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Spatial patterns of tungsten and cobalt in surface dust of Fallon, Nevada

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Abstract Spatial patterns of tungsten and cobalt are described for surface dust of Fallon, Nevada, where a cluster of childhood leukemia has been ongoing since 1997. In earlier research, airborne tungsten and cobalt was shown to be elevated in total suspended particulates in Fallon. To fine-tune the spatial patterns of tungsten and cobalt deposition in Fallon, surface dust was collected in a grid pattern within as well as outside of Fallon to establish background concentrations of metals. In surface dust, tungsten and cobalt show sharp peaks (934 ppm and 98 ppm, respectively) within Fallon just north of highway 50 and west of highway 95. These two peaks overlap spatially, and given the grid pattern used for collecting surface dust, the source area of these two airborne metals can be pinpointed to the vicinity of hard-metal industry located north of highway 50 and west of highway 95. Fallon is distinctive in west central Nevada because of high airborne tungsten and cobalt particulates, and given its cluster of childhood leukemia, it stands to reason that additional biomedical research is in order to test directly the leukogenicity of combined airborne tungsten and cobalt particulates.

Keywords Fallon · Nevada · Childhood leukemia · Tungsten · Cobalt · Surface dust chemistry

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Introduction

Spatial patterns of tungsten and cobalt are described for surface dust of Fallon, Nevada (Fig. 1), where a cluster of childhood leukemia has been ongoing since 1997. Officially, 16 cases of childhood leukemia were diagnosed from 1997 to 2002 inclusive (Expert Panel 2004), and one additional case was reported in December 2004 (Nevada State Health Division 2004). Given Fallon's pediatric population of ~2,500 children up to 19 years in age (U.S. Census 2000) and a national expected rate of childhood leukemia of

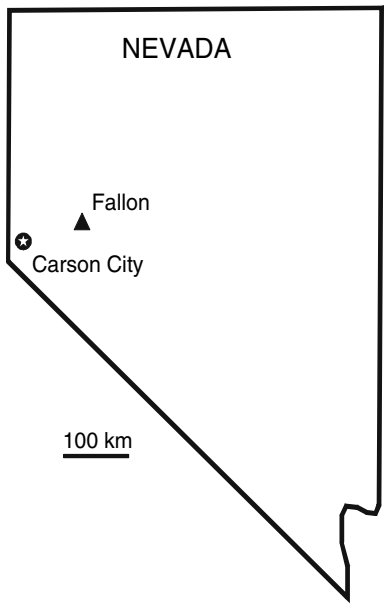

Fig. 1 Map of Nevada showing the location of Fallon

4.1 cases per 100,000 children up to 19 years in age per year (US NCI 2003), the expected rate of childhood leukemia for Fallon should be only one case every 10 years.

This cluster, deemed ''one of the most unique ever reported'' (Steinmaus et al. 2004), has prompted extensive research in an effort to determine if an environmental cause might be responsible. Prior research has included drinking water (Moore et al. 2002), jet fuel (US ATSDR 2002), pesticides (US CDC 2003a), surface water (U.S. ATSDR 2003a), outdoor air (US ATSDR 2003b), surface soil and indoor dust (US ATSDR 2003c), potential lingering effects of underground nuclear bomb testing in the area (Seiler 2004), and groundwater (Seiler et al. 2005). Although few definitive conclusions have been made, significantly elevated airborne tungsten and cobalt levels have been identified in airborne particulates within Fallon relative to comparison towns (Sheppard et al. 2006) and in lichens within Fallon compared to outlying desert areas (Sheppard et al., 2007). Toxicological research on effects of tungsten and cobalt on leukemia has been recommended, as has additional environmental research in Fallon to improve understanding of its airborne tungsten and cobalt.

An additional environmental monitoring technique that is applicable to Fallon is surface dust chemistry, the measurement and interpretation of element concentrations in fine sediments that accumulate on outdoor surfaces. Surface dust is an ideal indicator of atmospheric deposition, especially for heavy metals. Surface dust is easy to collect, so large spatial arrays of samples can be obtained quickly. Surface dust reflects the chemical composition of recent deposition, on the order of days to weeks depending on precipitation frequency. By collecting surface dust in a grid pattern, it is possible to map differing concentrations of atmospheric deposition of heavy metals and thereby pinpoint sources of unusual airborne metals (Mielke et al. 1999; Lee et al. 2006). Paired studies of surface dust and total suspended particulates can be particularly fruitful for confirming airborne chemistry and identifying spatial patterns of metals (Muskett and Jones 1980; Boudissa et al. 2006). Many case studies exist worldwide of using surface dust chemistry to quantify atmospheric loading of heavy metals and/or identify spatial patterns of deposition (Harrison 1979; Duggan 1984; Fergusson and Ryan 1984; Thornton et al. 1985; Rapsomanikis and Donard 1985; Davies et al. 1987; Tam et al. 1987; Wong and Mak 1997; Benin et al. 1999; Reid et al. 2003; Clark et al. 2005). Accordingly, surface dust chemistry in and around Fallon was used to establish the spatial patterns of tungsten and cobalt deposition, with the objective of pinpointing the source or sources of elevated airborne tungsten and cobalt particulates within Fallon.

Methods

Fallon is a small, rural farming community (Greater Fallon Area Chamber of Commerce 2005) located in west central Nevada (Fig. 1). Its climate is cool to mild and dry, with a mean annual temperature and precipitation of 10.7°C and 127 mm, respectively, as typified from meteorological data from Fallon (monthly data from 1931 to 2004 obtained online from the National Climatic Data Center, NOAA 2006). Along with service industries and small businesses, Fallon has

an industrial facility that carries out hard-metal metallurgy, including tungsten carbide and cobalt (Harris and Humphreys 1983). This hard-metal facility has been suggested as a candidate source of tungsten within Fallon generally (Reno Gazette-Journal, 5 February 2003) and more specifically for elevated airborne tungsten and cobalt in total suspended particulates of Fallon (Sheppard et al. 2006).

The strategy for surface dust chemistry was to map concentrations of elements in outdoor dust deposits within and around Fallon. Fieldwork took place in March, 2005, at which time Fallon had not experienced a substantial rain storm for three weeks (daily data obtained online from the National Climatic Data Center, NOAA 2006), allowing surface dust to accumulate for that length of time. In total, 125 surface dust samples were collected along a grid pattern within Fallon as well as outside of Fallon to establish background concentrations of metals (Amini et al. 2005; Rimmer et al. 2006; Biasioli et al. 2006). Within Fallon, the grid cell length was 0.5 km, which maximized the sampling density inside the town. Outside of Fallon, the grid cell length was relatively coarser (2.0–5.0 km), which maximized the spatial extent of sampling away from Fallon. The high density of sampling points and large overall spatial coverage throughout Fallon and outside of the town was intended to account for variation in surface dust chemistry that might be due to wind and/or unusual distribution of the population. Geographic coordinates were recorded for each sample to facilitate spatial analysis of metal concentrations across Fallon and the surrounding area and to map distributions of metals.

Surface dust samples were collected mostly from paved surfaces using a clean brush and clean paper as a dustpan (Harrison 1979; Duggan 1984; Thornton et al. 1985; Harrop et al. 1990). Samples were stored in clean polyethylene vials. Outside of Fallon, where surfaces are not paved, samples were collected from undisturbed desert and therefore contained some coarse sand. In all cases, samples were sieved to isolate the fine fraction (<0.150 mm) for analysis to make them comparable. Sieved samples were transferred to clean, glass, screw-cap vials and oven-dried at 100°C for 24 h. Coarse and fine fractions from each sample are archived in the event that future analyses are necessary.

Two aliquots of each sample were prepared for analysis by instrumental neutron activation analysis (INAA). Approximately 150 mg of powder was weighed into clean high-density polyethylene vials used for short irradiations. At the same time, 200 mg of each sample was weighed into clean high-purity quartz vials used for long irradiations. Individual sample weights were recorded to ±0.01 mg. Both vials were sealed prior to irradiation. Certified standard reference materials of SRM-1633a (coal fly ash), SRM-1648 (urban particulate matter), and SRM-688 (basalt rock) were similarly prepared, as were quality-control samples (e.g., standards treated as unknowns) of SRM-1648 (urban particulate matter), SRM-278 (obsidian rock) and Ohio Red Clay (a standard developed for in-house applications).

INAA of sediments, which consists of two irradiations and a total of three gamma counts, constitutes a superset of procedures used at most INAA laboratories (Glascock 1992; Neff 1992, 2000). A short irradiation was carried out through a pneumatic tube irradiation system (Glascock 1992). Samples in polyvials were sequentially irradiated, two at a time, for 5 s by a neutron flux of 8×10^{13} n cm^{-2} s^{-1}. The 720-s count yielded gamma spectra containing peaks for nine short-lived elements: aluminum, barium, calcium, dysprosium, potassium, manganese, sodium, titanium, and vanadium. For the second irradiation, samples were encapsulated in high-purity quartz vials and were subjected to a 12-h irradiation at a neutron flux of 5×10^{13} n cm^{-2} s^{-1}. This long irradiation is analogous to the single irradiation utilized at most other laboratories. After the long irradiation, samples decayed for five days and were then counted for 1,800 s (the middle count) on a high-resolution germanium detector coupled to an automatic sample changer. The middle count yielded determinations of eight medium-half-life elements: arsenic, lanthanum, lutetium, neodymium, samarium, tungsten, uranium, and ytterbium. After an additional three-week decay, a final count of 10,000 s was carried out on each sample. The latter measurement yielded 17

long-half-life elements: cerium, cobalt, chromium, cesium, europium, iron, hafnium, nickel, rubidium, antimony, scandium, strontium, tantalum, terbium, thorium, zinc, and zirconium. Data were standardized using the standard-comparator method in which the concentrations of the unknown samples (i.e., dust samples) were determined by ratioing the measured activities per unit weight of the unknown sample to those for a reference standard (SRM-1633a, SRM-1648, and SRM-688) with known concentrations. The median recovery from the urban particulate matter SRM for all measured elements with a certified value was 96%, indicating excellent performance of INAA on surface dust. Based on the analyses of thousands of separate quality control measurements made during the past 15 years, the precision of INAA for this matrix is generally better than 5% for most elements (with the exception of As, Nd, Ni, Sr, Zn, and Zr).

Results

Most of the elements measured in surface dust show little variability across sampling points within and around Fallon, with coefficients of variation (standard deviation standardized to the mean, Sokal and Rohlf 1981) of less than 50% (Fig. 2). This establishes a background pattern with little spatial variability in atmospheric deposition of metal particulates across the greater Fallon area. In sharp contrast to this pattern of little spatial variability, tungsten shows large spatial variability, with a coefficient of variability of 732%. After tungsten, cobalt has a coefficient of variability of 77% followed by arsenic at 71%. Tungsten and cobalt have several sampling points with high values, but arsenic has only one high value.

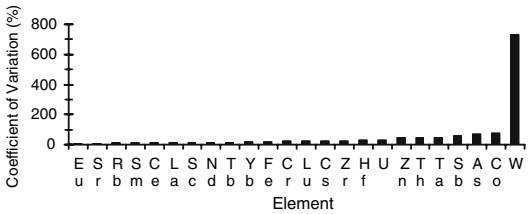

Fig. 2 Coefficients of variation (standard deviation standardized to the mean, Sokal and Rohlf 1981) of elements measured in surface dust of Fallon. For all elements, $n = 125$. Bromine, molybdenum, and nickel are not included here because they had many values below the detection limits

Tungsten and cobalt show sharp peaks (934 ppm and 98 ppm, respectively) within Fallon just north of highway 50 and west of highway 95 (Fig. 3). Spatially, these two peaks overlap exactly, with the same sampling location having the highest value for both metals. Adjacent sampling locations in north-central downtown Fallon also have notably high concentrations of tungsten and cobalt. The other sampling locations in and around Fallon have concentrations of <10 ppm tungsten and <15 ppm cobalt, which establishes natural background levels for these two metals in the area. These background values are roughly similar to typical crustal values of 1.2 ppm for W and 22 ppm for Co (Krauskopf 1995).

Discussion

Previous environmental research related to the cluster of childhood leukemia in Fallon, Nevada, has noted elevated tungsten. Tungsten has been high in drinking water (US CDC 2003b), in groundwater (Seiler et al. 2005), and in blood and urine samples of residents (US CDC 2003b). However, a consensus has been that tungsten is not unusual enough in Fallon to merit additional concern related to the cluster of childhood leukemia there. Tungsten is ubiquitous throughout northern Nevada (Stager and Tingley 1988), and elevated tungsten in and around Fallon has been suggested to be a result of the natural geology of the region (Expert Panel 2004; Pardus et al. 2005).

However, the high peak values of tungsten and cobalt in the surface dust of central Fallon are not likely to be a result of tungsten and cobalt found in rocks and/or soils of the area. Rocks collected within Fallon as well as from desert sites around Fallon show normal concentrations of tungsten and cobalt (Sheppard et al., 2007). Soils of residential Fallon and nonresidential outlying

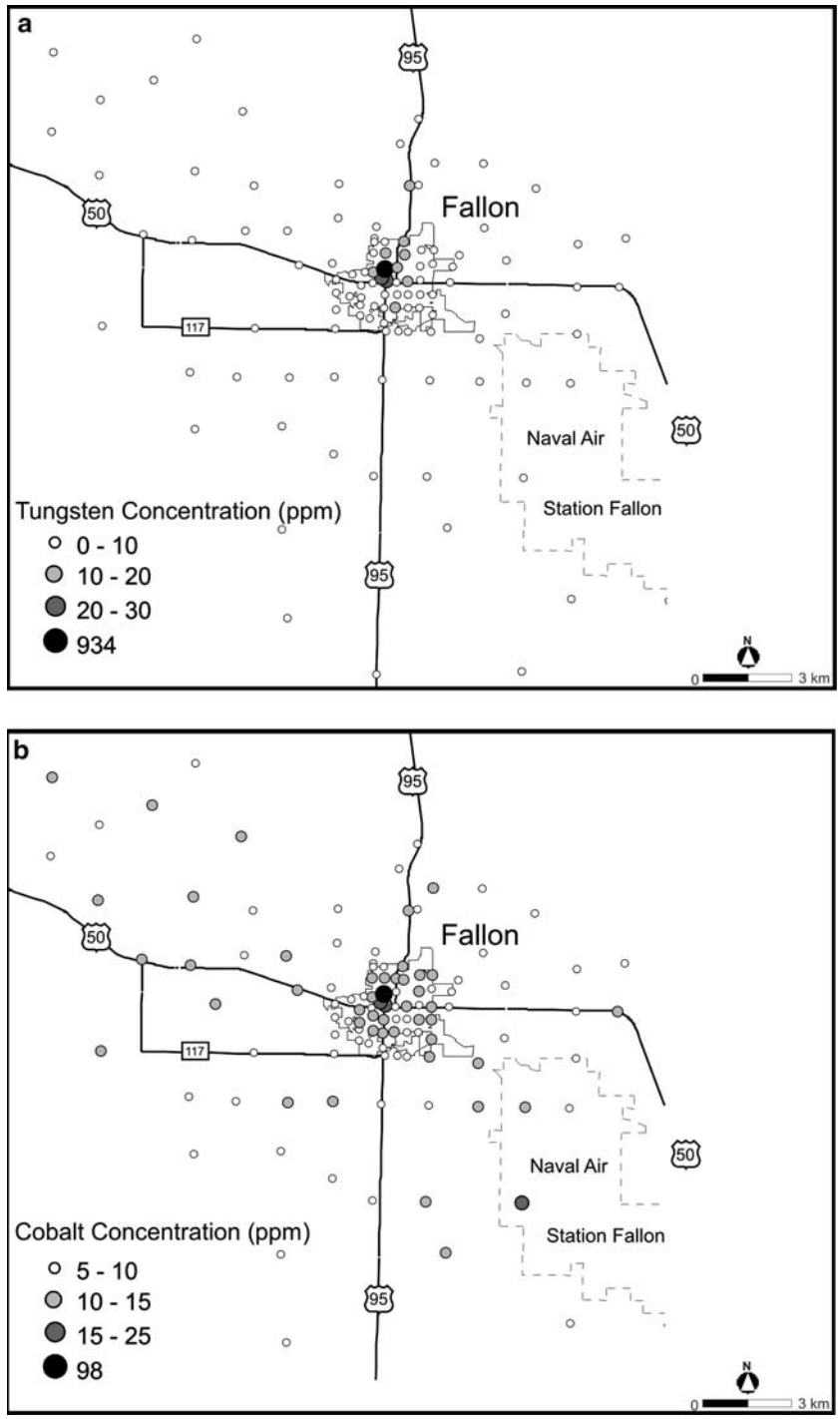

Fig. 3 Surface dust concentrations of (**a**) tungsten and (**b**) cobalt

desert areas are also not notable for high levels of any metals, presumably including tungsten and cobalt (US ATSDR 2003c). Alternatively, the peak sampling location coincides spatially with the hard-metal facility located within Fallon, which supports the suggestion that the hard-metal facility in Fallon be considered as a candidate source of Fallon's elevated airborne tungsten and cobalt particulates (Sheppard et al. 2006, 2007).

Conclusions

Fallon is distinctive from nearby comparison towns in Nevada as well as from outlying desert areas because of significantly elevated tungsten and cobalt in total suspended particulates and in lichens (Sheppard et al. 2006, 2007). This present study fine-tunes the spatial patterns of tungsten and cobalt using surface dust within Fallon. Elevated levels of these two metals, which are otherwise unrelated geologically in Nevada, suggest a local point source instead of widespread co-occurrence. Given the grid pattern used for collecting surface dust, the source area of these two metals can be pinpointed to the area of the hard-metal facility that is located just north of highway 50 and west of highway 95. This area merits direct monitoring to determine the exact source of airborne tungsten and cobalt particulates.

It cannot be concluded from only environmental data that elevated airborne tungsten and/or cobalt cause childhood leukemia. Such a connection requires biomedical research. Nonetheless, given that childhood leukemia in Fallon is the ''most unique cluster ever reported'' (Steinmaus et al. 2004) and that Fallon is distinctive because of elevated airborne tungsten and cobalt particulates, it stands to reason that additional biomedical research is in order to test directly the leukogenicity of airborne tungsten and cobalt.

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