2.1 | 1.9 | 2.1 | 2.1 | 1.7 | 1.9 | 1.7 | 1.8 | | | |
| all ages | n=29 | min | 239 | 419 | 1 | 2 | 362 | 72 | 482 | 120 | 582 | 865 | |
| | | max | 5982 | 11192 | 485 | 971 | 4706 | 4191 | 6078 | 5563 | 1089 | 1467 | |
| | | avg | 1953 | 3757 | 121 | 241 | 1390 | 1089 | 1964 | 1663 | **810** | 1221 | |
| | | std | 1536 | 2764 | 106 | 216 | 985 | 815 | 1353 | 1182 | not reported | 170 | |
| | | gmn | 1437 | 2733 | 76 | 152 | 1118 | **804** | 1582 | **1251** | not reported | **1208** | |
| | | gsd | 3.3 | 2.4 | 3.3 | 3.4 | 2.0 | 2.4 | 2.0 | 2.4 | not reported | 1.2 | |

Table 3.4. Lead intake, uptake, and resulting BLLs based on the two intake scenarios. HK = Harley Kneip coefficient for lead based on age; abs. = absorption; BLL = blood lead level.

| **age (months), HK, n** | | **Lead intake from foods based on 2.5 servings/day (g/day)** | | | | | | | | | **BLL (µg/dl)** | | **Lead intake from foods based on 5 servings/day (g/day)** | | | | | | | | | **BLL (µg/dl)** | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | maize | g. corn | millet | local rice | white rice | legume | bread | tomato | total | 30% abs | 50% abs | maize | g. corn | millet | local rice | white rice | legume | bread | tomato | total | 30% abs | 50% abs |
| 12-23, 0.404, 3 | min | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.0 | 0.0 | 0.3 | 0.0 | 0.1 | 0.2 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.0 | 0.0 | 0.4 | 0.1 | 0.1 |
| | max | 0.4 | 6.6 | 0.2 | 2.5 | 2.3 | 3.3 | 1.5 | 0.2 | 10.1 | 1.2 | 2.0 | 0.8 | 13.1 | 0.3 | 5.0 | 4.7 | 6.6 | 1.5 | 0.2 | 19.2 | 2.3 | 3.9 |
| | **avg** | **0.2** | **2.4** | **0.1** | **1.4** | **0.8** | **1.2** | **0.6** | **0.1** | **6.7** | **0.8** | **1.4** | **0.4** | **4.8** | **0.1** | **2.7** | **1.6** | **2.4** | **0.6** | **0.1** | **12.7** | **1.5** | **2.6** |
| | std | 0.2 | 3.6 | 0.1 | 1.3 | 1.3 | 1.8 | 0.8 | 0.1 | 5.6 | 0.7 | 1.1 | 0.3 | 7.2 | 0.2 | 2.5 | 2.7 | 3.7 | 0.8 | 0.1 | 10.7 | 1.3 | 2.2 |
| 24-25, 0.366, 6 | min | 0.9 | 2.1 | 0.0 | 1.9 | 0.0 | 0.4 | 0.3 | 0.1 | 7.0 | 0.8 | 1.3 | 1.8 | 4.2 | 0.0 | 3.7 | 0.0 | 0.7 | 0.3 | 0.1 | 12.8 | 1.4 | 2.3 |
| | max | 15.2 | 85.9 | 0.3 | 12.0 | 2.4 | 1.3 | 7.2 | 0.8 | 115.5 | 12.7 | 21.1 | 30.3 | 171.7 | 0.6 | 24.0 | 4.9 | 2.5 | 7.2 | 0.8 | 230.4 | 25.3 | 42.2 |
| | **avg** | **4.1** | **19.7** | **0.1** | **6.2** | **0.7** | **1.0** | **2.4** | **0.3** | **34.4** | **3.8** | **6.3** | **8.1** | **39.3** | **0.2** | **12.4** | **1.4** | **2.0** | **2.4** | **0.3** | **66.1** | **7.3** | **12.1** |
| | std | 5.5 | 32.6 | 0.1 | 4.6 | 0.9 | 0.3 | 2.6 | 0.3 | 40.6 | 4.5 | 7.4 | 11.0 | 65.3 | 0.2 | 9.2 | 1.7 | 0.7 | 2.6 | 0.3 | 81.8 | 9.0 | 15.0 |
| 36-47, 0.350, 9 | min | 0.3 | 3.4 | 0.0 | 2.9 | 0.0 | 0.6 | 0.0 | 0.0 | 13.1 | 1.4 | 2.3 | 0.5 | 6.7 | 0.0 | 5.8 | 0.0 | 1.2 | 0.0 | 0.0 | 26.1 | 2.7 | 4.6 |
| | max | 33.0 | 59.8 | 1.7 | 12.1 | 2.9 | 4.9 | 50.8 | 5.9 | 105.7 | 11.1 | 18.5 | 65.9 | 119.5 | 3.4 | 24.1 | 5.9 | 9.9 | 50.8 | 5.9 | 164.7 | 17.3 | 28.8 |
| | **avg** | **6.7** | **28.5** | **0.5** | **7.1** | **1.3** | **2.4** | **10.6** | **0.8** | **57.8** | **6.1** | **10.1** | **13.3** | **57.0** | **1.0** | **14.2** | **2.5** | **4.8** | **10.6** | **0.8** | **104.2** | **10.9** | **18.2** |
| | std | 10.1 | 17.7 | 0.7 | 3.6 | 1.1 | 1.5 | 16.8 | 1.9 | 31.6 | 3.3 | 5.5 | 20.2 | 35.3 | 1.4 | 7.1 | 2.2 | 3.0 | 16.8 | 1.9 | 54.2 | 5.7 | 9.5 |
| 48-59, 0.363, 5 | min | 0.8 | 1.5 | 0.0 | 4.2 | 0.5 | 1.2 | 0.0 | 0.2 | 12.5 | 1.4 | 2.3 | 1.7 | 3.0 | 0.1 | 8.4 | 1.0 | 2.4 | 0.0 | 0.2 | 23.2 | 2.5 | 4.2 |
| | max | 19.3 | 13.7 | 0.5 | 26.7 | 6.2 | 7.0 | 16.0 | 8.0 | 60.8 | 6.6 | 11.0 | 38.6 | 27.4 | 1.0 | 53.4 | 12.3 | 14.1 | 16.0 | 8.0 | 105.5 | 11.5 | 19.1 |
| | **avg** | **5.9** | **7.8** | **0.2** | **14.5** | **2.8** | **2.7** | **4.2** | **1.8** | **39.8** | **4.3** | **7.2** | **11.8** | **15.6** | **0.4** | **29.0** | **5.5** | **5.4** | **4.2** | **1.8** | **73.7** | **8.0** | **13.4** |
| | std | 7.6 | 4.6 | 0.2 | 11.1 | 2.4 | 2.5 | 6.7 | 3.4 | 21.8 | 2.4 | 4.0 | 15.2 | 9.2 | 0.4 | 22.2 | 4.7 | 5.0 | 6.7 | 3.4 | 40.0 | 4.4 | 7.3 |
| 60-71, 0.345, 6 | min | 5.5 | 10.2 | 0.3 | 2.1 | 0.8 | 1.6 | 0.2 | 0.0 | 25.0 | 2.6 | 4.3 | 11.0 | 20.4 | 0.6 | 4.1 | 1.6 | 3.3 | 0.2 | 0.0 | 48.5 | 5.0 | 8.4 |
| | max | 59.1 | 93.9 | 7.4 | 51.0 | 11.1 | 6.6 | 8.7 | 6.4 | 175.2 | 18.1 | 30.2 | 118.2 | 187.9 | 14.9 | 101.9 | 22.2 | 13.2 | 8.7 | 6.4 | 349.6 | 36.2 | 60.3 |
| | **avg** | **19.9** | **31.1** | **2.2** | **14.8** | **2.9** | **3.5** | **2.6** | **1.2** | **78.2** | **8.1** | **13.5** | **39.8** | **62.2** | **4.4** | **29.6** | **5.9** | **7.1** | **2.6** | **1.2** | **152.6** | **15.8** | **26.3** |
| | std | 20.4 | 31.6 | 2.7 | 18.9 | 4.1 | 2.2 | 3.2 | 2.5 | 55.2 | 5.7 | 9.5 | 40.8 | 63.2 | 5.3 | 37.8 | 8.1 | 4.4 | 3.2 | 2.5 | 110.8 | 11.5 | 19.1 |
| all ages | min | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.0 | 0.0 | 0.3 | 0.0 | 0.1 | 0.2 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.0 | 0.0 | 0.4 | 0.1 | 0.1 |
| | max | 59.1 | 93.9 | 7.4 | 51.0 | 11.1 | 7.0 | 50.8 | 8.0 | 175.2 | 18.1 | 30.2 | 118.2 | 187.9 | 14.9 | 101.9 | 22.2 | 14.1 | 50.8 | 8.0 | 349.6 | 36.2 | 60.3 |
| | **avg** | **8.1** | **20.9** | **0.7** | **9.2** | **1.7** | **2.3** | **5.1** | **0.9** | **48.8** | **5.2** | **8.6** | **16.1** | **41.9** | **1.3** | **18.4** | **3.4** | **4.5** | **5.1** | **0.9** | **91.6** | **9.7** | **16.2** |
| | std | 12.37 | 23.46 | 1.42 | 10.33 | 2.25 | 1.84 | 10.05 | 2.02 | 39.95 | 4.17 | 6.95 | 25.18 | 47.75 | 2.88 | 21.02 | 4.58 | 3.75 | 10.23 | 2.05 | 78.58 | 8.21 | 13.68 |

Figure 3.1. Lead dust contamination of food and the post-harvest system for grains in Zamfara.

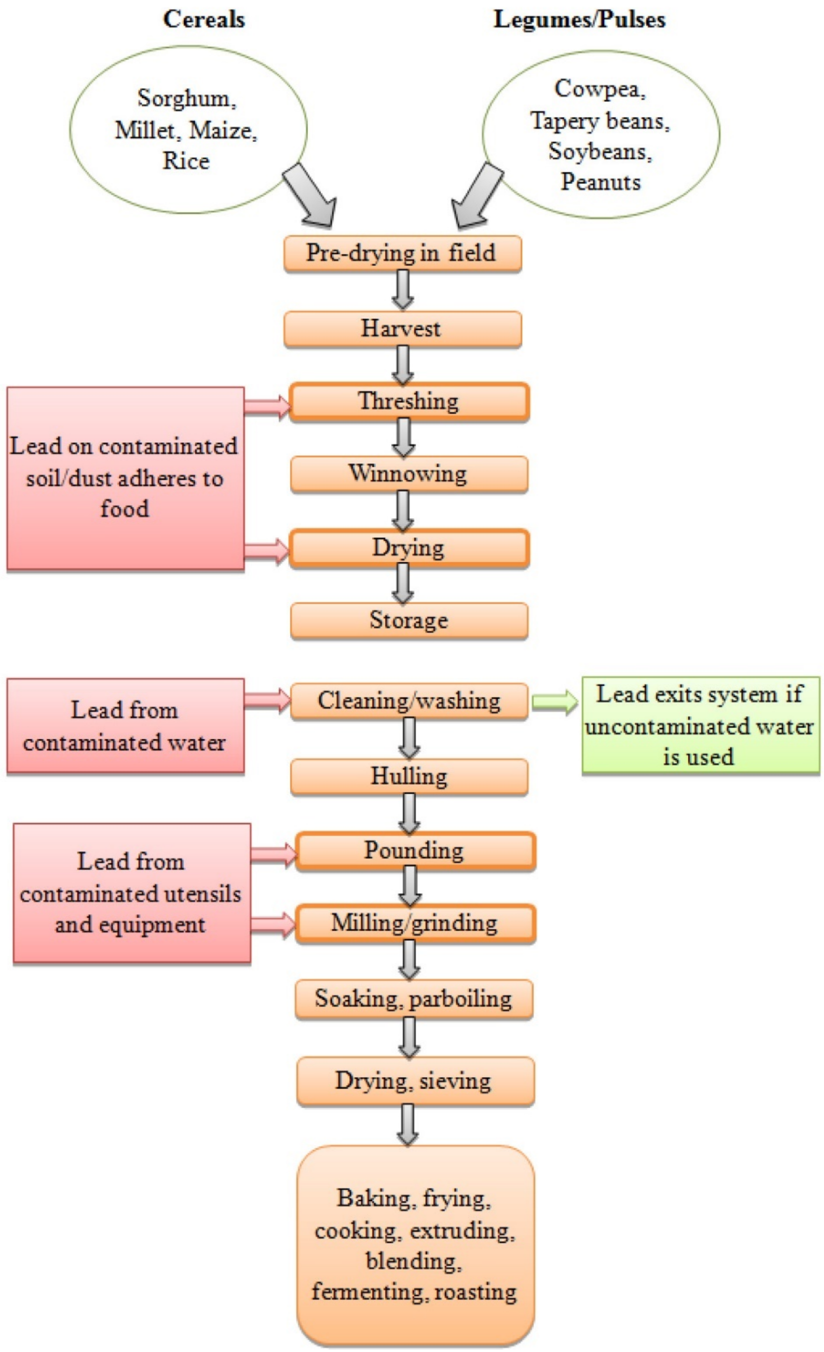


Figure 3.1 Legend. Lead dust contamination of food may occur at several stages of post-harvest food processing, including threshing, drying, pounding, and milling. The only stage where contamination of food may be reduced is during cleaning/washing, assuming uncontaminated water is used. The cleaning of foods occurs before pounding and milling and lead may be re-introduced during that stage of food preparation.

# CHAPTER 4 – CLEAN DIRT: ASSESSMENT OF SOIL REMEDIATION AND PROJECT RESILIENCE IN VILLAGES EFFECTED BY SEVERE LEAD POISONING OUTBREAK IN NIGERIA

**Abstract**

**Background:** Since 2010, federal, state, and local governments have collaborated with international health agencies and NGOs to address epidemic lead poisoning associated with artisanal gold mining in Nigeria. More than 400 children died and thousands of people have been severely poisoned. Response activities have included emergency soil remediation to reduce unprecedented exposures, chelation treatment in out-patient clinics, and advocacy programs promoting health education. Remediation involved removing contaminated soils and covering areas with “clean” backfill soils. Eight villages in Zamfara State and two villages in Niger State have been remediated over the course of six years. **Objectives:** During the environmental response, four remedial effectiveness evaluations (REEs) were carried out in Zamfara State to assess: the efficacy of remediation in reducing blood lead levels (BLLs); the degree of recontamination; the effectiveness of institutional controls in sustaining the remedy; and the technical and institutional capacity of Nigerian governments to prevent and respond to future crises. **Methods:** Four of eight remediated villages were assessed during different REE events. Two subsets of homes were evaluated through interviews and environmental sampling: i) targeted homes of children not responding positively to chelation treatment, and ii) randomly selected homes. Public areas throughout the villages were also assessed and informal discussions were held with multiple levels of leadership. Retrospective (ex-post) social impact assessment (SIA) was used to analyze interview and observation results. **Results:** Significant differences were found between pre- and post-remediation lead levels in all villages. BLLs are progressing satisfactorily in two villages while significant soil recontamination and persistently high BLLs remain in the other two villages. Six categories of SIA variables show intentional and unintentional project impacts and the influence of social factors on the long-term sustainability of the remedy. Residents in all villages are aware of methods to reduce para-occupational exposures and recontamination, yet there is little implementation of basic occupational hygiene practices. Broader efforts to prevent future lead poisoning outbreaks are lacking. **Conclusions:** The combined six-year remediation and health response substantially reduced environmental exposures and BLLs in all villages, but recontamination and childhood lead poisoning persist in two villages. While the technical capacity has grown considerably, institutional controls to prevent resumption of processing activities and low-level para-occupational exposures are needed.

## 4.1 Introduction

### 4.1.1 Environmental Health Response

A severe childhood lead poisoning epidemic was discovered in March 2010 by Médecins Sans Frontières (MSF, Doctors Without Borders) in remote villages of Zamfara State, Nigeria (Moszynski, 2010; MSF, 2010). In May 2010, a team of Nigerian, World Health Organization (WHO), and United States (US) health professionals conducted environmental and epidemiological investigations, finding that hundreds of children had died from and thousands more were at risk of severe lead poisoning (Bartrem et al., 2014; Dooyema et al., 2011; Greig et al., 2014; MSF, 2010; Plumlee et al., 2013; Thurtle et al., 2014; Tirima et al., 2016; von Lindern et al., 2011; WHO, 2011). The outbreak was attributed to artisanal processing of gold ore containing up to 10% lead. The ores were crushed, washed, and dried within residential walled compounds and public areas throughout the villages, exposing young children, pregnant women, and nursing mothers to lead in airborne dust, soil, and food prepared in these homes (Dooyema et al., 2011; von Lindern et al., 2011). The traditional Emirate leadership halted ore processing within the villages in April 2010, but residual soil and dust contamination posed a continuing health threat and preempted chelation therapy (Bartrem et al., 2014; Tirima et al., 2016; von Lindern et al., 2011). The geometric mean blood lead level (BLL) in May 2010 was 149 µg/dl compared to the US Centers for Disease Control and Prevention (CDC) reference BLL of 5 µg/dl (CDC, 2012). There is no safe level of lead in blood, with dose-dependent impacts ranging from subtle, latent neurocognitive deficits to renal impairment, motor skill losses, encephalopathy, and death (ATSDR, 2007).

In June 2010, international organizations, Nigerian health authorities, local civil and traditional governments, and village residents collaborated to remediate the villages and treat poisoned children (Tirima et al., 2016; von Lindern et al., 2011). Phase I emergency response actions were undertaken in two villages in June-July 2010. Following the rainy season, five additional Phase II villages were remediated from September 2010 – February 2011. Phase III cleanup of the largest affected village was undertaken in 2013 (von Lindern et al., 2011). Pre-remediation geometric mean *in situ* soil lead levels in homes during Phase I, II, and III were 1642 mg/kg, 442 mg/kg, and 360 mg/kg, respectively (Tirima et al., 2016). Remediation in the seven Zamfara State villages achieved an overall 89% reduction in soil lead exposures. Geometric mean pre-treatment BLLs decreased from 149 µg/dl to 15 µg/dl (Tirima et al., 2016). In 2015, MSF discovered a second lead poisoning outbreak in neighboring Niger State. Two villages with a combined population of 2500 were subsequently characterized and remediated in 2016 after funding was secured from the Nigerian Federal Government (Ukwu, 2016).

Each of these Nigerian cleanups utilized protocols and risk mitigation strategies developed at the Bunker Hill Superfund Site (BHSS) in Idaho, USA, and oversight was provided by the US organization TerraGraphics International Foundation (TIFO). The BHSS cleanup model integrates remediation, institutional controls, lead awareness, biological monitoring, and health and medical follow-up to reduce lead exposure and BLLs (USEPA, 2010, 2005, 1999). This model was adapted for northern Nigeria relying on local institutions, labor practices, technology, and equipment (NRC, 2005; Sheldrake and Stifelman, 2003; Tirima et al., 2016; von Lindern et al., 2011, 2003). Personnel from state and local governments were trained to supervise remediation, administer payrolls, procure supplies and equipment, confirm adequate contaminant removal, and track remediation data (Tirima et al., 2016). Remediation goals were to reduce exposures via excavation and replacement of contaminated soils with background (<25 mg/kg Pb) “clean” soils, facilitate MSF’s medical treatment program, and build local technical capacity for future responses (Tirima et al., 2016). Details regarding the cleanup protocols and procedures can be found in Tirima et al. (2016) and von Lindern et al. (2011).

### 4.1.2 Remedial Effectiveness Evaluation

Successfully adapting US Superfund protocols to the remote areas in Zamfara required frequent field modifications as Nigerian experience and capabilities evolved. Over the six-year period, four formal Remedial Effectiveness Evaluations (REEs) were jointly conducted by TIFO, MSF and Nigerian government personnel in 2010, 2011, 2012 and 2016. Each REE provided insight into the efficacy of the cleanups and treatment programs, the transfer of capacity and responsibility, and essential social and technical information for modifying and adapting subsequent environmental health response activities. These REEs were based on US protocols at the BHSS and National US Lead Abatement Programs, which successfully reduced children’s mean BLLs from 70 µg/dl in 1974 to 2 µg/dl in 2002. (von Lindern et al., 2016, 2003). BHSS children’s BLLs were monitored annually and follow-up in-home investigations were conducted for children whose BLL exceeded the health criteria at the time (ranging from 40 µg/dl to 10 µg/dl over the 28 years). In Zamfara, REEs also incorporated observations and interviews as part of a retrospective or ex-post social impact assessment (SIA) to evaluate intentional and unintentional consequences of the environmental health response and the influence of social factors on the efficacy of the remedy.

### 4.2. Objectives

Broadly, the goals of the REEs were to assess project success, address challenges, and improve future responses. Specific objectives were to evaluate: 1) the efficacy of remediation in reducing BLLs; 2) the degree and extent of recontamination; 3) the effectiveness of institutional controls in keeping ore processing activities out of the villages; and 4) the technical and institutional capacity of Nigerian governments to respond to and prevent future crises. This evaluation was accomplished utilizing both qualitative and quantitative data.

### 4.3. Data Sources and Methods

#### 4.3.1 Soil Sampling and Interviews

Environmental sampling data for this investigation were obtained from both the original cleanup effort (Tirima et al., 2016) and four REEs conducted from 2010 to 2016. Table 1 summarizes the remediation statistics and the status of medical treatment in the Zamfara villages. The 2010 and 2011 REEs focused on Phase I and II villages where persistently high BLLs suggested possible post-remediation increases in environmental exposures. Targeted homes were selected based on adverse blood lead trends noted by MSF and the REEs utilized BHSS follow-up protocols (USEPA, 2010, 2005, 1999). The 2012 REE was an audit of an abbreviated remediation effort in Bagega village conducted independently by Zamfara State without international oversight. Ten village homes were fully remediated and three homes were partially cleaned before the effort was curtailed due to funding issues. The assessment focused on the State's capabilities to carry out remedial activities after two years of training and capacity building during Phase I and II. Three fully remediated homes and three partially remediated homes were tested. Environmental results, project procurement, sequencing, quality control, and data management were reviewed with project personnel.

The 2016 REE was initiated in response to both persistently high BLLs in two Zamfara State villages as well as the lead poisoning outbreak in neighboring Niger State in which 28 children died (Zinggl, 2016). This REE emulated the US Superfund five-year review (FYR) protocols (USEPA, 2001). A FYR assesses if a remedy is functioning as intended, if exposure assumptions, toxicity data, and cleanup criteria are still valid, and highlights new information that could compromise the overall effectiveness of the remediation (USEPA, 2001). The 2016 REE was originally designed to both target villages with persistently high BLLs and to assess villages where the chelation treatment program was successfully completed. However, security concerns later prevented access to all but the two villages with persistent blood lead problems – Abare and Dareta. No surveys were undertaken in villages where the clinics were closed and the remedy is believed to be sustained, based on MSF's exit blood lead observations.

Homes targeted for follow-up investigation met at least one of following criteria:

- ≥15 courses of chelation treatment *and* most recent BLL ≥45 µg/dl *and* difference between first ever BLL and most recent BLL ≥10 µg/dl (see Thurtle et al. (2014) for description of treatment type, length, and other protocols)
- Any BLL ≥65 µg/dl that increased twice or more *and* didn't reduce by 20 µg/dl during treatment course
- BLL within 100-119 µg/dl after 4 courses of treatment
- MSF Health Promotion team witnessed processing inside the home within the last year

In 2016, 12% and 17% percent of all Abare and Dareta homes, respectively, were investigated based on target criteria. An additional 18% and 20% percent, respectively, were selected randomly. Informed consent for the survey was obtained from the head of household and semi-structured interviews were conducted with community leaders and parents in the local Hausa language. The interviews were designed to elicit dialog reconstructing a day, a week, and a month in the child's life to explore possible lead exposure sources and routes. Discussions included diet, hygiene habits, where meals were prepared and consumed, sleep and play areas within the home, and locations children visited on a regular or occasional basis, including village public areas and processing sites (*dabas*). Children were observed by one member of the interview team during the assessment and sometimes followed outside to observe play habits, activities, and locations visited. Parents were asked about employment in the mining industry and about implementation of basic occupational hygiene. Interviewers also asked about the use of galena as a cosmetic in the home and if the household was involved in any trade utilizing or recycling lead. The source and storage of the household food (mostly grains) were evaluated and samples were collected for later testing at the discretion of the interviewer. Results of food sampling can be found in Plumlee et al. (2013) and Tirima et al. (2017). Field notes were reviewed post-interview and summary forms and checklists developed to allow for ex-post SIA analysis of results (Vanclay, 2012, 2002). These results were used to assess possible lead exposure sources and routes, and to determine social impacts of the project and social influences on project success.

Environmental sampling was performed using portable x-ray fluorescent spectrometer (PXRF) to determine *in situ* surface soil heavy metal concentrations. Several models of PXRF were used during remediation and REE sampling, including InnovX (Olympus) Alpha 4000 and Delta 4000 and Niton (Thermo Fisher) XL3. Sampling protocol followed characterization and post-remediation sampling used during the cleanup efforts (Tirima et al., 2016). REE-specific PXRF testing was accomplished in children's high-use areas, village public areas, locations where mining tools were observed, and areas where ore processing is more likely to occur, including under shade trees or near water sources. Information regarding the resumption, if any, of processing activities at these locations was obtained from community members who helped to identify public areas for characterization. Additionally, randomly selected areas in homes and public areas were tested at the samplers' discretion.

During the 2016 REE, soil samples were collected for calculating the conversion from *in situ* PXRF lead concentration to *ex situ* sieved ICPMS concentration. E*x situ* bulk sample aliquots were collected directly from the top 2 cm of random *in situ* PXRF test at sub-locations at 52 sites. These samples were homogenized in plastic sample packs and three PXRF readings were obtained through the bags. The samples were then submitted to a laboratory, sieved to minus 80 standard mesh (179 µm) and analyzed by inductively coupled plasma mass spectrometry (ICP-MS) for Pb, As, Mn, Cd, and Hg.

**4.3.2 Data Analysis**

PXRF logs from Phase I, II, III, and the REEs were downloaded daily and categorized by village, home compound, or public area. During remediation, maps were prepared for each location designating sub-areas where *in situ* soil lead concentrations exceeded the excavation and/or clean soil cover criteria. Soils >1000 mg/kg were removed until PXRF testing showed lead levels <400 mg/kg. Both excavated locations and areas with concentrations 400-1000 mg/kg were covered with 5-8 cm of "clean" backfill soil (Tirima et al., 2016). A nominal backfill lead concentration of 25 mg/kg was used in all analyses, but generally values were non-detect (ND) with a PXRF detection limit of ~12 mg/kg. PXRF lead concentration logs were available for both pre- and post-excavation surface soils. The pre-remediation results were potentially biased toward values near the remedy determination levels (400 mg/kg and 1000 mg/kg) because these areas were often more intensively tested to identify boundaries and avoid false negative results (Type II error). To assess this possible bias, representative pre-remediation *in situ* soil exposure indices were developed by estimating area weighted average (AWA) soil lead concentrations and comparing the results to the PXRF records. Remediation maps for 40 randomly selected home compounds were spatially divided into three pre-remedial categories reflecting the action criteria (i.e., ND-399 mg/kg, 400-999 mg/kg, and >1000 mg/kg). Areal extent and average soil lead concentrations from all PXRF readings in each sub-area were determined allowing calculation of the pre-remediation *in situ* AWA soil lead concentration for each compound. Post-excavation (i.e., pre-clean soil backfill) AWA soil lead concentrations were calculated by averaging *in situ* results in excavated sub-areas and retaining the un-remediated values for the ND-399mg/kg category. Post-remediation (i.e., post-clean soil backfill) AWA soil concentrations were determined by substituting clean soil values (25 mg/kg Pb) for all remove/replace and cover locations. These AWAs were then compared to various central-tendency statistics from *in situ* data to identify the most-representative exposure metric. The best pre-and post-remediation measurement was selected and used as an *in situ* exposure index in all compounds tested and or/ remediated in all Phases and REEs. Village, compound and public area means and distribution statistics were generated by SAS® 9.4 software. Multiple comparisons of soil exposure metrics for both targeted and randomly selected homes were accomplished by analysis of variance (ANOVA) techniques. For *ex situ* soil samples collected in 2016, the *in situ*, *ex situ* bulk, and ICP-MS sieved results were then compared by linear regression to establish an empirical correlation coefficient to estimate minus 80-mesh soil concentrations from *in situ* results.

Questionnaires and notes from semi-structured interviews in 2011 and 2016 were analyzed using a SIA framework from Vanclay (2002). While SIA is typically used prior to planned interventions, specific aspects of the framework can be adapted for retrospective analysis (Li et al., 2014; Vanclay, 2012). Broadly, SIA is a tool for analyzing, monitoring, and managing both intentional and unintentional impacts of interventions. One of its primary goals is to elicit a more sustainable and equitable outcome for interventions (IAIA, 2009; Vanclay, 2002). SIA framework refers to alterations to the environment as biophysical changes (Slootweg et al., 2001). Biophysical changes result in intentional and unintentional social impacts but social influences also result in impacts to the biophysical environment– or in this study, the remediation and health response. Six categories of indicator variables were modified from Vanclay (2002) to determine social impacts of and on the environmental health response project, including: way of life, culture, community, political systems, environment, and health (Table 2). Relevant indicator variables were grouped into these six categories and given a positive, negative, neutral, or combination of positive and negative based on how the project impacted each indicator and how each indicator impacted the project.

## 4.4 Results

### 4.4.1 Area Weighted *In Situ* Soil Lead Levels

Table 3 compares pre- and post-remediation *in situ* soil lead concentrations and central tendency metrics to the results for the 40 compounds with AWA analyses. Two outlier homes were excluded from the analysis based on residual results. The best PXRF central tendency statistic for pre-remediation AWA is the arithmetic mean of all readings obtained for the home (slope = 1.04, $R^2$ = 0.99, intercept not significant, N=38). This 1:1 relationship confirmed the use of PXRF home arithmetic means as an appropriate estimate of *in situ* soil lead concentration. Typically, arithmetic mean is the favored central tendency statistic for exposure and risk assessment (Crump, 1998; Parkhurst, 1998; Powell, 2003). Table 3 summarizes this *in situ* exposure metric by village for all pre-remediation, post-remediation, and REE homes. ANOVAs comparing overall pre- and post-remediation geometric means by village show soil lead exposures were reduced by 96% in Dareta and by 97% in Yargalma in Phase I, and by 81% in Abare in Phase II (Table 3 and Figure 1). Pre-remediation soil Pb estimates were previously found to significantly differ by Phase due to the dilution effect of the intervening rainy season, with Phase I concentrations about 74% greater than Phase II (Tirima, et al. 2016). Post-remediation AWA estimates are not significantly different between Phases, indicating the remedy was similarly effective in all villages. However, it is important to note that post-excavation concentrations were significantly greater than post-remediation levels following application of clean soil ($p<0.0001$), emphasizing the importance of soil cover to protectiveness of the remedy and human health.

### 4.4.2 REE Environmental Results

Figure 1 shows pre- and post-remediation and REE geometric mean *in situ* PXRF soil lead levels by village. Histograms of pre- and post-remediation average home lead concentration are presented in Figures 2 and 3, respectively. ANOVAs comparing REE soil concentrations to pre- and post-remedial exposures indicate significant recontamination has occurred in some homes. The September 2010 REE undertaken three months following Phase I remediation in Dareta showed geometric mean *in situ* lead levels in randomly selected homes had increased significantly, following the intermittent rainy season, from 58 mg/kg post-remediation to 325 mg/kg ($p<0.0001$), but remained considerably below pre-remediation levels. However, the 2010 REE geometric mean exposure (3389 mg/kg) in homes identified by MSF had returned to pre-remediation geometric mean conditions (1366 mg/kg). These families had re-engaged in mineral processing in the home with attendant adverse blood lead outcomes. The 2011 REE conducted 5 months after completion of Phase II remediation investigated only homes identified by MSF as those with children not responding positively to chelation therapy. Geometric mean *in situ* soil lead exposures in these homes were found to be 665 mg/kg, 439 mg/kg, and 929 mg/kg in Abare, Dareta, and Yargalma, respectively, indicating significant increases from post-remediation levels ($p<0.0001$). However, multiple comparisons indicated that most of the recontamination among the 27 compounds tested was confined to 3 families that continued to engage in mineral processing in the home, despite the admonitions of local authorities. Figure 4 shows histograms of residential compound means for the three villages assessed during the 2011 REE.

PXRF sampling during the 2012 REE in Bagega village (data not shown) found overall in *situ* lead levels in the six homes decreased from 1012 mg/kg pre-remediation to 377 mg/kg. In the three homes where remediation was considered complete, average *in situ* lead was 237 mg/kg, though maximum lead concentration under the clean soil cover in two of the three homes was >1000 mg/kg. In two of the three homes where excavation was incomplete, max *in situ* lead exceeded 10,000 mg/kg, with 697 mg/kg average lead for the three homes. During the remediation effort, PXRF data were not entered for tracking progress or confirmation of remediation. Further, homes selected for soil removal were not sequenced spatially to avoid cross contamination of remediated areas when transferring excavated soils to the landfill.

The most extensive REE was the FYR conducted in Abare and Dareta in March of 2016. Figure 5 shows *in situ* soil lead concentration histograms for pre- and post-remediation and both random and MSF targeted 2016 REE compounds for both villages. Geometric mean *in situ* soil lead levels for randomly selected homes in Dareta (276 mg/kg) did not significantly increase from the 2010 REE (325 mg/kg), whereas levels in randomly selected Abare homes (612 mg/kg) were recontaminated to levels not significantly different from targeted homes (883 mg/kg) or from pre-remediation lead concentrations ($p<0.0001$). Dareta targeted homes showed significantly higher concentrations (1323 mg/kg) than randomly selected homes (276 mg/kg) ($p<0.0001$). These results indicate the remedy remains functional for most of Dareta but is failing in much of Abare. By February 2017, MSF had closed treatment clinics in four of the eight Zamfara State villages after children's BLLs dropped below the chelation threshold of 30 μg/dl. Two villages, Bagega and Yargalma, are expected to close in 2017.

### 4.4.3 Comparison of *In Situ* PXRF and Sieved *Ex Situ* Soil Lead Levels

Comparisons of *ex situ* bulk to *in situ* values at all concentrations showed a slope of 1.17, $R^2$ of 0.96, and insignificant intercept ($p<0.0001$). At concentrations <2000 mg/kg the slope was 1.10 and $R^2$ 0.86 with no intercept value ($p<0.0001$), indicating *ex situ* samples were appropriately collected at *in situ* PXRF locations. For the *in situ* to ICP-MS conversion, model forms included simple correlations and log transformation of both variables at various concentration ranges. In general, *in situ* XRF readings show lower concentrations than sieved laboratory results and the relationship is concentration dependent. Several generalizable and site-specific factors, including soil moisture content, gradation, matrix effects, relative metals levels, and XRF performance characteristics may be reflected in this relationship. As a result, it was determined to develop a simple empirical conversion coefficient using a linear model accommodating the remedial action criteria (400 mg/kg and 1000 mg/kg) and the majority of concentration readings observed during remediation (ND-2000 mg/kg *in situ* PXRF). This range includes 64% of all Yargalma readings which had the highest observed pre-remediation mean PXRF soil lead concentration (geomean 2206 mg/kg). The following model was selected as an empirical conversion factor to estimate minus 80-mesh (179 µm) sieved fraction ICP-MS concentrations from *in situ* PXRF values for determination of dose estimates or comparison to literature values (slope = 2.8, $R^2$ = 0.85, intercept not significant $p<.0001$, N=9): Sieved ICP-MS (mg/kg) = 2.8* *in situ* PXRF (mg/kg)

Applying this conversion factor to the *in situ* PXRF readings utilized in remediation significantly increases lead concentrations when applied as a soil exposure metric (Table 4). Estimated pre-remediation and REE fine soil fraction concentrations are 2.8 times greater than those reported in Tables 2 and 3 and Figures 2-5, and in earlier reports (von Lindern et al. 2011; Tirima, et al. 2016). The relationship is not straightforward for post-remediation concentration estimates, as the conversion factor does not apply to ND levels for clean soil, although it is likely pertinent to the (ND-399 mg/kg category) soils that remained exposed following remediation. It should be noted that this adjustment slightly increases both post-remediation exposures and the percent reduction in soil lead exposures achieved through remediation.

### 4.4.4 REE Qualitative Results

Observations from the SIA are presented in Table 6. For each indicator, the effect (positive, negative, or neutral) of the project on the indicator and the social impact on project efficacy is provided with an example. In 2011, miners reported dissatisfaction with a lack of options for safe storage of working materials and ore at the *dabas*. While most households repeated, nearly verbatim, the rules of not bringing mining tools and ore home, in 2011, working materials were observed in 50%, 80%, and 55% of homes in Abare, Dareta, and Yargalma, respectively. Much of the moderate Pb levels (400-1000 mg/kg) found in homes occurred in areas where mining materials were stored daily. In 2016, working materials were observed in 31% and 81% of homes in Abare and Dareta, respectively, despite people from only 16% and 37% of households interviewed admitting to storing these materials inside their homes. Although 63% of households reported that miners washed their clothes outside the home, laundry and bathing areas consistently tested high for Pb (>1000 mg/kg). In 2011 and 2016, the team observed that young children regularly visit the *dabas*. Most parents (92%) reported that children spend up to 5 hours per day away from the home, suggesting considerable risk of exposure.

One of the critical causes of the lead poisoning outbreak in 2010 was that ore had to be processed in the homes for women to participate in the lucrative industry. In this region, Sharia law includes *purdah,* the sequestration of women after marriage, making it unusual for women to go outside the home for work, especially in rural areas. When ore processing within villages was banned by traditional leadership, women lost the ability to participate in the gold industry and instead returned to cooking food for their young daughters (approximate ages 5-13) to sell. Inadvertently, this may have created a greater push for young girls to visit *dabas* to sell food for their mothers. This practice also results in unsold (and likely contaminated) food being brought home for consumption at the end of the day. During the 2016 REE, recontamination of homes in Dareta was linked to young men (roughly ages 13-20) processing in their family homes. It is likely these teenagers were previously exposed to dangerous levels of lead as children during the initial outbreak in 2010. These exposures put an entire generation of children at risk of behavioral, learning, and social problems, among other long-term health risks. As these children grow up, previous lead exposure in childhood, in addition to the lucrative draw of the mining boom, may further hamper efforts to keep them from processing ore in the villages.

Mitigation measures to address these issues were discussed with formal and informal leaders. An aggressive enforcement campaign by the local Emirate following 2010 recontamination in three Dareta homes required the offending families to self-remediate. In Abare and Dareta, weak village leadership was identified as a contributing factor to recontamination in 2011, when households found to be processing were similarly admonished and required to remediate. In response, Phase III remediation outreach in Bagega village included community-lead discussions on how best to incorporate “safer mining” into the mining camps and community. While mining leadership has been vigilant in enforcing bans on residential ore processing in Bagega (Tirima et al., 2016), similar levels of local enforcement are lacking in other locations.

The 2012 REE focused on assessing the technical capacity of Nigerian government personnel to implement and sustain the remedy. Several project successes were highlighted, including excellent community outreach and replication of technical protocols. Shortcomings were identified in geographical sequencing to avoid contaminating remediated areas and in management of PXRF data. The abbreviated remediation effort was halted prematurely due to a stoppage in funding, an issue that was generally attributed to government corruption, a common issue in Nigeria (Transparency International, 2015). The strategy for 2013 remediation in Bagega village and 2016 remediation in Niger State was adapted based on these findings; MSF required homes to be certified by TIFO as fully remediated before initiating medical treatment.

## 4.5 Discussion

### 4.5.1 Summary REE/SIA Findings

Each REE effort was conducted with limited resources and with specific, time-critical goals focused on medical outcomes. Sampling and exposure assessments concentrated on investigation of possible sources of residential or para-occupational exposures associated with persistent blood lead problems. Villages where BLLs consistently declined and programs were closed once the medical programmatic goals had been achieved were not investigated. As a result, the findings of this study are biased toward villages where the response has been less successful. It should be remembered that BLLs have been successfully reduced and the program is expected to close for more than 75% of the effected population by 2017. Environmental results from the REEs must be placed in the context of the findings of the SIA, as project resilience is heavily influenced by the socio-economic context of artisanal mining in these communities (Figure 6). As of the 2016 REE, the only apparent institutional controls for the sustaining the remedy in the remediated villages were informal (mining, religious, or youth leaders) or formal (village or emirate leaders) efforts. These methods appear to have been effective in most villages as BLLs declined and treatment programs closed, with Dareta and Abare being exceptions. The capacity to respond to lead poisoning outbreaks has been built in the traditional, local, and state governments, but the efforts to coordinate prevention activities at a regional level is lacking. This is apparent in Abare and Dareta, where both formal and informal institutions have failed, as well as in the 2016 Niger State lead poisoning outbreak, which was preventable given political dedication to a regional safer mining effort.

Dareta and Abare have been the focus of REEs since shortly after remediation was completed in each location. In Dareta in 2010, recontamination was found in isolated homes which subsequently self-remediated by order of the Emir. Concurrently, outreach and community engagement components of the remediation program were strengthened in Phase II villages. In Abare, Dareta, and Yargalma in 2011, moderate recontamination was identified as being largely linked to para-occupational and occupational hygiene issues. Advocacy programs in conjunction with MSF and local youth, mining, and religious leadership were enhanced and communication with mining leaders was an additional focus of the Phase III remediation. Stakeholders promoted moving *dabas* further from villages to discourage children from visiting and worked towards better local controls to keep mining out of villages. In Yargalma, these advocacy efforts were fully adopted by local leadership, who were successful in addressing these issues. However, Abare and Dareta were the focus of the 2016 REE targeting villages with persistently high BLLs in children: in Dareta, severe recontamination was observed in targeted homes and low-level para-occupational recontamination was observed in others; in Abare, in addition to severe individual situations, moderate recontamination occurred throughout the village. In these two villages, on-going chelation treatment may be enabling risky behavior by masking the effects of severe lead exposure. The lack of clinical symptoms in children, including those still under medical treatment for high BLLs, makes it more difficult to emphasize the dangers of lead exposure in childhood. Childhood malnutrition, malaria, and endemic disease are common in every village (Ahmad and Hassan, 2016; Alabi et al., 2016; Garba et al., 2014; UNICEF, 2016), but 91% of children on chelation treatment had no neurological symptoms of lead poisoning (Greig et al., 2014).

The second outbreak of lead poisoning in Niger State and the majority recontamination problems in Zamfara are exacerbated by endemic poverty. Agricultural yields are decreasing markedly due to climate change, market disruptions, and ethnic conflicts among farmers and herdsman (Oguamanam, 2016). Mining is one of the few income-producing alternatives to subsistence farming, migration to cities, or recruitment by extremist organizations. The loosely organized artisanal mining practices in these communities are technically illegal, but not rigorously regulated. Small miners generally oppose regulation, as it subjects them to registration, taxation, royalties and exploitation by corrupt government or industry institutions. Since the 2016 REE and Niger State remediation, cooperative efforts have been undertaken to develop safer mining methods that reduce occupational and para-occupational exposures. Efforts are underway to implement these practices at the village level where generally more effective local institutional controls can be employed. However, as noted in the recalcitrant Zamfara communities, effectiveness of local controls can vary and be deleterious in the absence of responsible leadership.

#### 4.5.2 PXRF and Remedy Protectiveness

Lead concentrations used as indices of exposure for young children generally refer to the fine fraction of the soil. Smaller particles are more conducive to the hand-to-mouth incidental ingestion pathway, where particles <150 µm adhere to the hands through electrostatic processes and are ingested as children mouth their hands or other objects (USEPA, 2016). Particles <100 µm are more likely to be suspended in the atmosphere, transported to children's environments, and subject to ingestion or inhalation (WHO, 1999). Particles <10 µm are subject to deposition in the lungs where higher absorption rates occur (WHO, 1999). The severe BLLs, observed mortality and morbidity, lack of laboratory services, remote location, and logistical constraints did not allow for analyses of lead concentration as a function of particle size. The Zamfara and Niger State remediation relied instead on *in situ* PXRF for rapid environmental media measurements. Although, i*n situ* PXRF soil and dust concentrations can be effective exposure indices, these are not directly comparable to levels generally observed in scientific literature and public health guidance. Results from additional analyses suggest that estimated -80 mesh small particle concentrations in the 2016 Zamfara REE are 2.8 times greater than the PXRF *in situ* values*.* Estimated geometric mean -80mesh soil lead concentrations in Phase I, II and III villages were 4679 mg/kg, 1233 mg/kg, and 1008 mg/kg, respectively. The divergence in PXRF and the adjusted exposure estimates, however, had little effect on the protectiveness of the applied remedy, as the Nigerian cleanups were designed to account for these potential differences by removing and/or replacing all soils >400 mg/kg Pb *in situ (*estimated 1120 mg/kg -80 mesh) with maximum 25 mg/kg clean soil, resulting in estimated 110 mg/kg to 242 mg/kg -80 mesh geometric means by village. These criteria and resultant aggregate soil/dust lead concentrations were similar to those employed at the BHSS (i.e., 1000 mg/kg removal threshold, 100 mg/kg clean soil, 350 mg/kg community mean) (USEPA, 2010, 2005, 1999). While these values are significantly lower than the USEPA standard for residential soils, soil/dust ingestion rates are likely higher in this region of the world, where people live in homes constructed with adobe bricks and dirt floors, eat on the ground, and have no reprieve from soil exposures.

This investigation confirmed findings from other studies that PXRF is an excellent tool for rapid, field-based decisions (Carr et al., 2008; Pyle et al., 1996; Stark et al., 2008). The emergency nature of the response required real-time soil heavy metal information to effectively implement the remediation and allow children access to medical treatment. Collecting representative soil samples from each of the 945 homes characterized and waiting for laboratory results would have been both time and cost prohibitive.

**4.5.3 REE and SIA Process**

The REEs highlighted the importance of both comprehensive environmental remediation goals and inclusion of all stakeholder groups in identifying and addressing the social, cultural, and economic challenges to sustaining the remedies. Most REE efforts focus on the technical aspects and resultant environmental media and exposure metrics, neglecting the social and political aspects that often determine the long-term effectiveness of remediation (Elias and Gulson, 2003). SIA is a tool adapted for assessing and addressing some of the social and political issues often neglected by REEs. Biophysical and social changes are equally important components of any intervention (Figure 7). This study demonstrates that not only does remediation have social impacts, but social impacts have critical importance to the long-term success of remediation.

While initiating REEs and SIAs as early as possible in emergency responses is ideal (Watson, 2008), implementing a full assessment prior to initiating work in Zamfara was not feasible. Both practices involve methodological planning and assessment typically not conducive to emergency humanitarian intervention (Vanclay, 2003; Watson, 2008). One of the main constraints to successful impact assessment in emergency situations is time. Further, actors in emergency humanitarian responses often assume that all impacts are positive. Other than the Superfund FYR model, there are few, if any, standardized methods for conducting post-remediation evaluations (Apitz et al., 2005). The few documented REEs conducted outside the US have found that fragmented approaches to remediation, *in situ* stabilization or cap and cover techniques, and monitored natural recovery are not sustainable and are generally ineffectual at reducing overall risk to the humans and the environment (Boreland et al., 2009; Elfvendahl et al., 2004; Gray et al., 2006; Harvey et al., 2016; Phenrat et al., 2016). Research in Tanzania and Australia confirm that partial approaches to procedural aspects of remediation do not adequately protect human health (Boreland et al., 2009; Elfvendahl et al., 2004; Harvey et al., 2016). *In situ* stabilization and behavioral modification techniques implemented without long term community and industry support or follow-up in Kabwe, Zambia resulted in transient exposure reductions and children BLLs returned to pre-intervention levels (Bose-O'Reilly et al., 2016; Brink, 2016). Implementation and evaluation of remediation projects are more challenging in areas with limited involvement from government or mining leadership (Elias and Gulson, 2003), highlighting the importance of social influences on project resilience.

**4.6. Conclusions**

This ex-post analysis of four REEs confirms findings in Tirima et al. (2016) that soil remediation significantly reduced soil heavy metal concentrations, resulting in reduced lead exposures and children's BLLs. Utilizing PXRFs to characterize soil metal concentrations has proven to be a valuable and reliable method for rapid environmental risk assessment and remediation. In two of the eight Zamfara villages, resumption of ore processing and lack of leadership to enforce institutional controls has resulted in recontamination and elevated BLLs. Children's BLLs in the remaining villages have decreased, indicating that the primary remedial action goals are achievable. Further reductions in BLLs are dependent on maintaining minimum environmental media concentrations. Processing in the home is the most severe threat to sustaining the remedy.

Findings in this analysis highlight the interrelated nature of environmental and social impacts noted by other authors (Barrow, 2002; Elias and Gulson, 2003; Slootweg et al., 2001). Employing rigorous REE inclusive of retrospective SIA is crucial for attempting to understand the complex biophysical interactions of the intervention and the cultural contexts. The concept of using SIA to determine the social impacts on an intervention allows for more effective management of impacts and greater potential for addressing challenges to sustainability. For example, alternative sources of income in these communities are limited; artisanal mining was consistently reported as the major source of income in the affected communities. Long-term solutions must be adapted to be both protective of human health and sensitive to the economic and social realities. The results from the REEs and the 2016 lead poisoning outbreak in Niger State highlight the importance of addressing safer mining that, to date, has been lacking at higher levels of formal leadership. In some villages, mining and traditional leadership have successfully tackled this role, but to curtail future outbreaks nationally, formal and informal institutions must be supported.

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Table 4.1. Homes and public areas in the eight Zamfara villages affected by the lead poisoning outbreak. The number of homes and public areas assessed during characterization and REE efforts are indicated by village, along with the medical treatment status.

| | | **Remediation** | | | **Medical Treatment** | **2010 REE** | | | **2011 REE** | | | **2012 REE** | | **2016 REE** | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Village | Phase | Homes Tested | Homes Remediated | Public Areas Remediated | Status | Targeted Homes | Random Homes | Public Areas | Targeted Homes | Random Homes | Public Areas | Targeted Homes | Random Homes | Targeted Homes | Random Homes | Public Areas |
| Dareta | I | 94 | 85 | 13 | On-going | 3 | 11 | 6 | 9 | 0 | 5 | . | . | 19 | 22 | 28 |
| Yargalma | I | 66 | 63 | 11 | to close 2017 | . | . | . | 9 | 0 | 11 | . | . | . | . | . |
| Abare | II | 96 | 74 | 20 | On-going | . | . | . | 96 | 74 | 20 | . | . | 16 | 24 | 37 |
| Duza | II | 57 | 57 | 8 | complete | . | . | . | . | . | . | . | . | . | . | . |
| Sunke | II | 93 | 83 | 38 | complete | . | . | . | . | . | . | . | . | . | . | . |
| T. Daji | II | 78 | 75 | 31 | complete | . | . | . | . | . | . | . | . | . | . | . |
| T. Guru | II | 38 | 31 | 6 | complete | . | . | . | . | . | . | . | . | . | . | . |
| Bagega | III | 423 | 352 | 54 | to close 2017 | . | . | . | . | . | . | 3 | 3 | . | . | . |
| **Totals** | | **945** | **820** | **181** | **.** | **3** | **11** | **0** | **114** | **74** | **20** | **3** | **3** | **35** | **46** | **65** |

Table 4.2. Categories of indicator variables used in social impact assessment. Modified from Vanclay (2002).

| **Health and Well-Being** | **Culture** | **Community** | **Formal Institutions** | **Way of Life** | **Environment** |
|---|---|---|---|---|---|
| death in community | observance of religious practices | obligations to elders and leaders | increased workload for institutions | access to goods/services | adequacy of infrastructure |
| perceived health | participatory processes | changed demographics | integrity of agencies | occupational status | disruption to daily living |
| deviance labeling | children's behavior | social tension | capacity to respond | level of unemployment | access to healthcare |
| uncertainty | | | participation in decision making | economic vulnerability | |
| feelings towards intervention | | | | | |
| dissatisfaction | | | | | |
| fear of arrest/loss of income | | | | | |

Table 4.3. Regression results of area weighted average (AWA) lead levels compared to other central tendency statistics for pre-remediation *in situ* PXRF data in 38 homes (mg/kg).

| | | N | AWA | Central Tendency Statistics | | | |
|---|---|---|---|---|---|---|---|
| | | | | Arithmetic Mean | Q3 | Median | Geometric Mean |
| Phase | I | 12 | 3556 | 3466 | 3094 | 1603 | 1738 |
| | II | 12 | 1216 | 1141 | 1323 | 567 | 612 |
| | III | 14 | 2044 | 1906 | 1942 | 841 | 780 |
| | All | 38 | 2260 | 2157 | 2110 | 995 | 1030 |
| Comparison to AWA | | $R^2$ | | 0.99 | 0.64 | 0.65 | 0.50 |
| | | P | | <0.0001 | <0.0001 | <0.0001 | <0.0001 |
| | | Slope | | 1.04 | 1.55 | 2.01 | 2.95 |

Table 4.4. Geometric mean and geometric standard deviation (gsd) *in situ* PXRF soil lead concentrations by village and REE effort (mg/kg Pb; n/a = not available, data were not collected for this village during the relevant time period).

| | | **Abare** | | | **Dareta** | | | **Yargalma** | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | | N | geomean | gsd | N | Geomean | gsd | N | geomean | gsd |
| All Homes | Pre-rem | 96 | 457 | 4.1 | 91 | 1366 | 4.9 | 66 | 2206 | 3.5 |
| | Post-excavation | 50 | 114 | 1.4 | 12 | 111 | 1.5 | 20 | 148 | 1.6 |
| | Post-rem | 96 | 86 | 1.7 | 91 | 58 | 1.9 | 66 | 56 | 1.7 |
| Random Homes | 2010 REE | n/a | n/a | n/a | 11 | 325 | 4.3 | n/a | n/a | n/a |
| | 2016 REE | 25 | 612 | 3.4 | 31 | 276 | 3.2 | n/a | n/a | n/a |
| Targeted Homes | 2010 REE | n/a | n/a | n/a | 3 | 3389 | 3.0 | n/a | n/a | n/a |
| | 2011 REE | 9 | 665 | 2.4 | 9 | 439 | 9.0 | 9 | 929 | 2.5 |
| | 2016 REE | 16 | 883 | 2.2 | 20 | 1323 | 20.0 | n/a | n/a | n/a |
| Public Areas | Pre-rem | 26 | 808 | 5.0 | n/a | n/a | n/a | 10 | 3275 | 2.1 |
| | Post-rem | 26 | 78 | 2.1 | n/a | n/a | n/a | 10 | 46 | 1.6 |
| | 2010 REE | n/a | n/a | n/a | 6 | 256 | 2.7 | n/a | n/a | n/a |
| | 2011 REE | 4 | 2454 | 3.5 | 5 | 434 | 1.9 | 11 | 741 | 3.0 |
| | 2016 REE | 37 | 1241 | 5.3 | 28 | 1147 | 6.1 | n/a | n/a | n/a |

Table 4.5. Pre-remediation, post-excavation, and post-remediation (clean soil cover) statistics for *in situ* PXRF results and the conversion to analogous *ex situ* sieved (-80 mesh) ICP-MS results (mg/kg).

| | **Pre-Remediation *in situ*** | | | | | **Pre-Remediation Converted** | | | |
|---|---|---|---|---|---|---|---|---|---|
| Village | N | Mean | std | Geomean | gstd | Mean | std | Geomean | gstd |
| Dareta | 91 | 3490 | 4421 | 1366 | 4.9 | 9773 | 12379 | 3826 | 4.9 |
| Yargalma | 66 | 4143 | 4786 | 2206 | 3.5 | 11601 | 13400 | 6177 | 3.5 |
| Phase I | 157 | 3765 | 4574 | 1671 | 4.3 | 10541 | 12808 | 4679 | 4.3 |
| Abare | 96 | 1343 | 2724 | 457 | 4.1 | 3761 | 7628 | 1280 | 4.1 |
| Duza | 42 | 300 | 367 | 179 | 2.8 | 841 | 1027 | 501 | 2.8 |
| Sunke | 82 | 861 | 879 | 547 | 2.7 | 2410 | 2460 | 1531 | 2.7 |
| Tungar daji | 75 | 780 | 848 | 522 | 2.4 | 2183 | 2374 | 1461 | 2.4 |
| Tungar guru | 38 | 1118 | 2087 | 486 | 3.9 | 3129 | 5843 | 1360 | 3.9 |
| Phase II | 333 | 940 | 1756 | 440 | 3.3 | 2633 | 4917 | 1233 | 3.3 |
| Bagega / Phase III | 423 | 831 | 1574 | 360 | 3.3 | 2328 | 4407 | 1008 | 3.3 |
| | **Post-Excavation XRF *in situ*** | | | | | **Post-Excavation XRF Converted** | | | |
| Village | N | Mean | std | Geomean | gstd | Mean | std | Geomean | Gstd |
| Dareta | 12 | 120 | 43 | 111 | 1.5 | 336 | 121 | 312 | 1.5 |
| Yargalma | 20 | 161 | 64 | 148 | 1.6 | 451 | 179 | 414 | 1.6 |
| Phase I | 32 | 146 | 60 | 133 | 1.6 | 408 | 167 | 372 | 1.6 |
| Abare | 50 | 118 | 30 | 114 | 1.4 | 331 | 85 | 318 | 1.4 |
| Duza | 26 | 116 | 44 | 106 | 1.6 | 323 | 124 | 296 | 1.6 |
| Sunke | 68 | 163 | 120 | 130 | 2.0 | 457 | 335 | 365 | 2.0 |
| Tungar daji | 59 | 125 | 59 | 115 | 1.5 | 349 | 166 | 321 | 1.5 |
| Tungar guru | 26 | 209 | 103 | 190 | 1.5 | 584 | 287 | 532 | 1.5 |
| Phase II | 229 | 143 | 87 | 125 | 1.7 | 401 | 244 | 349 | 1.7 |
| Bagega / Phase III | 266 | 156 | 132 | 127 | 1.9 | 438 | 371 | 356 | 1.9 |
| | **Post-Remediation (Clean Soil) *in situ*** | | | | | **Post-Remediation (Clean Soil) Converted** | | | |
| Village | N | Mean | std | Geomean | gstd | Mean | std | Geomean | Gstd |
| Dareta | 91 | 69 | 42 | 58 | 1.9 | 165 | 130 | 111 | 2.7 |
| Yargalma | 66 | 64 | 39 | 56 | 1.7 | 144 | 119 | 108 | 2.2 |
| Phase I | 157 | 67 | 41 | 57 | 1.8 | 156 | 126 | 110 | 2.5 |
| Abare | 96 | 96 | 42 | 86 | 1.7 | 253 | 128 | 211 | 2.0 |
| Duza | 42 | 95 | 33 | 88 | 1.6 | 261 | 90 | 241 | 1.6 |
| Sunke | 82 | 107 | 37 | 100 | 1.5 | 283 | 110 | 253 | 1.7 |
| Tungar daji | 75 | 112 | 34 | 106 | 1.5 | 296 | 101 | 272 | 1.6 |
| Tungar guru | 38 | 108 | 42 | 97 | 1.7 | 284 | 119 | 252 | 1.7 |
| Phase II | 333 | 104 | 38 | 95 | 1.6 | 275 | 113 | 242 | 1.8 |
| Bagega / Phase III | 423 | 103 | 33 | 97 | 1.4 | 277 | 97 | 257 | 1.5 |

Table 4.6. Results of the ex-post SIA (using categories and indicator variables modified from Vanclay 2002).

| Categories | Indicators | | Social Impact of Environmental Health Project | | Social Impact on Project Sustainability |
|---|---|---|---|---|---|
| Health and Well-Being | death in community | +, - | tragically high mortality rates in children during the initial outbreak engendered strong community support for the environmental/health intervention; loss of life causes suffering in families and communities | +, - | fear of high mortality rates prevents adults from resuming ore processing; lack of clinical symptoms in children with elevated BLLs may create apathy, especially for youth |
| | perceived health | + | health messaging and community dialogues on lead exposure have increased awareness | - | lack of clinical symptoms in children with para-occupational exposures challenges behavior change efforts |
| | deviance labeling | + | project leaders continue to promote "safer mining" despite labeling by federal government | - | federal government considers artisanal mining to be "illegal", causing distrust and fear of reprisal |
| | uncertainty | Ø | no change | - | income and food stability remain uncertain; leftover food is not wasted - if it isn't sold at *dabas* it is brought home for consumption |
| | feelings towards intervention | + | community members, leaders, and formal government report appreciation for the intervention and continued support | + | positive feelings towards the intervention encourage future dialogue and collaboration, community dialogues about project challenges encourage local solutions |
| | dissatisfaction | +, - | most expectations were met or exceeded; some expectations, such as requests for storage at *dabas,* have not been met | +, - | dissatisfaction with certain actions (or lack thereof) by government hampers sustainability efforts |
| | fear of arrest/loss of income | + | project leaders have strongly discouraged any ban on artisanal mining and have worked with federal government to explore alternate solutions | +, - | locals understand the position of project leaders but are distrustful of government |
| Culture | observance of religious practices | + | religious views and practices, including Sharia law, are respected and accommodated | +, - | religious leadership plays important role in institutional controls; *purdah* and a banning of ore processing in residential areas prevents equitable access to mining jobs |
| | participatory processes | +, - | community engagement in early villages was limited due to emergency nature of response; later, extensive dialogues preceded remediation | +, - | community support and buy-in was stronger in Phase II and III villages, but possibly reduced in Phase I villages |
| | children's behavior | Ø | no change | - | children visit *dabas* |

| Categories | Indicators | | Social Impact of Environmental Health Project | | Social Impact on Project Sustainability |
|---|---|---|---|---|---|
| Community | obligations to elders and leaders | Ø | no change | +, - | cultural obligations to leaders aids resilience if leaders are engaged; weak leadership contributes to lack of institutional controls |
| | changed demographics | Ø | no change | - | outsiders living at mining camps have reduced incentive/obligations to community health |
| | social tension | - | possible increase in tension between family members where or neighbors when recontamination is attributed to specific individuals | +, - | peer-pressure may help with institutional controls; increased tension may result in loss of community participation in sustaining remediation |
| Formal Institutions | increased workload for institutions | + | project training increases capacity to sustain the remedy | - | increased responsibility of formal institutions is not matched with financial support from government |
| | integrity of agencies | +,- | project training encourages increased responsibility towards communities; outside assistance relieves government of responsibilities/accountability | - | endemic corruption a challenge to long-term sustainability |
| | capacity to respond | +, - | significant increase in technical capacity; international intervention required for data management and follow-through | +, - | trained Nigerian personnel results in fewer international staff needed and encourages local ownership; endemic corruption hampers assumption of responsibility by Nigerian government |
| | loss of subsidiarity | +, - | project leaders have strongly encouraged local solutions; international attention to the issue has resulted in federal statements about banning "illegal" mining | +, - | encouraging grassroots solutions increased community buy-in; fear of reprisal from federal level creates distrust |
| | participation in decision making | + | encourages ownership at all levels | + | encourages ownership at all levels |
| Way of Life | access to goods/services | + | improved roads, more clean water sources installed by government | + | increased goodwill towards intervention |
| | occupational status | Ø | project offered temporary employment only; mining an agriculture remain only employment options | +, - | temporary employment and income viewed positively; desire for more work could encourage recontamination |
| | level of unemployment | + | temporary employment, increased training | - | no other income options other than mining |
| | economic vulnerability | Ø | increased training, but no increase in job opportunities | - | no other income options other than mining |

| Categories | Indicators | | Social Impact of Environmental Health Project | | Social Impact on Project Sustainability |
|---|---|---|---|---|---|
| Environment | adequacy of infrastructure | + | infrastructure has improved in certain areas due to remediation and press coverage | - | overall lack of communication, transportation, health, and *daba* infrastructure presents challenges for sustainability |
| | disruption to daily living | +, - | repeated visits show commitment to supporting the communities; remediation and follow-up visits disrupt daily life, outside researchers may cause fatigue | +, - | rapport is built within the community; "research" (or intervention) fatigue is an issue |
| | access to healthcare | +, - | improves children's health; relieves government of responsibilities | + | engenders positive feelings towards project |

Figure 4.1. Geometric mean *in situ* PXRF soil lead concentrations by village and REE year. Groups with the same letter designation were analyzed via ANOVA and different numbers within letter groups indicate significant differences within that group. Significant differences were found between pre- and post-remediation soil lead concentrations within each village (i.e., between A1 and A2, between D1 and D2, and between Y1 and Y2). Significant differences were also found between post-remediation and REE soil concentrations within villages. Targeted homes in two of three villages (Abare and Dareta) and random homes in one village (Abare) have returned to pre-remediation conditions. (A=Abare, D=Dareta, Y=Yargalma, P=Phase, $p<0.0001$).

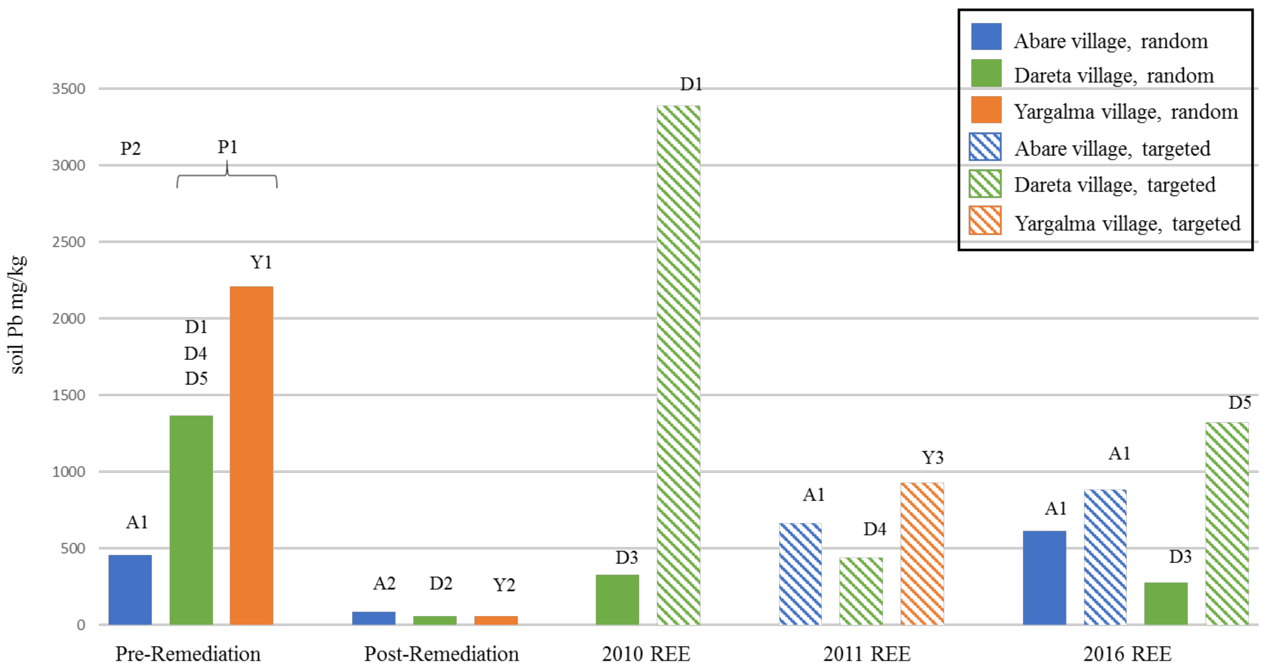


Figure 4.2. Histogram of pre-remediation *in situ* PXRF soil lead concentrations (mg/kg) in Abare, Dareta, and Yargalma homes. Data met assumptions for lognormal distribution and lognormal curves are fit to the histograms.

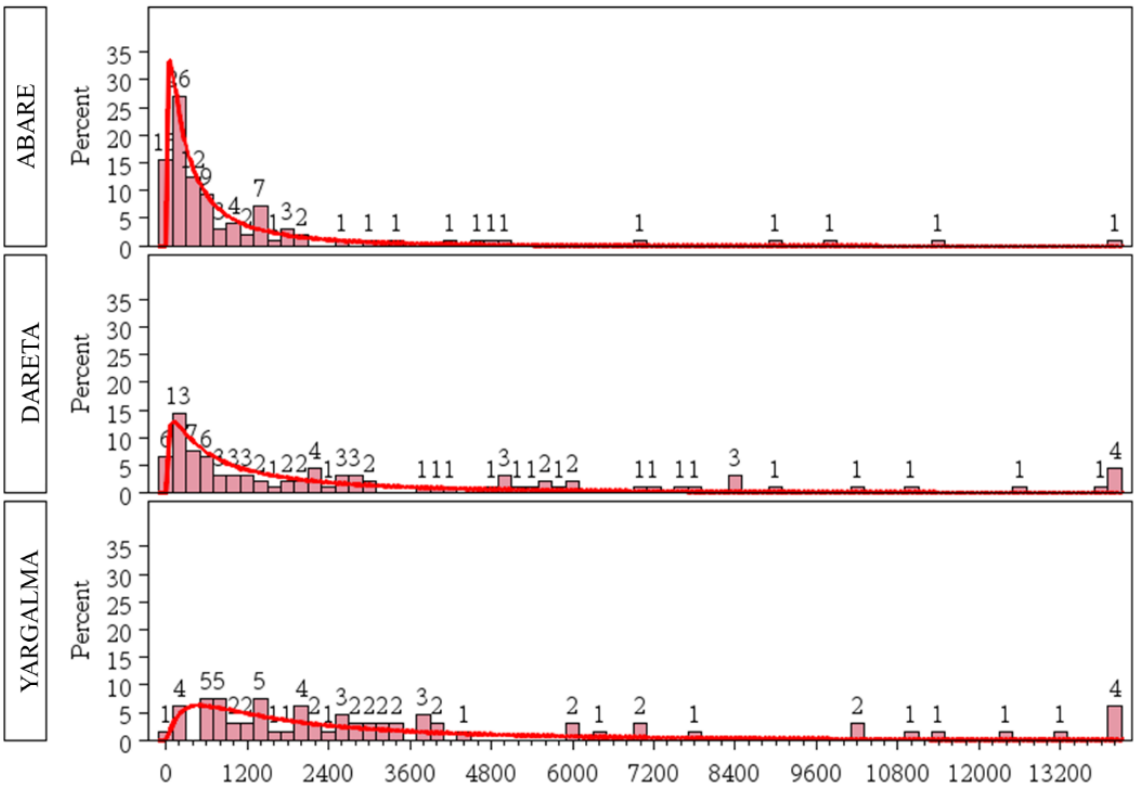


Figure 4.3. Histogram of post-remediation *in situ* PXRF arithmetic average soil lead concentrations (mg/kg) in Abare, Dareta, and Yargalma homes. Data did not meet assumptions for normal or lognormal distribution.

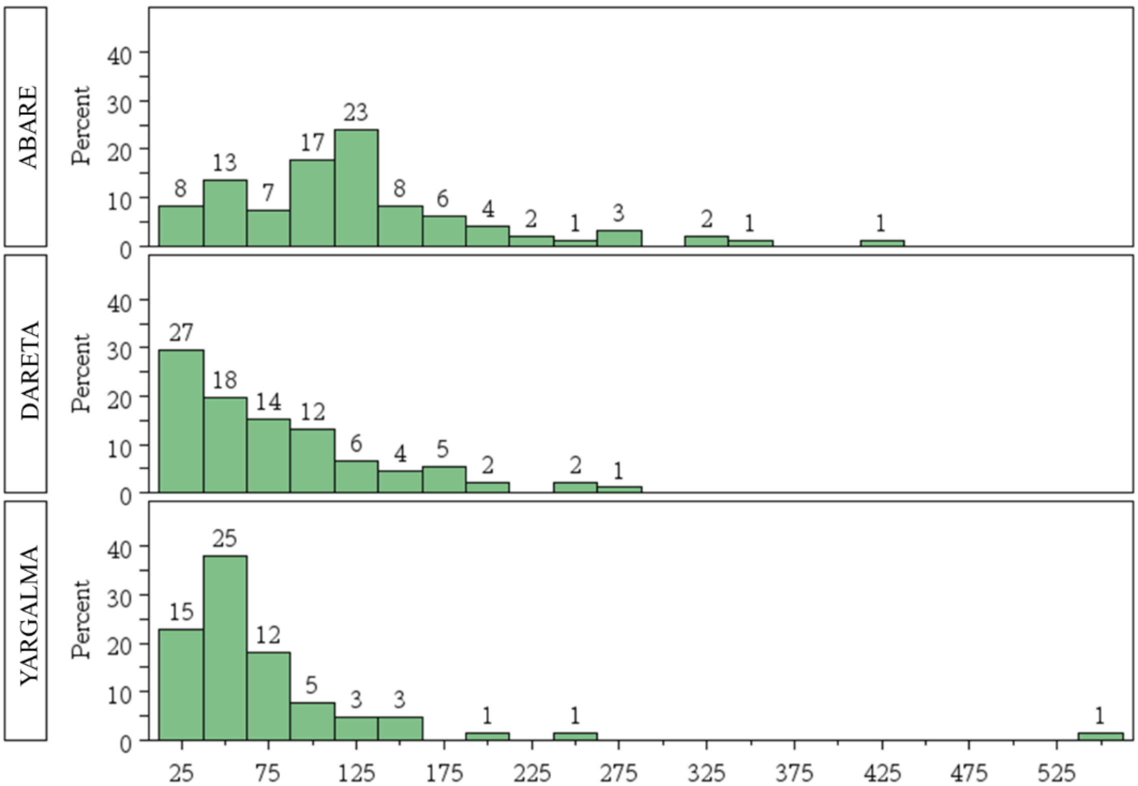


Figure 4.4. Histogram of 2011 REE *in situ* PXRF arithmetic average lead concentrations (mg/kg) in targeted Abare and Dareta homes (no randomly selected homes were assessed during this REE). Data met assumptions for lognormal distribution and lognormal curves are fit to the histograms.

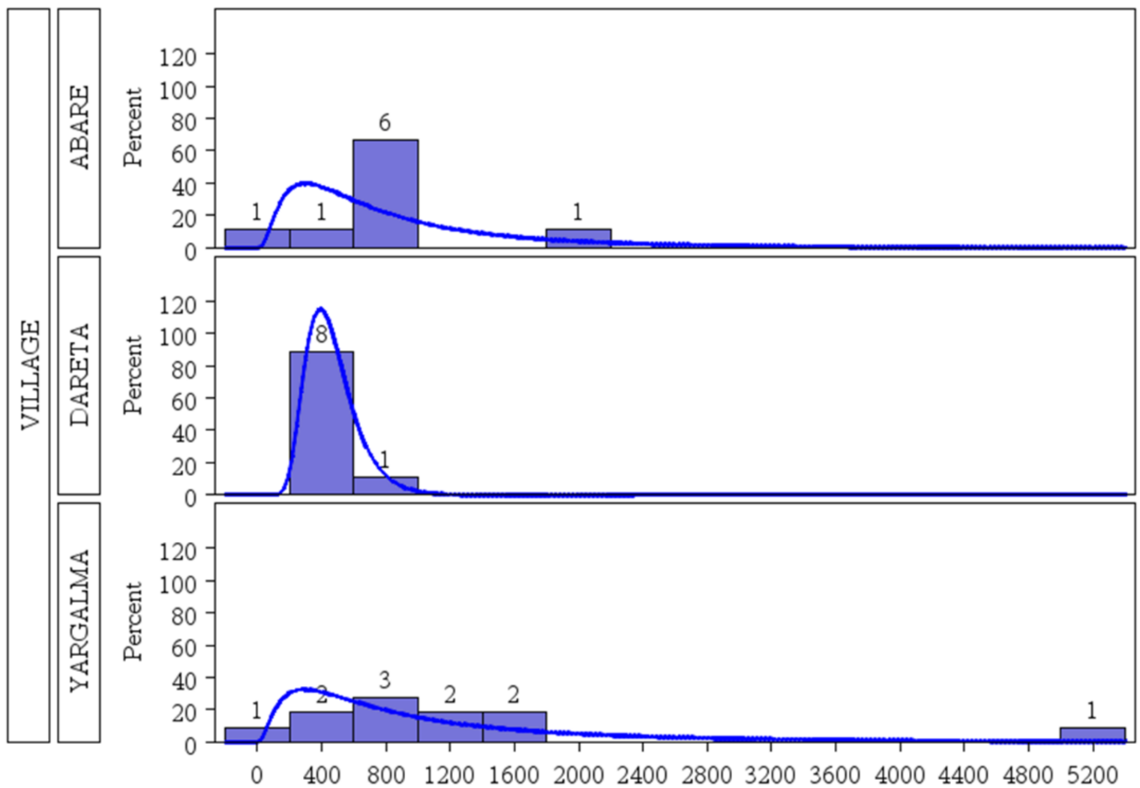


Figure 4.5. Histogram of 2016 REE *in situ* PXRF arithmetic average soil lead concentrations (mg/kg) in random and targeted Abare and Dareta homes. Data met assumptions for lognormal distribution and lognormal curves are fit to the histograms.

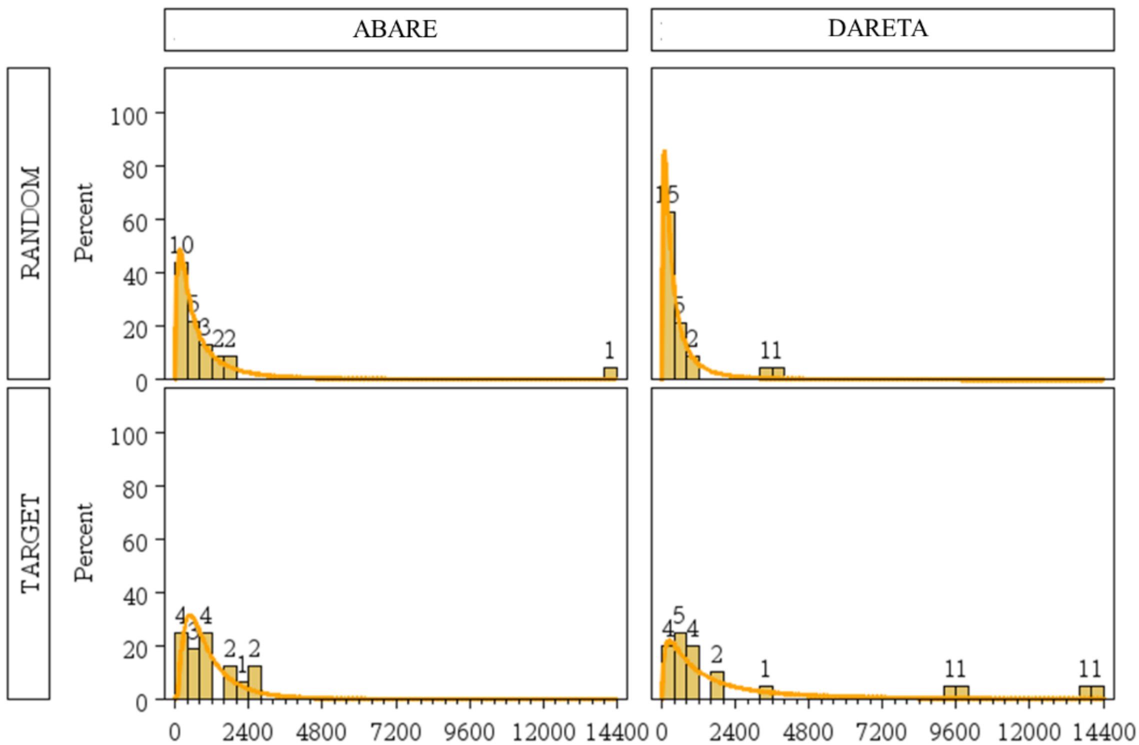


Figure 4.6. REE results in the framework of Social Impact Assessment. Observations and baseline data are the findings immediately apparent, influenced by the social, cultural, and economic context, and by non-mitigatable social impacts of artisanal mining and lead poisoning.

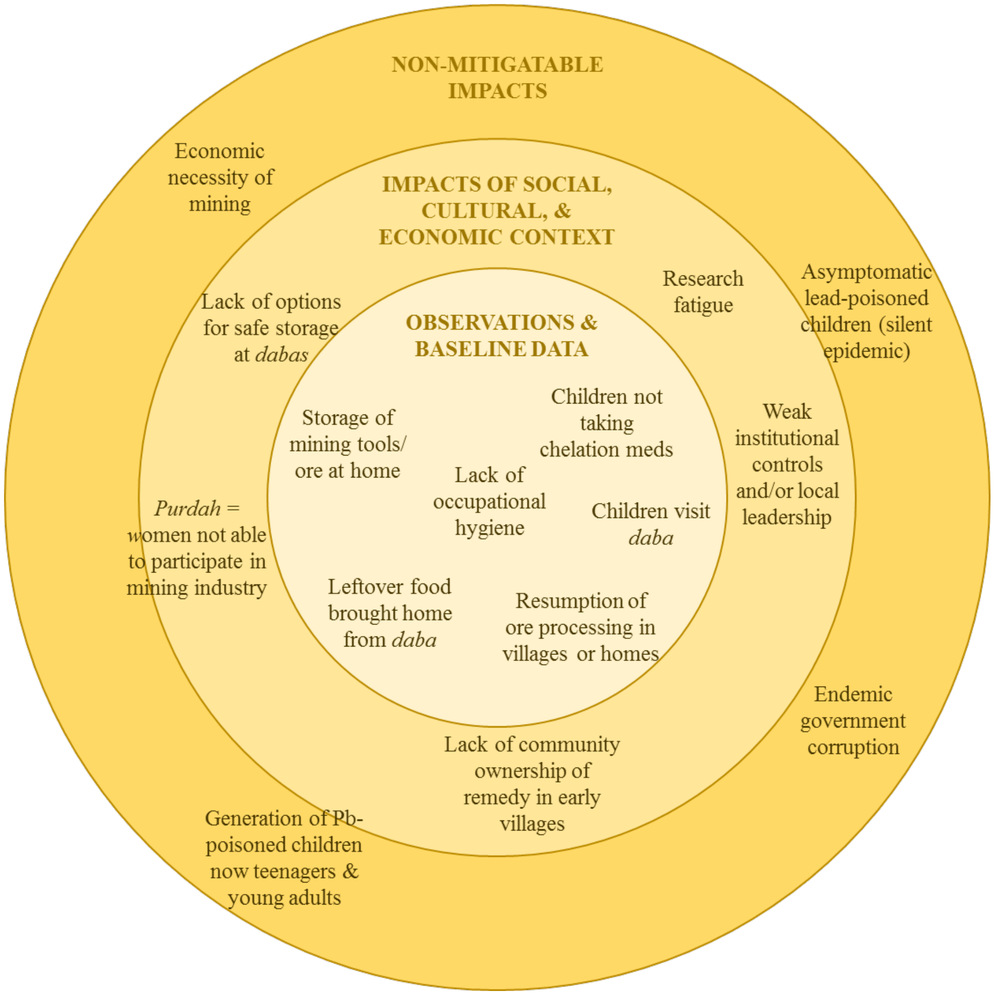
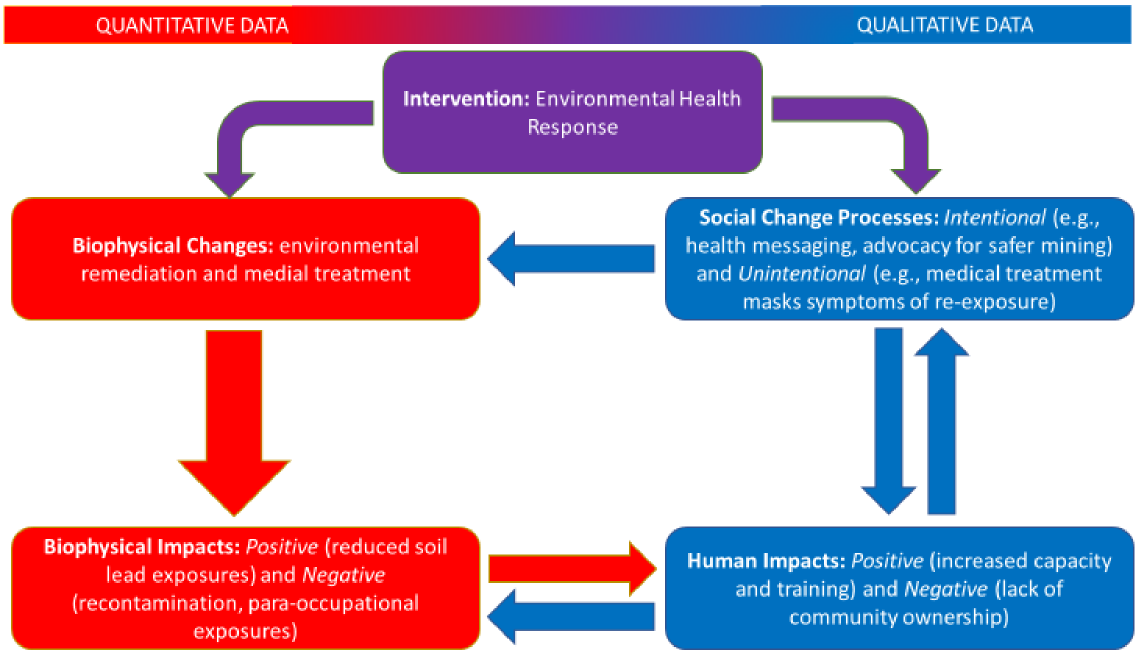


Figure 4.7. Schematic of biophysical (remediation and medical treatment) and social changes interacting to result in measurable physical and social impacts. Biophysical and social impacts also interact on and with each other and can be assessed via analysis of quantitative and qualitative data (Slootweg et al., 2001).



# CHAPTER 5 – LEGACY LEAD ARSENATE SOIL CONTAMINATION AT CHILDCARE CENTERS IN THE YAKIMA VALLEY, CENTRAL WASHINGTON, USA.

Durkee, J., Bartrem, C., Moller, G. "Legacy lead arsenate soil contamination at childcare centers in the Yakima Valley, Central Washington, USA." *Chemosphere,* vol. 168, 2017, pp. 1126-1135.

**Abstract**

**Background:** From the early 1900s to the 1950s, Yakima Valley orchards were commonly treated with lead arsenate (LA) insecticides. Lead (Pb) and arsenic (As) soil contamination has been identified on former orchard lands throughout Central Washington and pose a threat to human health and the environment. **Objectives:** The levels of Pb and As in soil and interior dust at participating childcare centers in the Upper Yakima Valley (Yakima County), Washington were sampled to explore exposure potential for young children. **Methods:** Childcare center soils were collected from two soil depths, homogenized, and analyzed in bulk by a field-portable X-ray fluorescence spectrometer (XRF). Interior dust wipes samples were collected from at least two locations in each facility. All soil samples >250 mg/kg Pb and/or >20 As mg/kg were sieved to 250 µm, tested by XRF a second time, and analyzed via acid digestion and inductively coupled plasma mass spectrometry (ICP-MS) analysis. **Results:** Bulk and sieved XRF results, as well as ICP-MS to XRF results were strongly correlated. Maximum Pb and As XRF results indicated that 4 (21%) and 8 (42%) of the 19 childcare centers surveyed exceeded the regulatory standard for Pb and As, respectively. Historic land use was significantly associated with elevated Pb and As levels. Interior dust loadings were below United States Environmental Protection Agency (EPA) guidelines. **Conclusions:** Childcare centers are areas of intensive use for children and when coupled with potential residential exposure in their homes, the total daily exposure is a potential hazard to children.

## 5.1 Introduction

### 5.1.1 Historic lead arsenate use

The use of lead arsenate (LA) pesticides on fruit orchards in the United States began in the early 1900s and continued until the 1950s or 1960s, when chlorinated pesticides, including DDT, became available as a more preferred option (Peryea, 1998). LA was more commonly used on orchards rather than other agricultural fields because of its high efficacy at reducing codling moths, which are common pests of apple trees (Veneman et al., 1983). LA was the most popular arsenical insecticide and was used worldwide in two forms, basic LA and acidic LA (Peryea, 1998). As insect populations developed a resistance to arsenic (As), application dosages and frequencies increased, as did soil contamination levels (Peryea, 1998). Application amounts were dependent upon species and persistence of insect populations, species of fruit tree, time of year, and use of any alternative pesticides (Peryea, 1998). Subsequently, many orchard lands were converted to residential or public use areas. This study evaluated the potential for exposure to lead (Pb) and As, especially for children on lands converted from orchards to residential and public use (see Figure 1).

This research focused on historic LA use in the Upper Yakima Valley, Washington State, but the compound was used throughout Central Washington, the United States, and globally. In New York, soils historically used for apple orchards were found to contain As concentrations ranging from 31 to 109 mg/kg (average = 72 mg/kg) and Pb concentrations ranging from 171 to 512 mg/kg (average = 339 mg/kg) (Aten et al., 1980). These orchards were sprayed with LA until 1965, usually at a rate of two (2) applications per year totaling 1010 kg of pesticide in a 12-year period (Aten et al., 1980). Studies of former orchard sites in Massachusetts confirmed elevated Pb and As concentrations as a result of LA use (Veneman et al., 1983). Orchard soils in southern Ontario, Canada were found to have average soil concentrations of 774 mg/kg and 121 mg/kg Pb and As, respectively (Frank et al., 1976a). Pb and As concentrations tend to be highest in former apple orchards, followed by cherry orchards and peach orchards (Frank et al., 1976a). Former orchard lands in Tasmania and Australia were examined for heavy metal concentrations and found to have levels of copper, Pb, and As 25e35 times the levels in comparable agricultural soils (Merry et al., 1983).

The association between Pb and As levels are strongly correlated in most LA studies, though some authors have found decreased levels of As, suggesting that As undergoes more leaching than Pb (Merry et al., 1983; Veneman et al., 1983; Peryea, 1998). As, also capable of biomethylation, is more environmentally mobile than Pb, hence Pb/As concentration ratios tend to decrease with increasing depth (Peryea, 1998). Frank et al. (1976a) found that not only does land use correlate with soil concentrations of Pb and As, but soil type is also a factor (Frank et al., 1976b). Loam and clay soils had higher concentrations of As than sandy or organic soils, and higher Pb concentrations were found in loam or sandy soils than clay soils (Frank et al., 1976b).

### 5.1.2 Exposure risks

The principal exposure route for children living and/or playing in LA contaminated soils is the incidental ingestion of soils and dusts. Indoor dusts may have elevated contaminant concentrations due to tracking in from outdoor locations. In the same way that exposure to soils occurs outdoors, exposure to contaminated dusts occurs indoors. Dust inhalation risks and therefore exposure risk is increased for young children that are not yet walking in comparison to children that are walking (Hunt and Johnson, 2012). Wolz et al. (2003) were the first to study household dust concentrations in Washington homes built on former orchard land contaminated with LA. They found that As and Pb concentrations of indoor dusts were correlated to soil loading levels, and that these indoor concentrations were elevated compared to areas without a history of LA application. Further analysis of the data indicated that intakes of contaminated dusts resulted in an exceedance of Agency for Toxic Substances & Disease Registry's (ATSDR) minimum risk level for As, 0.3 µg per kilogram per day. Soil concentrations ranged from <2.5 to 103 mg/kg and 1.2-594 mg/kg, for As and Pb, respectively, and house dust concentrations ranged from 2.3 to 49 mg/kg and 15-890 mg/kg for As and Pb, respectively. The mean house dust loadings ranged from 0.8 to 495 µg/$m^2$ and 14-5970 µg/$m^2$ for As and Pb respectively. Thirteen (13) of the 58 homes included in the study exceeded the Washington State Model Toxics Control Act (MTCA) Method A soil cleanup levels (regulatory standard) for unrestricted land uses (MTCA cleanup level), 250 mg/kg Pb and

20 mg/kg As (Wolz et al., 2003). Atmospheric soil As and Pb levels have been found to be the highest in the summer and fall when soil is dry. These season patterns appear to match blood Pb level patterns in children (Laidlaw et al., 2011).

The health impacts of exposures to Pb and As are well documented. Early childhood Pb exposure is directly linked to neurocognitive deficits and social problems in adulthood (ATSDR, 2007; Lanphear et al., 2005; Needleman et al., 1990). An estimated one to five IQ points are lost for each increase of 10 µg/dL in blood Pb level (BLL) (Hubbs-Tait et al., 2005). Centers for Disease Control and Prevention confirm that there is no safe threshold for Pb in blood (Centers for Disease Control and Prevention, 2012). As, in addition to being a developmental neurotoxin, is a known human carcinogen in its inorganic form. The overlap of toxic endpoints of the two metals indicates a potential for interaction in co-exposure settings, increasing risk for children exposed to LA contaminated soils and dusts. Limited studies exist examining toxicities resulting from simultaneous exposure, but some indicate synergistic interactions at low doses (Bae et al., 2001; Mejia et al., 1997).

Unfortunately, there is limited data to determine if exposures are resulting in direct health impacts in Central Washington residents (Hood, 2006). The Washington State Department of Health (DOH) conducted a study which included assessment of blood Pb levels for children throughout Washington. A total of 50,000 tests were conducted, 2% of which had blood levels above 10 µg/dL, which was the level of concern at the time of the study. The Centers for Disease Control and

Prevention abandoned the term “blood lead level of concern” for a reference value that is currently 5 µg/dL, and is based on the 97.5th percentile of the National Health and Nutrition Examination Survey (NHANES)-generated BLL distribution in children 1-5 years old (Centers for Disease Control and Prevention, 2012). The DOH identifies older homes, lower household incomes, Hispanic ethnicity, and Central Washington residency to be significant predictors of elevated BLLs (Community, Trade, and Economic Development, 2005).

**5.1.3 LA use and regulation in Washington State**

The economy of Central Washington is largely agriculturally based. Yakima County has been the largest producer of apples in the United States since 1964. Jobs in agriculture, forestry, and fishing account for more than 26% of jobs in Yakima County (Yakima County Development Association, 2013). Recently, as populations increased in Central Washington, homes, schools, and businesses were built on land that previously was orchard.

In Washington, LA was used from 1905 to 1947 (Wolz et al., 2003). On a per hectare basis, application rates reached 215 kg/year of Pb and 80 kg/year of As during this time period (Davenport and Peryea, 1991). Up to 76,081 ha of land in Washington may be affected (Hood, 2006). In 2002, an Area-Wide Task Force was assigned to investigate the Pb and As contamination in Washington and recommend actions to reduce the health risk of residents. The recommendations released in 2005 included prioritization of education and outreach for residents, use of soil covers, mandatory soil testing for new child use areas, and voluntary certification programs for childcare centers (CCs). Specifically mentioned in the recommendations was their concern for children's exposure to contaminated soils (Ecology, 2003).

Following these recommendations in 2005, the first version of Washington State House Bill 1605 directed state and local agencies to assist schools and CCs throughout Washington in areas impacted by area-wide contamination to reduce the exposure of children to contaminated soil. However, the version of the House Bill that was signed into law did not include a provision for the area-wide contamination east of the Cascades in Central Washington (areas in Central Washington that contain formerly orchard land primarily include the Yakima Valley, areas in and around the City of Wenatchee, and surrounding areas). Instead, the amended House Bill included enhancements on efforts for Western Washington, where the Pb and As contamination originated primarily from smelters (Figure 2). The law enlisted the technical and financial help of state and local agencies to assist schools and childcare facilities to reduce the exposure potential for children (Engrossed Second Substitute House Bill 1605 2005).

After House Bill 1605 was signed into law in April 2005, remediation efforts began in both Central and Western Washington. Extensive mapping and remediation of the contamination began in Western Washington, including schools, CCs, parks, and qualified residential properties. In Central Washington, 26 public schools and two (2) parks were remediated (Ecology, 2015). “During cleanup activities in Central Washington, maximum concentration levels for Pb and As were found to be 1650 mg/kg and 1100 mg/kg, respectively. The highest concentrations were found in the Manson and Wenatchee areas of Central Washington. Levels of Pb and As in the City of Yakima are also well above the MTCA standards, averaging 1080 mg/kg and 124 mg/kg, respectively” (N. Hepner, personal communication). The Washington State MTCA regulatory standard for soil Pb in unrestricted land use areas is 250 mg/kg, significantly lower than the U.S. Environmental Protection Agency (EPA) standard of 400 mg/kg. The MTCA standard for As is currently 20 mg/kg, however it is important to note that 20 mg/kg is greater than background levels in Washington, which have been found not to exceed 12.8 mg/kg. The average soil background level for As in Washington is 7 mg/kg (Ecology, 1994).

**5.2 Objectives**

The objective of this study was to identify As and Pb soil and dust concentrations at participating CCs in the Upper Yakima Valley in Washington. Children at these sites may be exposed to Pb and As in the soils during daily play activities. Schools in the affected area Figure 1. have been previously been tested and remediated as needed, but no data exists for childcare facilities and no testing program has been provided by government or private organizations. Results of this study were used to educate the owners/operators of the voluntarily participating childcare facilities on the levels of Pb and As found in their soil and indoor dust. Educational materials provided included information on the health risks of Pb and As exposure for themselves, their employees, and the children in their care. The immediate aim of this study is to raise awareness for facility owners. More broadly, the goal is also to increase the level of awareness concerning the contamination throughout the affected area.

## 5.3 Methods

### 5.3.1 Childcare center selection

Prior to sampling, the project and sampling plan were approved through the University of Idaho's Institutional Review Board (IRB). Included in the IRB application and sampling plan were provisions to guarantee confidentiality of CCs sampling results. The participating and non-participating CCs were kept confidential, and all results were de-identified in any reports and presentations.

Nineteen (19) CCs volunteered for and were included in the study from Yakima and surrounding communities. Thirteen (13) of the CCs were located in the City of Yakima and immediate vicinity. The other six (6) CCs were located in the nearby communities of East Valley, Terrace Heights, Union Gap, Selah, and Tieton. All of the licensed CCs (as of 2014) are shown in Figure 1. The yellow shading on Figure 1 represents the land thought to previously be orchard land, determined from analysis of historical aerial photographs. CCs that volunteered for the study were recruited at a monthly meeting of a community group of childcare providers in the Yakima area and individual visits to CCs. Nearly 200 childcare providers were contacted with educational materials and offered the opportunity to be included in this study. The majority of those contacted were at home providers. CCs were a priority for recruitment due to the high number of children served at these facilities. Each CC was provided and asked to complete a survey regarding site history, children's daily activities, cleaning frequency, and types of soil barriers in place in play areas.

### 5.3.2 Sample collection

Soil sampling methodology was based on the strategy employed by Washington State Department of Ecology (Ecology) during their sampling of area schools in 2002 (Ecology, 2011). Prior to sampling CCs were surveyed about their facilities and grounds, construction/ landscaping history, operations, child ages, interior cleaning frequency, and outside play activity. Each CC's outdoor play area was divided into exposure units (EU), which are discreet areas that children access daily. The number of samples taken in each EU was determined by total EU area (Table 1). EUs were drawn on aerial images of the CC and sampling locations were noted. Soil samples were collected from a freshly dug pit wall at 0-5 cm and 5-15 cm depths following removal of surface barriers (e.g. bark chips or sod). Organic material, gravel, woodchips, and other non-soil particles were avoided. Samples were placed in self-sealing polyethylene bags, labeled, and refrigerated.

Indoor dust wipe samples were collected from at least one windowsill and one floor surface following Housing and Urban Development (HUD) guidelines "Wipe Sampling of Settled Dust for Lead Determination" (U.S. Department of Housing and Urban Development, 2012). Disposable wipes meeting standards outlined in 40 CFR 745.63 were used (500" × 7.75", lanolin free, professional use Lead Dust Sampling Wipe, meeting ASTM E1792 standards). Samples were stored in hard-shell, non-sterilized, resealable containers. Samplers were trained in the appropriate methodology prior to sampling. Areas of high use were targeted, especially areas where children frequently placed their hands. One floor wipe sample and one windowsill wipe sample were taken unless a windowsill sample was not feasible/available, in which case two floor wipe samples were taken. The template used was reusable plastic and met HUD and EPA requirements. If a template could not be placed on the sample area (e.g., on the windowsill), the area to be sampled was delineated with masking tape and carefully measured.

### 5.3.3 Soil sample analyses

Refrigerated samples were transported to the University of Idaho (UI) Environmental Toxicology and Chemistry Laboratory for analysis. Soil samples were air dried and homogenized. A Niton XL3 Handheld X-ray fluorescence spectrometry (XRF) Analyzer was used to analyze soil samples for heavy metal content. After performing instrument calibration checks and testing standard reference materials, soil samples were tested 3 times for 30 s each test. Results were imported into Microsoft Excel® and subsequently into SAS®. Samples with Pb > 250 mg/kg and As >20 mg/kg were sieved to 250 µm and tested again using the same methodology. All sieved samples that tested for Pb > 250 mg/kg and/or As >20 mg/kg in either the sieved or bulk fractions were analyzed for As and Pb via inductively coupled mass spectrometry (ICP-MS) at the UI Analytical Sciences Laboratory.

### 5.3.4 Laboratory analyses

Additional laboratory analyses were performed by the UI Analytical Sciences Laboratory on samples with Pb and As XRF detections over MCTA cleanup levels. Soil and interior dust wipes were analyzed using EPA Standard Methods (SM) 3050 and 200.7 (ICP-MS).

### 5.3.5 Statistical analyses

XRF and ICP-MS results were imported into SAS® (Version 9.3). Average Pb and As XRF concentrations were calculated for each sample and used for further analyses as the standard deviations were lower than limits of detection (LOD). Linear regression was run to compare bulk As to sieved As, bulk Pb to sieved Pb, Pb to As in both bulk and sieved fractions, and XRF to ICP-MS results in bulk and sieved fractions for both As and Pb. Pb and As were also plotted against each other in the bulk fractions to graphically assess associations. Regression was also performed on bulk As and Pb results by site history and sample depth using method of least squares to fit general linear models.

**5.3.6 GIS analyses**

Using aerial photographs from 1927 to 1947 of the Yakima Valley and surrounding areas, former orchard land was digitized and displayed on an ESRI GIS map. Licensed CC locations and in home CCs locations were plotted on the map. CCs that were sampled were not distinguished from those that were not sampled in order to protect identity of participating CC locations (Figure 1).

Two additional maps (Figures 2 and 3) were created plotting the maximum soil concentrations for Pb and As at participating CCs. Also indicated on the map were maximum Pb and As concentrations from the school remediation projects conducted by Ecology. This data was acquired from the Ecology Environmental Information Management Database (EIM). Using the soil concentration values, the ESRI Inverse Distance Weighting (IDW) geostatistical Contours are estimates and may not accurately predict the soil contamination at any given point. Pb and As concentrations can vary significantly, even within a particular location (Figures 3 and 4).

**5.4 Results**

**5.4.1 Statistical analyses**

Based on maximum Pb and As results for discrete sample locations, 21% and 42% of the 19 CCs surveyed exceeded the MTCA Method A soil cleanup levels for unrestricted land use for Pb and As, respectively (see Table 2). Correlations between bulk and sieved Pb and bulk and sieved As results show a strong positive linear correlation (r = 0.99 and 0.93, respectively; Figures 5 and 6). Comparison of Pb to As in the bulk fraction demonstrate a weak linear correlation (slope = 0.12, r = 0.61). Statistically significant differences between average metal values at non-orchard and former orchard lands were found (see Figure 7). There was no significant difference in either As or Pb by sample depth.

For Pb, ICP-MS compared well to XRF in both bulk (slope = 0.97, $R^2$ = 0.97) and sieved (slope = 0.90, $R^2$ = 0.99) fractions. The relationship was slightly less favorable for As, with bulk (slope = 0.78, $R^2$ = 0.89) and sieved (slope = 0.79, $R^2$ = 0.91) slopes further from 1 (data not shown).

Dust loading of wipe samples ranged from 0.4 to 6.3 µg/ft$^2$ for Pb, well below the EPA limits of 40 µg/ft$^2$ and 250 µg/ft2 for floors and windows, respectively. As levels were below the limit of detection (1.5 µg/ft$^2$) for all wipe samples.

**5.4.2 GIS analyses**

The extent of former orchard land in the Upper Yakima Valley is expansive, encompassing much of the area that is now residential. Using the 1927 and 1947 aerial photographs to define former orchard land 8 out of the 11 CCs that were sampled were on former orchard land, and 10 out of the 13 CCs that were not sampled were on former orchard land. A total 18 of 42 CCs (43%) were in former orchard land (Figure1). There were 300 in-home CCs total,116 of those were in former orchard land, 39% of the total (Figure 1). Contours generated using the ESRI IDW geostatistical tool and maximum soil concentrations for Pb and As indicate a relationship between former orchard lands and elevated concentration values (Figures 3 and 4). Areas that show elevated As values tend to also show elevated Pb values (Figures 3 and 4).

**5.4.3 Survey results**

Fifteen (15) of 19 CCs filled out a survey, though some fields were omitted. Average CC age was 11.7 years (SD ± 6), the number of children on site ranged from 5 to 400 (average 83.9, SD ± 93), and children's ages ranged from 1 month to 16 years. Time outdoors in summer and winter averaged 108.5 (SD ± 64) and 52 (SD ± 24) minutes, respectively. Total time at the CCs each day averaged 502 (SD ± 78) minutes. Cleaning frequencies were reported for wet mopping, dusting, and vacuuming at 9.5 (SD ± 4), 3.7 (SD ± 4), and 6.2 (SD ± 2) times per week, respectively. Nine (9) CCs reported no new soil introduction, 11 use sprinkler systems in the play area, nine (9) use some type of soil barrier or cover such as mulch or gravel, 13 reported no interior or exterior lead-based paint, six (6) reported some type of previous excavation had been completed (often for the initial CC construction), and seven (7) reported interior remodeling within the last 22 years or less. When asked if they were aware of the possibility of Pb and As soil contamination in the area none of the respondents had heard of the issue. Due to the low number of complete surveys (questions left blank or surveys not returned at all), regression analysis of questionnaire results against soil metal concentrations was not possible. Complete summary of survey information in Table 3a and b.

**5.5 Discussion**

The strong correlation between former land use and Pb and As levels in regression analyses supports the hypothesis that most of the Pb and As in the soils is a result of historic LA pesticide application. Sample depth does not appear to play an important role, perhaps due in part to leaching of As through the soil column over time. This association between land use history and Pb and As concentrations could be used in future education efforts in the region.

While the correlation between bulk and sieved results demonstrate strong correlation for both metals, the correlation between the two metals is not as strong, indicating potential differences in contaminant fate and transport mechanisms. This is in line with findings from other authors (Merry et al., 1983; Veneman et al., 1983; Peryea, 1998). Site history is the strongest indicator of contamination in linear regression analysis.

Several options exist to reduce exposures for Upper Yakima Valley residents. Some environmental Pb remediation methodologies promote the use of phosphorus to bind to Pb in the soil and make it less bioavailable to human and animal digestion (Peryea, 1998). However, As has been shown to become more mobile in Pb contaminated soils remediated with phosphorus, increasing the risk of both surface and groundwater contamination (Peryea, 1998; Davenport and Peryea, 1991). The addition of phosphate-based fertilizers has the same effect on As mobility (Davenport and Peryea, 1991).

Physical removal of contaminated soil is a common mitigation measure in many areas, including remediation of schools in Central Washington (Peryea, 1998). Capping of soils (encapsulation) is another option for reducing exposure risks (Peryea, 1998), though this involves long-term maintenance and control to keep the barrier intact. Mixing contaminated soils with “clean” soils is an additional option, one that is more viable with lower contaminant levels (Hood, 2006).

Reduction of exposure can also be achieved by reducing children's contact with contaminated soils and dust, where possible. The majority of CCs employed some degree of bark cover, four had bark covering the majority of the play area. Eleven (11) CCs reported watering lawns via a sprinkler system, improving grass cover. Given the contamination concentrations, these factors reduce the risk of soil exposure to children at those locations, a point which was discussed with facility owners. The lack of elevated Pb and As in indoor dusts may be explained by the frequent cleaning reported by facilities on the questionnaires.

It is possible that the recruitment method resulted in a sample bias towards facilities with good hygiene practices and grounds maintenance; i.e., facilities with less stringent cleaning regimens and without grass cover or established play areas may have been more reluctant to participate in the study. This could result in the data here biasing towards CCs with lower exposure risks for children.

There was also a sample bias towards the larger CCs verses in home CCs. All of the ~200 licensed CCs in the Upper Yakima Valley (as of 2014) were contacted by email to participate in the study. Only one in-home CC out of ~140 replied and agreed to participate. Most of the larger (non-home) CCs and school CCs were visited on site to recruit participation. Eighteen (18) out of the 32 larger/ school CCs agreed to allow sampling.

Some areas that have been converted from orchard use do not appear to have elevated levels of As and Pb. This could be the result of inaccuracies in the historic maps, landscaping/movement of soils, weathering, construction activities, and different soil tilling and LA application patterns during agricultural use.

**5.6 Conclusions**

The MTCA Method A soil cleanup levels for unrestricted land use for Pb and As are exceeded in some of the CCs participating in this study. The MTCA cleanup levels for Pb and As are regularly exceeded in soil samples analyzed in this study, consistent with previous observations of widespread contamination of former orchard lands in the Yakima Valley (Ecology, 2015). Pb and As have a common pathology in neurotoxicity, and thus are of particular concern in children's environmental health. CCs are areas of intensive use for children and when coupled with potential residential exposure in their homes, the total daily exposure suggests a hazard to children. Heavy metals in dusts are a major exposure risk to infants and children who exhibit greater hand to mouth activity (U.S. Environmental Protection Agency, 2013). All responding CC facility operators surveyed in this study were unaware of the LA contamination in the Yakima Valley. Previous remediation and assessment activities in the region targeted schools, but did not address other areas where children spend significant amounts of time outdoors. This study suggests the need for further review and action in addressing targeted public outreach and education and in reducing children's exposure to legacy LA in impacted areas. Childcare centers are areas of intensive use for children and when coupled with potential residential exposure in their homes, the total daily exposure is a potential hazard to children.

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Table 5.1. Number of samples taken per Exposure Unit (EU), dependent upon area size and intensity of use by children (Ecology, 2002).

| Exposure Unit Size ($m^2$) | Low Use | Casual Use | Intensive Use |
|---|---|---|---|
| 0 – 400 | 0 | 4 | 8 |
| 400 – 4,000 | 0 | 6 | 10 |
| 4,000 – 28,000 | 0 | 6 – 12[1] | N/A[2] |

[1]Six samples for the first 4,000 $m^2$, one additional sample for each additional 4,000 $m^2$
[2]Not applicable, intensive use areas assumed to be <4,000 $m^2$.

Table 5.2. Count of soil sample XRF results exceeding MTCA cleanup levels for Pb and As.

| | | Bulk | | Sieved | |
|---|---|---|---|---|---|
| | Total Count | Pb >250 mg/kg | As >20 mg/kg | Pb >250 mg/kg | As >20 mg/kg |
| **No. CC Sites** | 19 | 4 | 8 | 4 | 8 |
| **No. Sample Locations** | 187 | 13 | 55 | 13 | 49 |
| **No. Samples** | 356 | 18 | 90 | 19 | 73 |

Table 5.3a. Childcare Center (CC) site survey summary – demographics and cleaning information.

| Survey Question | # Responses | Range | Average | Stdv | Max | Min |
|---|---|---|---|---|---|---|
| Site Age (Years) | 15 | 24 | 11.7 | 6 | 26 | 2 |
| Years Since Remodeling | 3 | 19 | 14.3 | 10 | 22 | 3 |
| Child Count | 15 | 394.5 | 83.9 | 93 | 400 | 5.5 |
| Age Range of Children | 15 | 30 days to 16 years | | | | |
| Minutes Outside, Summer | 15 | 218 | 108.5 | 64 | 240 | 22 |
| Minutes Outside, Winter | 14 | 98 | 52.6 | 24 | 210 | 22 |
| Total Indoor Play (Minutes) | 15 | 683 | 377.6 | 177 | 720 | 37 |
| Total Time at Center (Minutes) | 15 | 270 | 502 | 78 | 600 | 330 |
| Days Per Week at Center | 14 | 1.5 | 4.8 | 1 | 5 | 3.5 |
| Wet Mop Freq (Times/ Week) | 15 | 11 | 9.5 | 4 | 14 | 3 |
| Dusting Freq (Times/ Week) | 13 | 13.75 | 3.7 | 4 | 14 | 0.25 |
| Vacuum Freq (Times/ Week) | 14 | 6 | 6.2 | 2 | 7 | 1 |

Table 5.3b. CC site survey summary – facility information.

| Survey Question | # Responses | Yes | No |
|---|---|---|---|
| Any Soil Introduced | 11 | 1 | 10 |
| Sprinkler System in Play Area | 13 | 11 | 2 |
| Soil Cover Used | 14 | 9 | 5 |
| Enviro. Testing Done | 11 | 1 | 10 |
| Presence of Lead Paint | 13 | 0 | 13 |
| Excavation on Site | 8 | 4 | 4 |
| Remodeling Done | 14 | 9 | 5 |
| Aware of Pb-As Issue | 8 | 0 | 8 |

Figure 5.1. Map of overlapping Childcare Centers and former orchard lands in the Upper Yakima Valley, as defined by digitizing 1927 and 1947 aerial photographs.

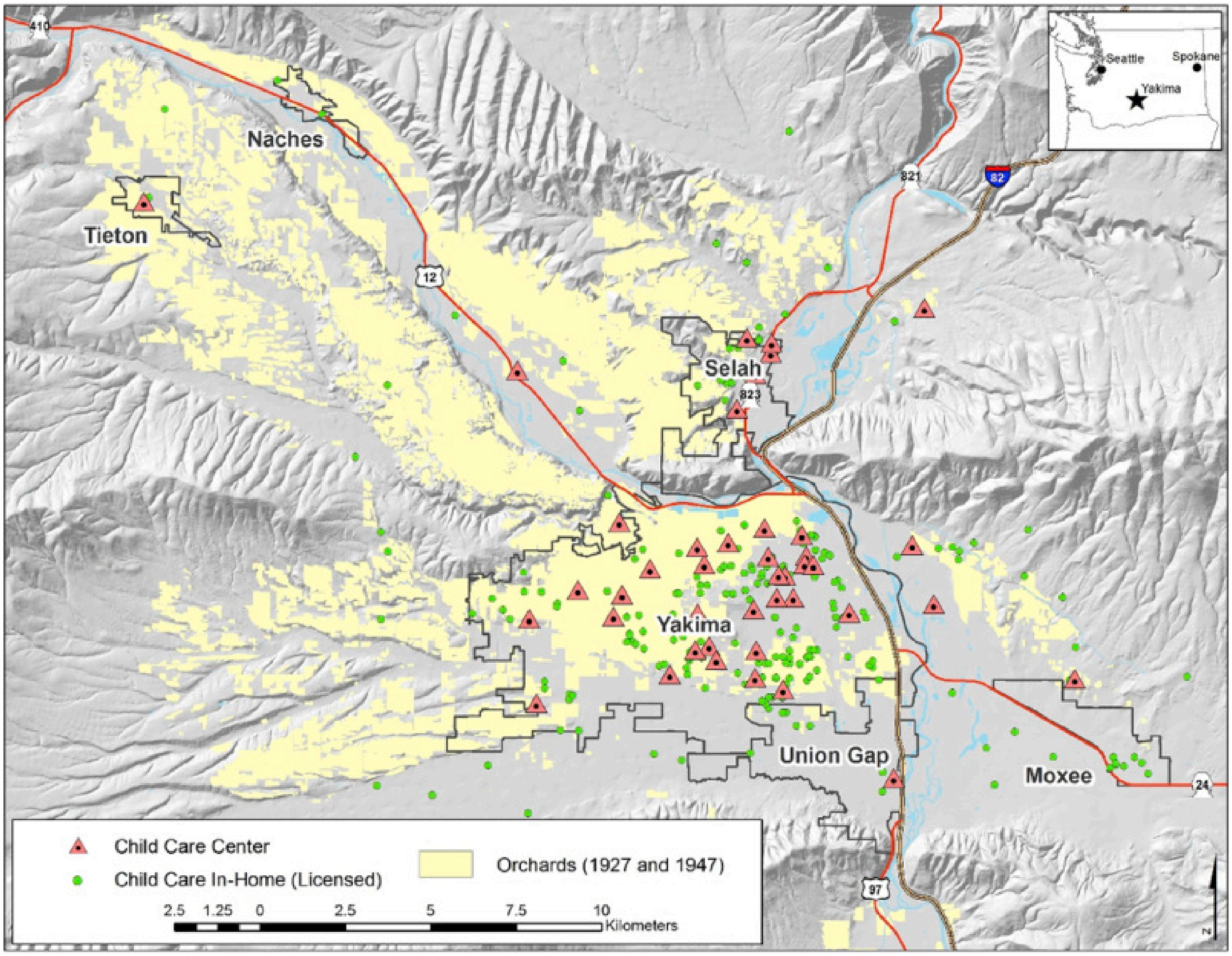


Figure 5.2. Map of Washington locating smelter and study area.

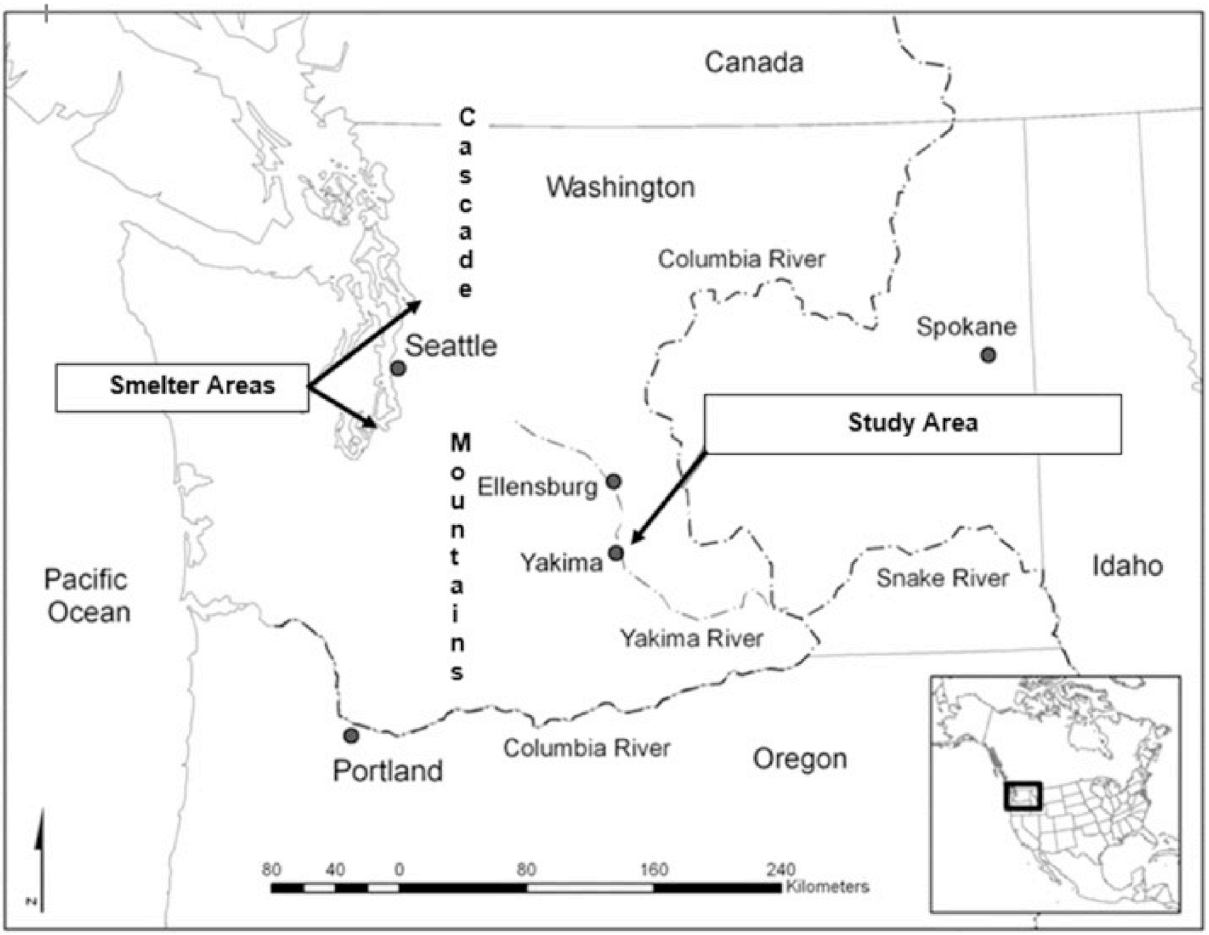


Figure 5.3. Inverse distance weighted contour maps of maximum Pb soil levels from sampled Childcare Centers combined with Washington State Department of Ecology data from regional schools surveyed before site remediation.

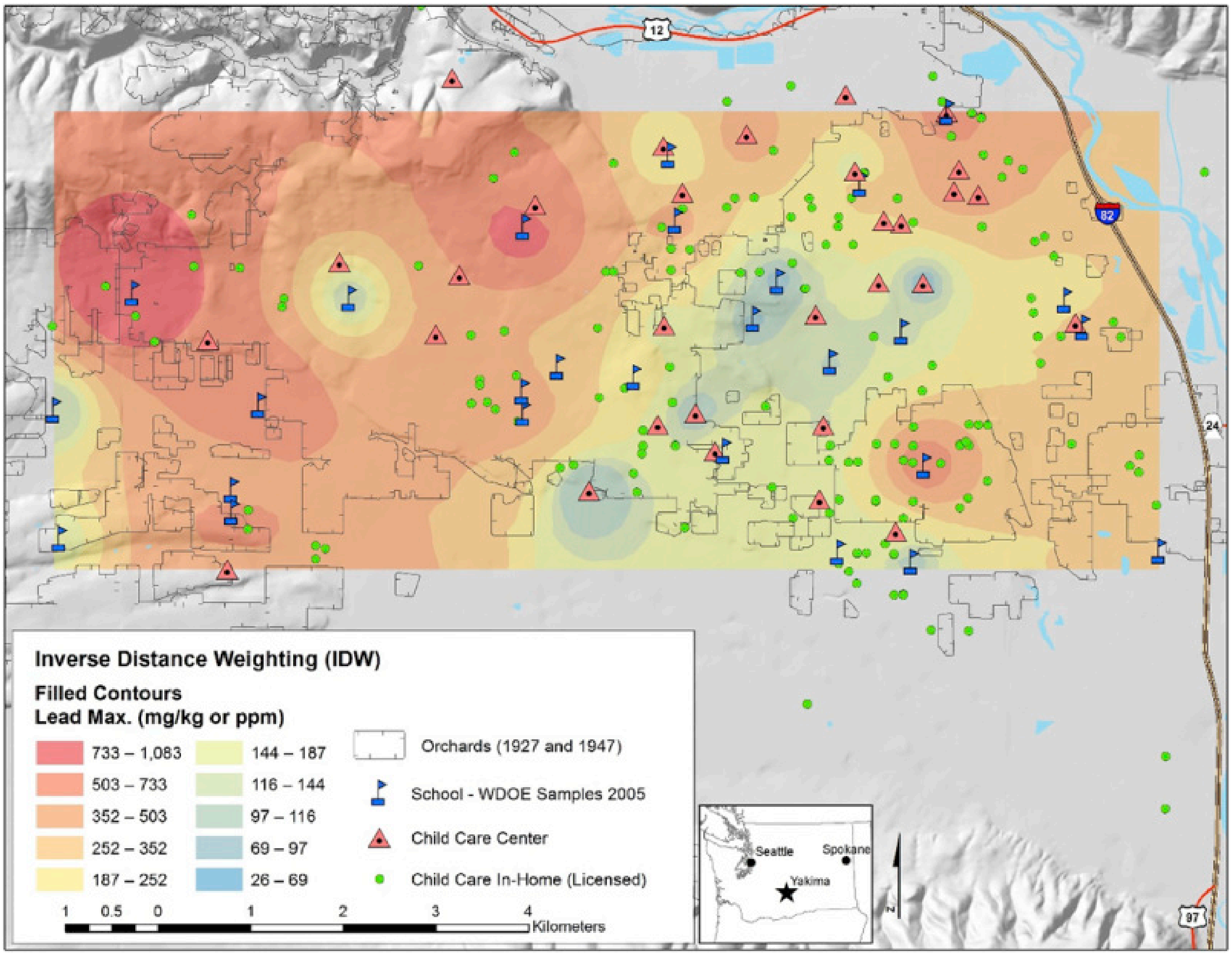


Figure 5.4. Inverse distance weighted contour maps of maximum As soil levels from sampled Childcare Centers combined with Washington State Department of Ecology data from regional schools surveyed before site remediation.

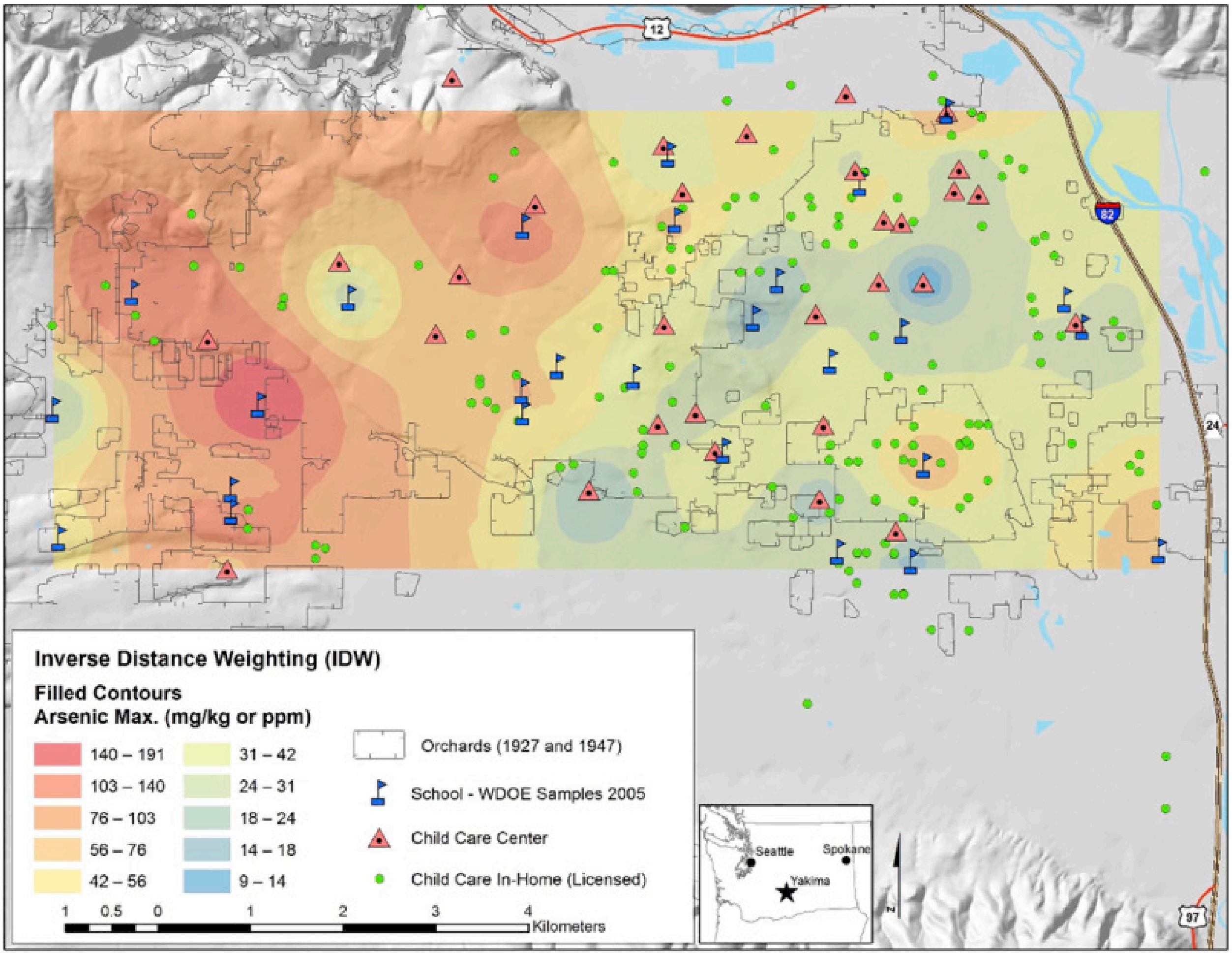


Figure 5.5. XRF results of Pb in bulk and 250 µm sieved fractions (mg/kg).

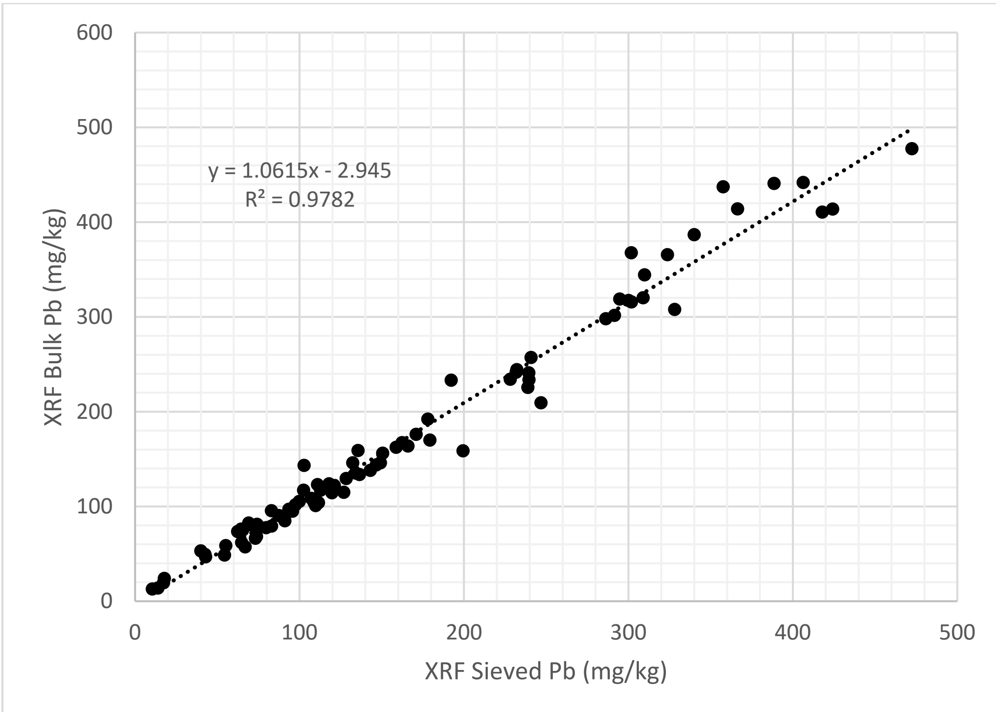


Figure 5.6. XRF results of As in bulk and 250 µm sieved fractions (mg/kg).

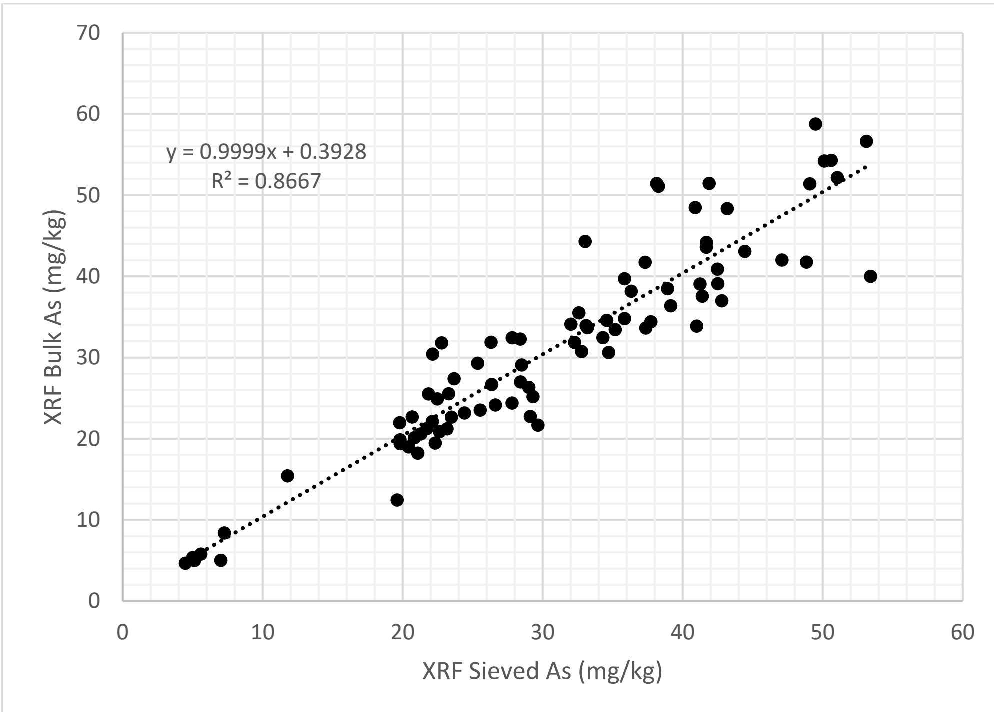
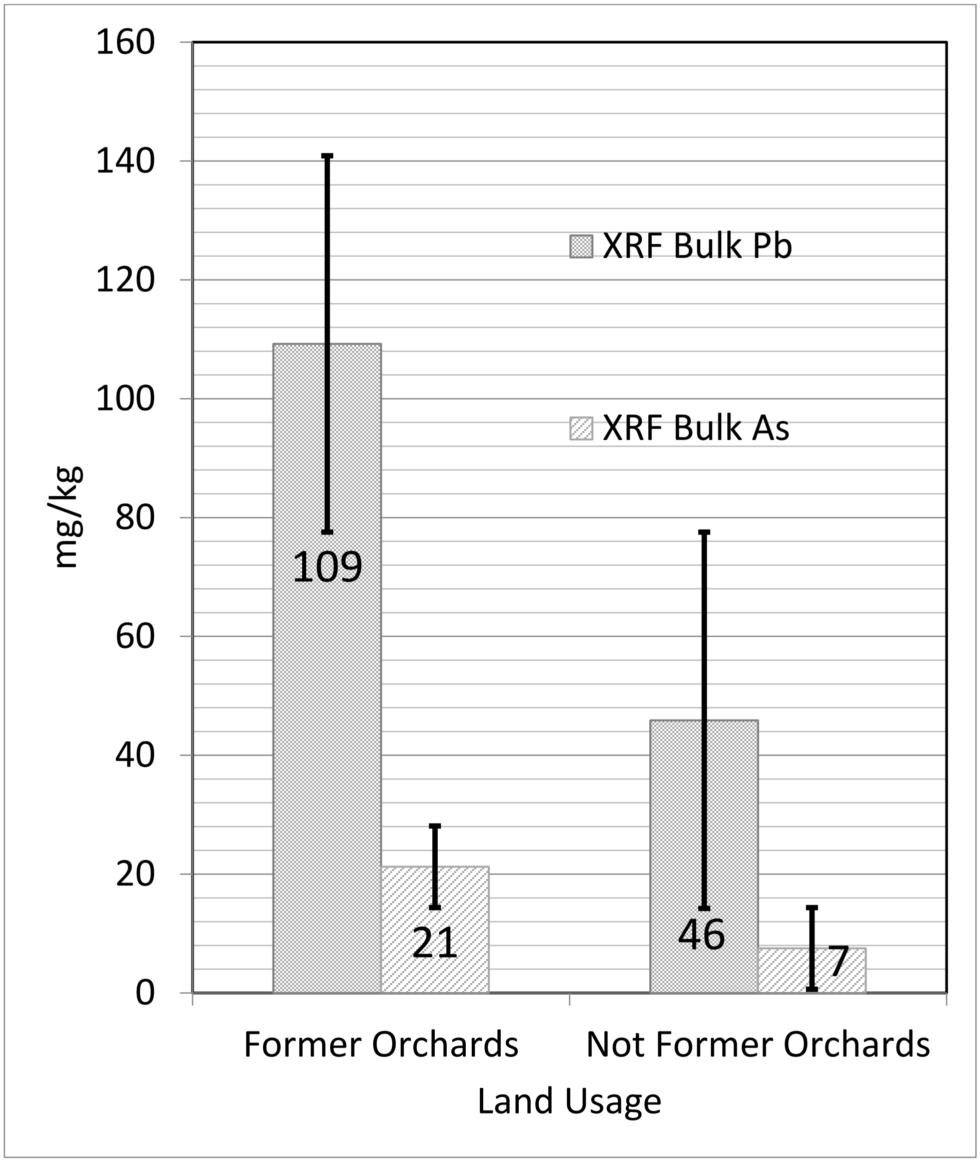


Figure 5.7. Average Pb and As results by historic land use, determined by review of aerial photographs from 1927 to 1947.



APPENDIX A

University *of* Idaho

**Office of Research Assurances**
**Institutional Review Board**
875 Perimeter Drive, MS 3010
Moscow ID 83844-3010
Phone: 208-885-6162
Fax: 208-885-5752
irb@uidaho.edu

To: Gregory Moller

cc: Margrit von Braun, Casey Lyn Bartrem

From: Jennifer Walker, IRB Coordinator

Approval Date: January 03, 201

Title: Characterization of the Blood Lead Absorption / Environmental Lead Exposure Relationship among Children Provided Chelation Treatment in the Lead Poisoning Epidemic in Zamfara, Northern Nigeria

Project: 16-146

Certified: Certified as exempt under category 4 at 45 CFR 46.101(b)(4).

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On behalf of the Institutional Review Board at the University of Idaho, I am pleased to inform you that the protocol for the research project Characterization of the Blood Lead Absorption / Environmental Lead Exposure Relationship among Children Provided Chelation Treatment in the Lead Poisoning Epidemic in Zamfara, Northern Nigeria has been certified as exempt under the category and reference number listed above.

This certification is valid only for the study protocol as it was submitted. Studies certified as Exempt are not subject to continuing review and this certification does not expire. However, if changes are made to the study protocol, you must submit the changes through VERAS for review before implementing the changes. Amendments may include but are not limited to, changes in study population, study personnel, study instruments, consent documents, recruitment materials, sites of research, etc. If you have any additional questions, please contact me through the VERAS messaging system by clicking the 'Reply' button.

As Principal Investigator, you are responsible for ensuring compliance with all applicable FERPA regulations, University of Idaho policies, state and federal regulations. Every effort should be made to ensure that the project is conducted in a manner consistent with the three fundamental principles identified in the Belmont Report: respect for persons; beneficence; and justice. The Principal Investigator is responsible for ensuring that all study personnel have completed the online human subjects training requirement.

You are required to timely notify the IRB if any unanticipated or adverse events occur during the study, if you experience and increased risk to the participants, or if you have participants withdraw or register complaints about the study.

APPENDIX B

**National Health Research Ethics Committee of Nigeria (NHREC)**

Promoting Highest Ethical and Scientific Standards for Health Research in Nigeria

Federal Ministry of Health

**NHREC Protocol Number:** NHREC/01/01/2007-26-05-2016 **NHREC Approval Number:** NHREC/01/01/2007-10/06/2016 **Date:** 12th June, 2016

**Re: Characterization of the Blood Lead Absorption / Environmental Lead Exposure Relationship among Children Provided Chelation Treatment in the Artisanal Lead Poisoning Epidemic in Zamfara, Northern Nigeria, 2010-2017**

Health Research Committee assigned number: NHREC/01/01/2007

Name of Principal Investigator: Dr. Ian von Lindern, P.E

Name of Co-Investigator: Dr. Nasir Sani-Gwarzo

Address of Co-Investigator: Port Health Services,
Federal Ministry of Health, Abuja
drgwarzo@gmail.com

Date of receipt of valid application: 26/05/2016
Date when final determination of research was made: 10/06/2016

**Notice of Research Exemption**

This is to inform you that the research described in the submitted protocol/documents have been reviewed and the Health Research Ethics Committee has determined that according to the National Code for Health Research Ethics, the activity described there-in meets the criteria for exemption and is therefore approved as exempt from NHREC oversight.

The National Code for Health Research Ethics requires you to comply with all institutional guidelines, rules and regulations and with the tenets of the Code. The HREC reserves the right to conduct compliance visit your research site without previous notification.

Signed

Clement Adebamowo BMChB Hons (Jos), FWACS, FACS, DSc (Harvard) Chairman, National Health Research Ethics Committee of Nigeria (NHREC)

Department of Health Planning, Research & Statistics
Federal Ministry of Health
11th Floor, Federal Secretariat Complex Phase III
Ahmadu Bello Way, Abuja

Tel: +234-09-523-8367
E-mail: chairman@nhrec.net, secretary@nhrec.net, deskofficer@nhrec.net,
URL: http://www.nhrec.net,

APPENDIX C

# Data Sharing Agreement

between

## Médecins Sans Frontières - Holland

and

TerraGraphics International Foundation – Idaho, USA

(TIFO)

This agreement between MSF-Holland (disclosing party) and TerraGraphics International Foundation (recipient party) is entered into on July 28 2014.

**Disclosing party:** **MSF-Holland (hereinafter referred to as "MSF")**

**Name of contact person: Sidney Wong, Medical Director**

Plantage Middenlaan 14

1018 DD
Amsterdam The
Netherlands

**Recipient party:** TerraGraphics International Foundation

220 E 5th St #8866

Moscow, Idaho US 83843

**Name of contact person:**

Ian H. von
Lindern
Executive
Director

(ian.vonlindern@terragraphics.com)

**Description/ name of data set: Summary of initial blood lead levels for children tested prior to treatment in the villages of Zamfara, Northern Nigeria during the artisanal mining lead poisoning epidemic from 2010-24.**

The data that is shared on the basis of this agreement has been collected by MSF as routine programmatic data.

MSF is the full owner of any data related to said study/ program.

**Purpose of use:** Characterization of the Blood Lead Absorption / Environmental Lead Exposure Relationship among Children Provided Chelation Treatment in the Lead Poisoning Epidemic in Zamfara, Northern Nigeria

**Method of transferring data: Data will be transmitted electronically from MSF to TIFO.** All data will be transferred in an encrypted format with the encryption key stored separate from the dataset.

Each party agrees to maintain this dataset either on the supplied CD or on a secure server.

The purpose of this Agreement is to set forth the terms, conditions, and obligations concerning the sharing of data between the parties.

Therefore, MSF and TIFO agree to share the aforementioned data under the following **conditions:**

1. MSF acknowledges that the dataset transferred may contain person identifiable data.
2. Both parties agree to maintain confidentiality and privacy safeguards that were originally created as part of the protocol or data collection. Both parties agree not to release information about specific identifiable subjects to anyone. Only people mentioned in the protocol will have access to the database.
3. During handling and storage of the data, all parties agree to abide by universally recognised ethical principles. In particular, due consideration will be given to issues of individual consent, confidentiality, and involvement of concerned communities.
4. Each party agrees to cooperate with the principle investigator (PI) in selective reporting of focused results so as to protect the integrity of subsequent research activities and uses of the shared data by the originating party.
5. The parties to this agreement agree not to use the transferred data for commercial purpose, and not to claim any intellectual property rights on the data.
6. Both parties view dissemination of research findings, both by publication and oral presentation, as an essential objective of the research. Therefore Parties are encouraged to publish the results of their work in a collaborative fashion for the benefit of the public.
7. A copy of each manuscript or abstract shall be submitted to MSF at least 30 days before submission for publication in a journal or presentation at an international meeting. MSF Publication shall be based on written consent of MSF and accompanied by an acknowledgement that MSF and the host country supplied the data. Guidelines for authorship of the International Committee of Medical Journal Editors Committee will be used to establish authorship.
8. The ownership of the data remains with MSF.
9. The information and data provided through this agreement shall only be used for the above mentioned purpose and according to the outlined terms and conditions. Neither party shall use the personal information/ data provided under this agreement for any purpose other than that set out in this agreement.

10. This Agreement shall be in full force and effect from the first date written above for a period of 5 years. This Agreement may be terminated with thirty (30) days written notice by either party or mutual agreement of the parties.
    Upon completion of the agreement, the recipient party will return all copies of the collected data and the cleaned database. MSF commits to storing the data for at least 5 years.
11. In the event of the termination of this Agreement, the personal information shared under this Agreement shall be returned to the disclosing party.

The undersigned individuals represent that they are fully authorized to execute this Agreement on behalf of the respective parties, perform the obligations under this Agreement, and make all representations, warranties, and grants as set forth herein.

MSF is willing to provide the data for use as indicated above according to the outlined conditions, and the recipient party and MSF agree to comply with those conditions.

**IN WITNESS WHEREOF** this Agreement has been signed on behalf of the disclosing party by the Medical Director of MSF:

Dr. Sidney Wong 29-July-2014

[Name] [Date]
[Signature]

**IN WITNESS WHEREOF** this Agreement has been signed on behalf of the recipient party by

Dr. Ian H. von Lindern July 28, 2014
[Name] [Date] [Signature]

Appendices (*Protocol or concept paper to be attached)*

# APPENDIX D

University *of* Idaho

**Office of Research Assurances**
**Institutional Review Board**
875 Perimeter Drive, MS 3010
Moscow ID 83844-3010
Phone: 208-885-6162
Fax: 208-885-5752
irb@uidaho.edu

To: Gregory Moller

From: Traci Craig, Ph.D.,

Chair, University of Idaho Institutional Review Board

University Research Office

Moscow, ID 83843-3010

Date: 4/29/2014 1:29:32 PM

Title: Washington Soil Lead Arsenate Study

Project: 14-142

Approved: April 29, 2014

Renewal: April 28, 2015

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On behalf of the Institutional Review Board at the University of Idaho, I am pleased to inform you that the protocol for the above-named research project is approved as offering no significant risk to human subjects.

This study may be conducted according to the protocol described in the application without further review by the IRB. As specific instruments are developed, each should be forwarded to the ORA, in order to allow the IRB to maintain current records. Every effort should be made to ensure that the project is conducted in a manner consistent with the three fundamental principles identified in the Belmont Report: respect for persons; beneficence; and justice.

This IRB approval is not to be construed as authorization to recruit participants or conduct research in schools or other institutions, including on Native Reserved lands or within Native Institutions, which have their own policies that require approvals before Human Participants Research Projects can begin. This authorization must be obtained from the appropriate Tribal Government (or equivalent) and/or Institutional Administration. This may include independent review by a tribal or institutional IRB or equivalent. It is the investigator's responsibility to obtain all such necessary approvals and provide copies of these approvals to ORA, in order to allow the IRB to maintain current records.

As Principal Investigator, you are responsible for ensuring compliance with all applicable FERPA regulations, University of Idaho policies, state and federal regulations.

# APPENDIX E

**Supplemental Material Table S3.1.** Foods reported by parents during the food survey. Foods are grouped into subcategories and names are listed in Hausa and English, where available. Foods not consumed within the past 4 weeks were not included in intake calculations.

| | **English** | ***Hausa*** | **Number of children consuming food** | **Number consuming within 4 weeks of survey** |
|---|---|---|---|---|
| breast milk | breast milk | *nonon yara* | 4 | 4 |
| cow milk | plain | *nonon shanu* | 11 | 11 |
| | in porridge | *fura* | 28 | 28 |
| | thickened milk | *kindirmo* | 5 | 5 |
| | yogurt | | 19 | 19 |
| | cottage cheese | *dakashi* | 1 | 1 |
| goat milk | plain | *nonon akunya* | 0 | 0 |
| | in porridge | *nonon akunya da fura* | 4 | 4 |
| | with cow milk and porridge | *nonon akuya da na shanu da fura* | 7 | 7 |
| | cottage cheese | *dakashi* | 6 | 2 |
| maize (corn) | boiled | *dafaffe masara* | 21 | 4 |
| | roasted | *gasassa massara* | 21 | 4 |
| | cooked, pounded | *tuwo masara* | 28 | 27 |
| | cake | *masar masara* | 23 | 23 |
| | grits roasted | *dambu masara* | 6 | 3 |
| | porridge | *pap* | 7 | 7 |
| | | *bula* | 22 | 21 |
| | bread | *hoche masara* | 14 | 12 |
| | Pounded with ground nut cake | *datun tuwo masara da kuli kuli* | 11 | 10 |
| | grits fried | *faten zakin masara* | 9 | 6 |
| guinea corn (sorghum) | boiled | *dawa* | 1 | 1 |
| | cooked, pounded, | *tuwon dawa* | 27 | 27 |
| | cake | *masar dawa* | 21 | 21 |
| | porridge | *puran dawa* | 19 | 17 |
| | grits | *dambun dawa* | 15 | 9 |
| | bread | *hoche dawa* | 24 | 24 |
| | pounded with ground nut cake | *datun tuwon dawa da kuli kuli* | 11 | 11 |
| | grits fried | *faten tsakin dawa* | 10 | 10 |
| millet | roasted | *tumu* | 10 | 1 |
| | cooked, pounded | *tuwon gero* | 4 | 2 |
| | cake | *masar gero* | 4 | 4 |
| | porridge | *puran gero* | 26 | 26 |
| | grits | *dambun gero* | 5 | 3 |
| | | *bula* | 13 | 11 |
| | bread | *hoche gero* | 1 | 1 |
| | pounded with ground nut cake | *datun tuwon gero da kuli kuli* | 2 | 0 |
| | porridge | *pap* | 0 | 0 |
| locally grown rice | plain | *shinkafa gidan* | 4 | 4 |
| | cooked, pounded | *tuwon shinkafa* | 27 | 26 |
| | cake | *masar shinkafa* | 11 | 9 |
| | grits | *dambun shinkafa* | 1 | 1 |
| | jolof | *dafaduka* | 26 | 26 |
| | with sauce | *shinkafa de mia* | 24 | 24 |
| | with ground nut cake | *shinkafa da kuli kuli* | 3 | 3 |
| white rice | plain | *shinkafa yar gawamati* | 3 | 3 |
| | cooked, pounded | *tuwon shinkafa* | 23 | 22 |

| | English | *Hausa* | Number of children consuming food | Number consuming within 4 weeks of survey |
|---|---|---|---|---|
| | cake | *masar shinkafa* | 3 | 3 |
| | grits | *dambun shinkafa* | 1 | 1 |
| | jolof | *dafaduka* | 25 | 23 |
| | with sauce | *shinkafa de mia* | 22 | 20 |
| | with ground nut cake | *shinkafa da kuli kuli* | 2 | 2 |
| cow pea | plain | *wake kawai/farin wake* | 6 | 6 |
| | beans and rice | *shinkafa de wake* | 27 | 27 |
| | fried bean cake | *kosai* | 26 | 26 |
| | boiled bean cake | *alala* | 20 | 19 |
| | mashed beans | *faten wake* | 7 | 7 |
| | ground, boiled, with oil and pepper | *dan wake* | 10 | 7 |
| | bean cake | *masar wake* | 12 | 11 |
| tapery bean | plain | *jan wake* | 0 | 0 |
| | beans and rice | *shinkafa de wake* | 5 | 4 |
| | fried bean cake | *kosai* | 3 | 3 |
| | boiled bean cake | *alala* | 2 | 1 |
| | mashed beans | *faten wake* | 0 | 0 |
| | ground, boiled, with oil and pepper | *dan wake* | 0 | 0 |
| peas | plain | *gujiya/kwaruru* | 20 | 4 |
| soy bean | soy bean | *wake suya/wara* | 6 | 6 |
| | tofu | *awara* | 27 | 27 |
| | porridge | *pap* | 0 | 0 |
| sweet potato | plain | *kudaku/dankali* | 22 | 21 |
| | pounded, with ground nut cake | *luluyan dankali* | 11 | 10 |
| | fried with chilies | *soyayyen dankali* | 24 | 24 |
| | mixed with rice | *shinkafan de dankali* | 9 | 9 |
| yam | plain | *doya* | 22 | 21 |
| | pounded, with ground nut cake | *luluyan doya* | 9 | 8 |
| | fried with chilies | *soyayyen doya* | 13 | 13 |
| | mixed with rice | *shinkafan de doya* | 21 | 20 |
| | mashed | *faten doya* | 5 | 5 |
| | pounded, with ground nut cake | *luluyan mankani* | 6 | 5 |
| | mixed with rice | *shinkafan de mankani* | 2 | 1 |
| Irish potato | plain | *dankalin turawa* | 4 | 2 |
| | pounded with ground nut cake | *luluya* | 0 | 0 |
| | fried with chilies | *soyayyen dankalin turawa* | 2 | 2 |
| | mixed with rice | *shinkafan de dankalin turawa* | 0 | 0 |
| | Irish potato fried with egg | *dankalin turawa da kwai* | 1 | 1 |
| cassava | plain | *rogo* | 16 | 9 |
| | cassava with ground nut cake | *rogo da kuli kuli* | 17 | 9 |
| | cassava premade | *gari kwaki* | 8 | 4 |
| | cassava premade with sugar | *gari da suga* | 10 | 8 |
| | premade with ground nut cake | *gari da kuli kuli* | 11 | 8 |
| | cassava, cooked and pounded | *taiba* | 5 | 3 |
| ground nut | roasted | *soyayye gyada* | 21 | 17 |
| | boiled | *dafaffen gyada* | 19 | 5 |
| | cake | *kuli kuli* | 23 | 23 |
| | oil | *mangyada* | 22 | 22 |
| | porridge | *kunun gyada* | 0 | 0 |
| | soup | *mia* | 17 | 17 |
| | with leaf | *mia kuli da ganye* | 18 | 17 |
| tomato | plain | *tomati* | 8 | 8 |
| | tomato soup | *tomati da mia* | 21 | 21 |

| | English | *Hausa* | Number of children consuming food | Number consuming within 4 weeks of survey |
|---|---|---|---|---|
| | tomato with ground nut oil | *tomati da mai* | 8 | 8 |
| | cooked, mixed with leaves | *ganye da feffe* | 11 | 8 |
| other vegetables | peppers | *yaji* | 8 | 8 |
| | pepper soup | *miyar yaji* | 10 | 10 |
| | carrots | *karas* | 8 | 5 |
| | green beans | *hakin wake* | 4 | 4 |
| | cabbage, raw and cooked | *kabeji* | 18 | 10 |
| | pumpkin soup | *kabushi* | 14 | 8 |
| | lettuce | *latas* | 22 | 21 |
| | onion | *albasa* | 17 | 17 |
| | cucumber, raw | *?* | 1 | 1 |
| okra | plain | *kubewe/guro* | 3 | 3 |
| | okra soup | *miyan guro* | 26 | 25 |
| amaranth | cooked | *aleafu* | 11 | 7 |
| | amaranth soup | *miyan aleafu* | 18 | 16 |
| juice | juice | *juse* | 16 | 16 |
| | sure | *zutu* | 8 | 4 |
| | sure juice | *zobo* | 14 | 13 |
| fruit | can fruit | *abincin gwanwani* | 2 | 2 |
| | sour lemon | *lemun tsami* | 5 | 3 |
| | banana | *ayaba* | 16 | 15 |
| | apple | *apple* | 3 | 2 |
| | orange | *lemu* | 25 | 24 |
| | papaya/paw paw | *gwadda* | 5 | 5 |
| | guava | *gwaiba* | 16 | 11 |
| | pineapple | *abarba* | 12 | 7 |
| | water melon | *kankana* | 27 | 24 |
| | mango | *mangwano* | 20 | 11 |
| baobab leaves | cooked | *kuka* | 4 | 4 |
| | soup | *miyan kuka* | 25 | 25 |
| local tree or plant leaves | cooked with kuli kuli and oil | *zogala* | 23 | 14 |
| | cooked with kuli kuli and oil | *rama* | 21 | 7 |
| | cooked with kuli kuli and oil | *yadiya* | 7 | 2 |
| | tree leaf, with kuli kuli and oil | *dinkim* | 12 | 8 |
| | cooked with kuli kuli and oil | *tafasa* | 21 | 11 |
| | cooked with kuli kuli and oil | *lalo* | 18 | 2 |
| | cooked with kuli kuli and oil | *zutu* | 14 | 10 |
| | leaves of plant, cooked | *hakin wake* | 18 | 11 |
| fruit tree leaf mix | cooked | *kayan magani* | 15 | 7 |
| wild plant seeds | cooked, pounded | *dadawa* | 23 | 23 |
| red meat | cow | *naman shanu* | 24 | 24 |
| | camel | *rakumi* | 11 | 7 |
| | horse | *doki* | 0 | 0 |
| | donkey | *jaki* | 0 | 0 |
| | goat | *akuya, bunsuru* | 25 | 24 |
| white meat | chicken | *kaza* | 24 | 24 |
| | guinea fowl | *zabo* | 15 | 13 |
| | birds | *tsuntsaye* | 4 | 2 |
| wild game | lizard | *damo, tsari* | 9 | 5 |
| | rabbit | *zomo* | 5 | 4 |
| | snake | *maciji* | 1 | 1 |

| | English | *Hausa* | Number of children consuming food | Number consuming within 4 weeks of survey |
|---|---|---|---|---|
| | rat | *bera* | 6 | 3 |
| | mice | *kahiya* | 4 | 3 |
| | crocodile | *kada* | 0 | 0 |
| | porcupine | *bushiya* | 7 | 6 |
| egg | chicken | *kwan kaji* | 22 | 19 |
| | guinea fowl | *kwan zabo* | 18 | 10 |
| fish | locally caught | *kifi* | 25 | 23 |
| | canned sardines | *kifin gwangwani* | 4 | 3 |
| spices / flavoring | soil for flavor | *jarkasa* | 2 | 2 |
| | soil for flavor | *kanwa* | 14 | 14 |
| | ginger | *citta* | 10 | 10 |
| | tamarind | *tsamiya* | 12 | 9 |
| purchased / processed foods | biscuit | *biskin* | 24 | 24 |
| | bread | *biredi* | 25 | 25 |
| | macaroni/spaghetti | *taliya* | 20 | 19 |
| | indomie | *taliya* | 14 | 13 |
| | coffee or tea | *shayi* | 19 | 17 |
| | soda | *coca cola/fanta* | 12 | 12 |
| | margarine | *blue band* | 0 | 0 |
| | chewing gum | *cingam* | 23 | 23 |
| | sweets | *minti* | 20 | 20 |
| | ice cream | | 0 | 0 |

# APPENDIX F

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Best wishes

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# APPENDIX G

Chapter 2 is reprinted from Environmental Health Perspectives (124:9), *Environmental Remediation to Address Childhood Lead Poisoning Epidemic due to Artisanal Gold Mining in Zamfara, Nigeria*.

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To: Casey Bartrem <bart1432@vandals.uidaho.edu>, "Glazener, Tracey (NIH/NIEHS) [C]" <glazener@niehs.nih.gov>, "Dimes, Martha (NIH/NIEHS) [E]" <dimes@niehs.nih.gov>
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# APPENDIX I

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