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Research Paper

Assessing human metal accumulations in an urban superfund site

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ABSTRACT

Butte, Montana is part of the largest superfund site in the continental United States. Open-pit mining continues in close proximity to Butte's urban population. This study seeks to establish baseline metal concentrations in the hair and blood of individuals living in Butte, MT and possible routes of exposure. Volunteers from Butte (n = 116) and Bozeman (n = 86) were recruited to submit hair and blood samples and asked to complete a lifestyle survey. Elemental analysis of hair and blood samples was performed by ICP-MS. Three air monitors were stationed in Butte to collect particulate and filters were analyzed by ICP-MS. Soil samples from the yards of Butte volunteers were quantified by ICP-MS. Hair analysis revealed concentrations of Al, As, Cd, Cu, Mn, Mo, and U to be statistically elevated in Butte's population. Blood analysis revealed that the concentration of As was also statistically elevated in the Butte population. Multiple regression analysis was performed for the elements As, Cu, and Mn for hair and blood samples. Soil samples revealed detectable levels of As, Pb, Cu, Mn, and Cd, with As and Cu levels being higher than expected in some of the samples. Air sampling revealed consistently elevated As and Mn levels in the larger particulate sampled as compared to average U.S. ambient air data.

1. Introduction

Butte, MT a town with over a century of copper and molybdenum mining activities is home to approximately 30,000 individuals and is also part of the largest Superfund complex in the United States to date. Butte was given Superfund designation in 1983. In 1994 the United States Environmental Protection Agency (EPA) issued a Record of Decision designating Butte, along with the neighboring town and mining site of Anaconda, MT, and 120 miles of Montana's Clark Fork River, as a single Superfund complex (EPA, 1994). The EPA identified potential health threats from direct contact with, or ingestion of, contaminated soils, water, or inhalation of contaminated air (EPA, 1994). Epidemiological studies conducted in the 1990s and the early 2000s suggested that cancer rates (National Cancer Institute, 2009), as well as neurodegenerative diseases (e.g. Multiple sclerosis, Parkinson's disease) (Satterly, 1995), are higher than average in Butte (Silver-Bow County) as compared to the rest of Montana and the nation. More current epidemiological reports from The National Cancer Institute now list Silver Bow County as a priority 8, or declining trend, county (National Cancer Institute, 2017). In addition to the historical mining activity and waste, current open-pit mining activities continue to occur within the city limits of Butte. While most studies have focused on the historical waste issues in Butte, little is known about the effects of current surface copper and molybdenum mining within the city limits.

Exposure assessment is often considered the weakest link in risk assessment (Kakkar and Jaffery, 2005). However, the relationship between exposure of a population to a pollutant and the subsequent biological effect is an important aspect of environmental epidemiology. Metals are known to cause a variety of disease states depending on the amount and type of metal exposure, the length of exposure time, and the individuals' genetic predisposition (Yoon et al., 2008; Jomova and Valko, 2011). Chronically high levels of redox active metals are known to participate in inflammatory response and oxidative stress (Gaetke and Chow, 2003). In addition, aberrant levels of metals, such as high copper and low zinc, are implicated in a variety of diseases, especially neurodegenerative diseases (Lim et al., 2010). Many metals or metalloids, such as lead, mercury, cadmium and arsenic can lead to toxic effects in humans, even at low levels of exposure (Zhang et al., 2015). Metals that are essential trace micronutrients, such as copper, zinc and manganese, have been implicated in protein dysregulation and disease

Abbreviations: ICP-MS, inductively coupled plasma–mass spectrometry; TSP, total suspended particulate; ppm, parts per million; ppb, parts per billion; Al, aluminum; As, arsenic; Cd, cadmium; Cu, copper; Pb, lead; Mn, manganese; Mo, molybdenum; U, uranium; Zn, zinc

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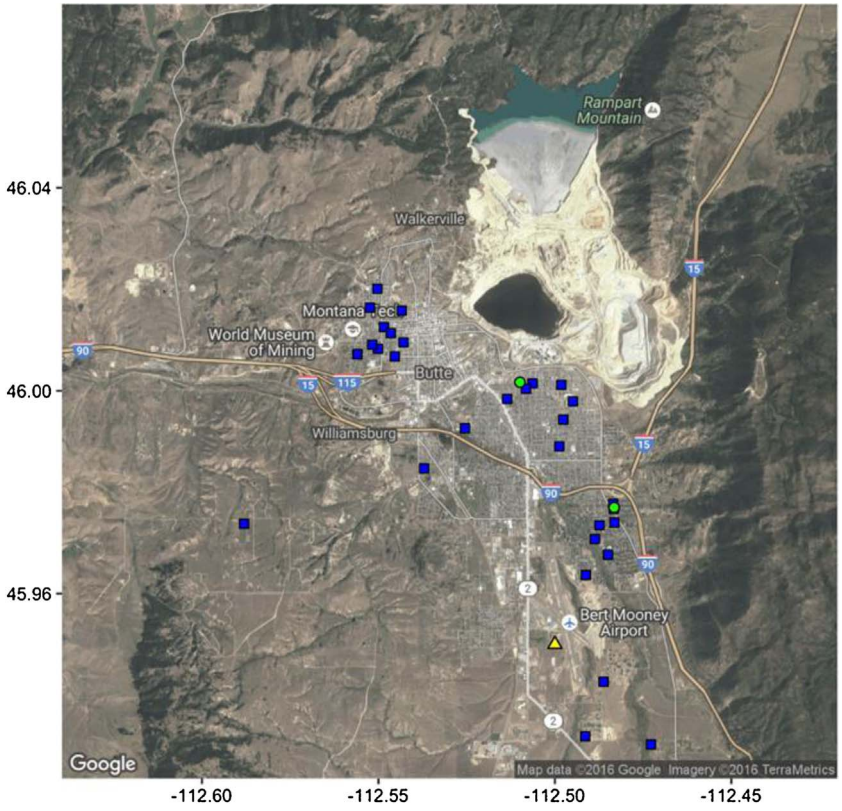

Fig. 1. Butte, MT study area wherein soil (blue squares) and air (green circles) samples were collected. Soil samples were collected once and air samples were collected weekly during the period of May–October 2015. Weather information collected at Bert Mooney Airport weather station (yellow triangle). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

states when found in elevated concentrations outside of the optimum range (Gaetke and Chow, 2003).

Very little human sampling has been undertaken in Butte, MT, the nation's largest Superfund Site, and an area with extensive and on-going metal contaminate release to the environment. This study seeks to establish current metal concentrations in individuals living within close proximity to the surface copper mine in Butte, MT by sampling both blood and hair of volunteers. To date, no study has evaluated the contribution to metal exposures through inhalation or ingestion of dust from the surface mine in the presence of other primary pathways, such as smoking or diet. Additionally, correlations among different metals, which may reveal potential antagonistic or synergetic sources, have been completely unexplored.

Butte, MT sits at an elevation of approximately 5500 ft above sea level, has a semi-arid climate, and is surrounded on three sides by the Continental Divide. Open pit mining began in 1955 with the creation of the Berkeley Pit which operated until 1982. Superfund designation was given in 1983 and then interestingly the Continental Pit, which is adjacent and to the east of the Berkeley Pit, opened in 1986 (Pit Watch, 2016). The Continental Pit is still currently in operation. Fig. 1 is a Google Earth image illustrating the layout of the town of Butte as well as the size and location of the Berkeley Pit, Continental Pit, and the Tailings Pond north of town. In addition, locations 1 and 2 (Fig. 1, green circles) are identified on the map as the 2 locations of the air monitoring stations that were in use from May 2015–October 2015. Other locations mapped show approximate locations of soil samples that were collected throughout town (Fig. 1, blue squares).

This study seeks to establish baseline metal accumulations in the Butte population through a variety of mechanisms. First, volunteers were recruited from Butte to submit hair and blood samples to be digested and analyzed for metal content by inductively-coupled plasma mass spectrometry (ICP-MS). These data were compared to a control population of volunteers recruited from Bozeman, MT (Gallatin County). Bozeman is a town approximately 90 miles to the east of Butte with no historical or current mining practices.

Biological monitoring is a commonly used method to assess the potential accumulation of emissions in a population. Blood, urine, feces, hair and nails are all biological materials that are commonly used to quantify trace metal accumulations (Elinder et al., 1988; Kucers et al., 1995) and all methods have advantages and limitations in their use. One of the major limitations in biological monitoring is the lack of correlation of a trace element between matrices type (i.e. hair vs blood or whole blood vs serum) (Hall et al., 2006; Schultze et al., 2014) with blood giving information for acute exposure (Pomroy et al., 1980) and hair as a way to monitor longer exposure times (Bencko, 1995; Iyengar, 1998; Klaassen, 2001). Hair is an attractive, non-invasive, highly mineralized biological monitoring source that reflects minerals and chemicals contained in the organism. This means that hair elemental concentrations correlate well to the actual internal levels of those chemical substances in the body (Szynkowska et al., 2015).

In addition to the human sampling, soil samples were collected from residences of 32 of the Butte volunteers. Soil samples were dried, sieved, acid digested and analyzed for metal content by ICP-MS. To more fully understand the potential environmental routes of exposure, three air monitors were placed in two locations in Butte from late May–October 1, 2015. These three monitors were set to continuously collect either PM 10 μm or total suspended particulate (TSP). Large air particles, PM 10 and larger, were monitored in this study since dust production from the current mining activity is of major concern rather than combustion of particles into the smaller, 2.5 μm particle size. Each week, the air filters were weighed and portions were acid digested and analyzed for metal content by ICP-MS.

Data collected suggests that current open-pit mining practices within the city limits of Butte, MT may be causing chronic inhalation and/or ingestion of low levels of various metals. Both hair and blood data revealed elevated levels of As in the Butte population as compared to Bozeman. In addition, Al, Cu, Cd, Mn, Mo, and U were found to be elevated in hair samples of Butte residents as compared to Bozeman residents. These elevated metals could be associated with aberrant biological regulation of important enzymatic and biological cofactors