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Spatial patterns of tungsten and cobalt on leaf surfaces of trees in Fallon, Nevada

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Abstract

Spatial patterns of airborne tungsten and cobalt are described from leaf-surface chemistry of trees in Fallon, Nevada, where a cluster of childhood leukemia has been ongoing since 1997. In earlier research, airborne tungsten and cobalt have been shown to be elevated in total suspended particulates, surface dust, and lichens from Fallon. To update data on the spatial patterns of airborne tungsten and cobalt in Fallon, leaves were collected in October 2007 from trees growing throughout Fallon. Collected leaves were measured for metals accumulated onto their surfaces. On Fallon leaf surfaces, tungsten and cobalt show maxima of 17 ppm and 6 ppm, respectively, near the center of town, north of Highway 50 and west of Highway 95. These two peaks overlap spatially, and given the dense and widespread pattern of collection, the source area of these two airborne metals can be pinpointed to the vicinity of a hard-metal industry located north of Highway 50 and west of Highway 95. Fallon is distinctive in west central Nevada for its elevated airborne tungsten and cobalt particulates, and given its cluster of childhood leukemia cases, it stands to reason that additional biomedical research is in order to test directly the leukogenicity of combined airborne tungsten and cobalt particulates.

Key words: childhood leukemia, cobalt, Fallon, leaf-surface chemistry, Nevada, tungsten

INTRODUCTION

Spatial patterns of airborne tungsten and cobalt are described from leaf-surface chemistry of trees in Fallon, Nevada (Figure 1), where a cluster of childhood leukemia cases 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 about 2500 children up to 19 years in age (U.S. Census 2000) and a national expected rate of childhood leukemia of 4.1 cases per 100 000 children up to 19 years in age per year (U.S. NCI 2003), the expected rate of childhood leukemia for Fallon should be only one case every ten years.

This cluster, deemed 'one of the most unique ever reported' (Steinmaus *et al*. 2004), has prompted multiple investigations to determine if an environmental cause might be responsible. Prior research has focused on drinking water (Moore *et al*. 2002), jet fuel (U.S. ATSDR 2002), pesticides (U.S. CDC 2003; Rubin *et al*. 2007), surface water (U.S. ATSDR 2003*a*), outdoor air (U.S. ATSDR 2003*b*), surface soil and indoor dust (U.S. ATSDR 2003*c*), 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, elevated tungsten and cobalt have been identified in total suspended particulates of Fallon relative to comparison towns (Sheppard *et al*. 2006*a*) and in lichens within Fallon compared to outlying desert areas (Sheppard *et al*. 2007*b*). Tungsten and cobalt maxima have been

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found in surface dust near the center of Fallon, just north and west of the crossroads of the main highways (Sheppard *et al.* 2007*a*). Dendrochemistry has shown that tungsten had begun increasing in Fallon tree rings by the mid-1990s, coinciding roughly with the onset of the cluster of childhood leukemia (Sheppard *et al.* 2007*c*). From direct microscopic analysis of airborne tungsten particles in Fallon, they are anthropogenic in origin, not natural (Sheppard *et al.* 2007*d*). Because of this co-occurrence in Fallon of elevated airborne tungsten and cobalt, shown with multiple lines of evidence, and an extraordinary cluster of human disease, toxicological research on effects of exposure to tungsten and cobalt particulates on leukemia is warranted.

Additional environmental research in Fallon is also warranted to continue monitoring airborne tungsten and cobalt there. An additional environmental monitoring technique that is applicable in Fallon is leaf-surface chemistry: the measurement and interpretation of element concentrations in particulates that accumulate on surfaces of leaves of trees and other plants. Leaf-surface chemistry is an ideal indicator of atmospheric chemistry (Wittig 1993), especially for heavy metals (Rautio *et al.* 1998). Leaves are easy to collect (Aksoy *et al.* 1999), so large spatial arrays of samples can be obtained quickly (Loppi *et al.* 1997). Leaf-surface particulates reflect the chemical composition of recent accumulations, of the order of weeks to months or perhaps an entire growing season, depending on the occurrence of precipitation (Alfani *et al.* 1996*b*). By collecting leaves across a region, differing accumulations of airborne metals can be mapped, thereby pinpointing source areas (Aboal *et al.* 2004). Paired studies of leaf-surface accumulations with ground surface dust and/or total suspended particulates can be particularly fruitful for confirming airborne chemistry and identifying spatial patterns of metals (Bargagli 1993; Čeburnis and Steinnes 2000). Because of these advantages, many case studies exist worldwide using leaf-surface chemistry to quantify atmospheric loading of heavy metals and/or identify their spatial patterns (e.g. Ward 1977; Dasch 1987; Alfani *et al.* 1996*a*; Aksoy and Öztürk 1997; Aksoy *et al.* 1999; Gupta *et al.* 2004; Salve *et al.* 2006; Rossini Oliva and Mingorance 2006). Accordingly, leaf-surface chemistry was used in Fallon to update spatial patterns of airborne tungsten and cobalt and to confirm their source area.

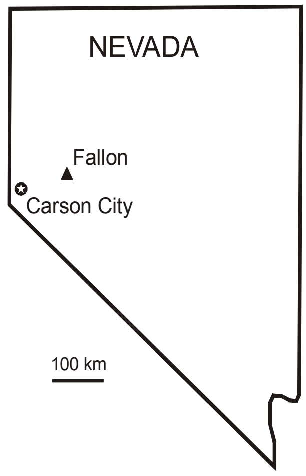

Figure 1. Map of Nevada, showing the location of Fallon.

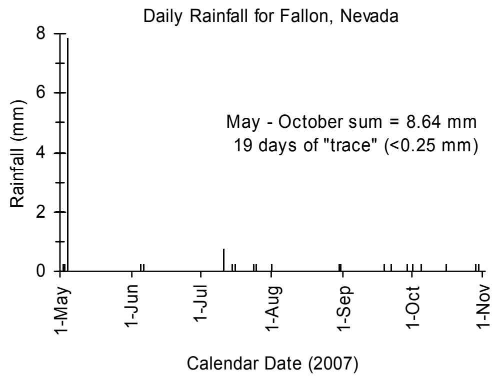

Figure 2. Daily rainfall in Fallon during May through October, 2007. Short marks indicate days with only a trace amount of rain. Daily data from NCDC, NOAA.

METHODS

Fallon is a small, rural, farming community (Greater Fallon Area Chamber of Commerce 2008) located in west central Nevada (Figure 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 2008 obtained on-line from the National Climatic Data Center, NOAA 2008). Along with service industries and small businesses, Fallon has a hard-metal metallurgical facility, which includes the production of tungsten carbide and cobalt (Harris and Humphreys 1983). The hard-metal facility has been considered a candidate source of tungsten within Fallon generally (Mullen 2003) and more specifically of elevated tungsten and cobalt in total suspended particulates and in the surface dust of Fallon (Sheppard *et al*. 2006*a*, 2007*a*).

Tree leaves were collected in mid-October 2007. All trees sampled were deciduous species, so the results of this study reflect particulate accumulations onto leaf surfaces during just the growing season of 2007. During May through October 2007, measurable rainfall was recorded at the nearby Fallon Naval Air Station on only two days (Figure 2; daily data obtained on-line from the National Climatic Data Center, NOAA 2008). Trace rainfall was recorded on 19 other days, but ultimately very little rain fell on or near Fallon during this period (8.64 mm total), and most of that (7.87 mm) fell during a single day in early May. Consequently, this study reflects particulate accumulations onto leaf surfaces during most of the growing season of May through October (Alfani *et al*. 1996*b*).

All trees sampled were broadleaved species, in part because conifer species are not common in Fallon but also because broad leaves provide an ample surface for accumulating atmospheric particulates. Tree species was not held constant because no single species predominates throughout all parts of town. The urban for-

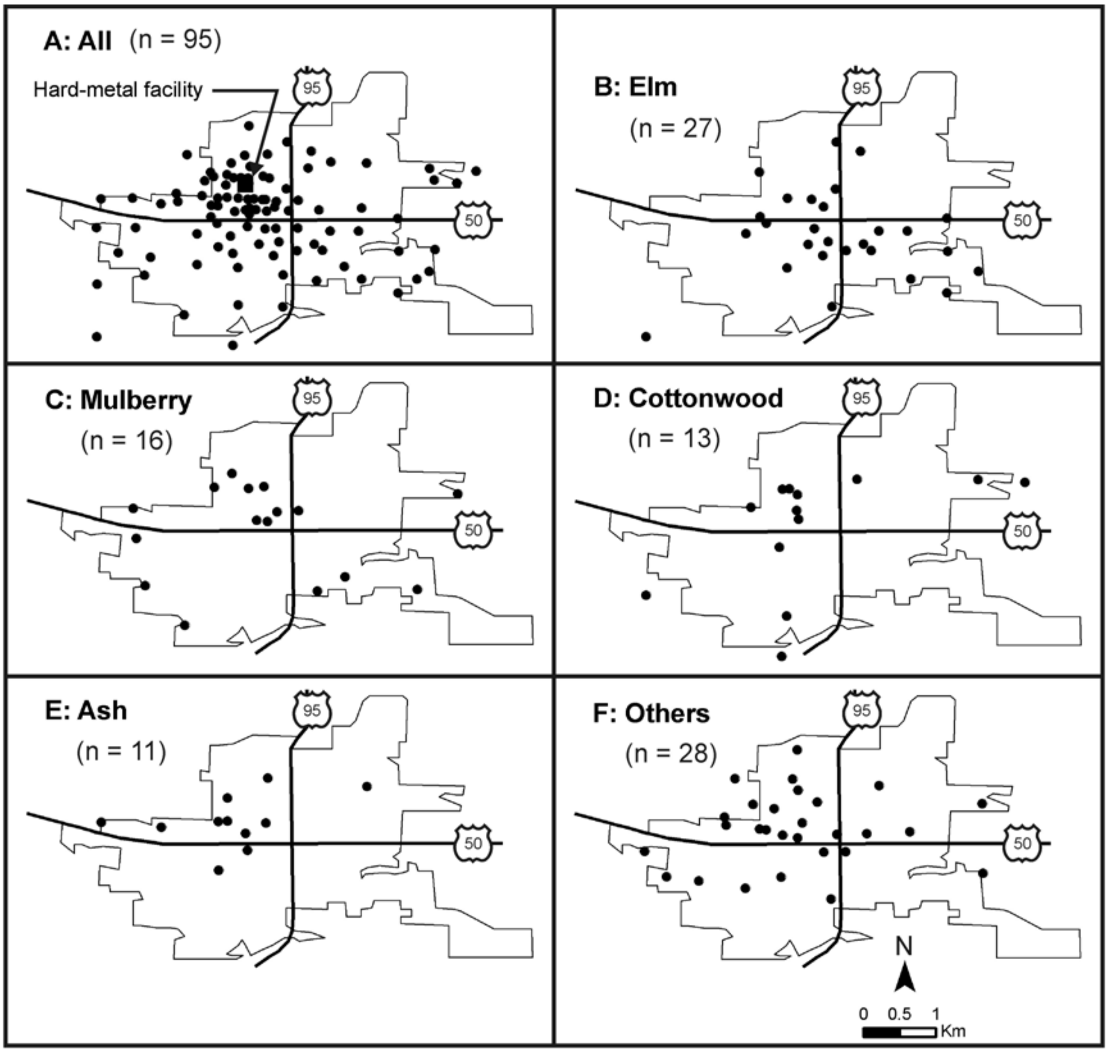

Figure 3. Maps of all sampling spots in Fallon (A), and sampling spots by dominant tree species (B–F). In all maps, the irregular polygon is the outline of Fallon. In (F), other species include box elder, Japanese maple, poplar, plantain, catalpa, locust, cherry, willow, and undetermined.

est of Fallon contains many kinds of trees, and elm (*Ulmus*), mulberry (*Morus*), cottonwood (*Populus*), and ash (*Fraxinus*) were the most common tree types sampled (Figure 3B–E). Accumulation of aerosols onto leaves can be affected by leaf characteristics such as roughness, pubescence, moisture, and stickiness (Wedding *et al*. 1977), but these leaf characteristics do not vary appreciably across the tree species sampled in this study.

Trees were selected for sampling at spatial densities that varied with distance from the hard-metal facility located near the center of Fallon (Figure 3A). Within 0.5 km of the facility, trees were sampled on average about 150 m apart (41 trees km^{-2}). From 0.5 to 1.0 km from the facility, trees were sampled on average about 270 m apart (13.6 trees km^{-2}). From 1.0 to 2.0+ km from the facility, trees were sampled on average about 510 m apart (3.1 trees km^{-2}). Sampling at variable densities provided fine resolution where spatial variability in airborne tungsten was thought to be high, as well as full coverage of the outskirts of Fallon with relatively coarser resolution (Sheppard *et al*. 2006*b*). In total, leaves from 95 trees were sampled, which is over three times the minimum of 30 suggested for contamination monitoring with leaves (Aboal *et al*. 2001). Geographic coordinates were recorded for each sampled tree to facilitate mapping concentrations of metals in particulates on leaf surfaces.

From each tree sampled, an outer branchlet of several leaves was clipped off with pruning cutters from a height of about 2 m above ground. The aspect of each tree sampled was not held constant or patterned, so sampling was effectively random across trees. None of the trees sampled was next to other trees, so there is no forest canopy effect in this study (Dasch 1987). Branchlets were stored in clean paper bags during fieldwork. Later, leaves were trimmed from their petioles using clean, ceramic (non-metal) scissors.

Leaf tissues themselves were not measured for metals content, but rather rinse-water solutions of particulates from the leaf surfaces were measured. Consequently, this study reflects airborne metals that accumulated on leaf surfaces, not soil-derived metals that moved through the trees to leaf tissues (Wolterbeek and Bode 1995). Trimmed leaves were placed in clean, 50-mL polyurethane vials, and tepid, de-ionized water was added to completely submerge the leaves. The vials were capped tightly and shaken lightly for two hours (Little 1973). Rinse solutions were poured into new, clean polyurethane vials. Rinsed leaves were then oven dried at 50°C for several days and weighed to ±0.0001 g.

Rinse solutions were filtered with acid-washed GHP Acrodisc syringe filters (<0.2 µm) and acidified to pH <2 with certified pure nitric acid. Measurement was performed using inductively coupled plasma, mass spectroscopy (ICP–MS) (Elan DRC-II, PerkinElmer,

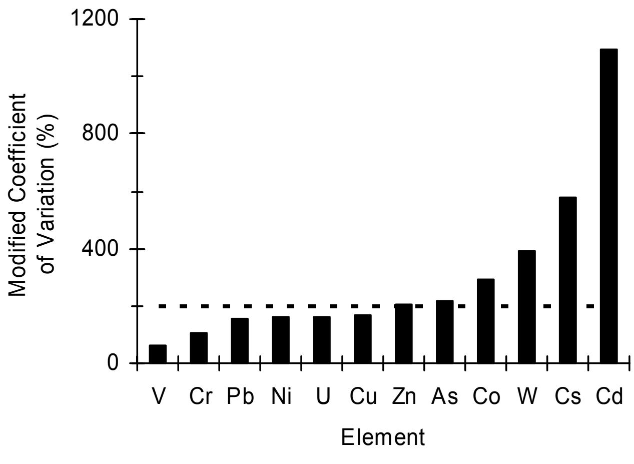

Figure 4. Modified coefficients of variation (standard deviation standardized to the mean; Sokal and Rohlf 1981) of elements measured in leaf-surface particulates of Fallon. The modification is that the median is used instead of the mean. For all elements, *n* = 95. Dashed horizontal line indicates 200% coefficient of variation.

Shelton, CT). Most analytes were measured in standard mode (vanadium, nickel, copper, zinc, arsenic, cadmium, cesium, tungsten, lead and uranium), while chromium and cobalt were measured using the dynamic reaction cell (DRC) flushed with ammonia gas. For all elements measured, detection limits were sub-ppb based on three standard deviations from the mean of eleven replicate measurements. Measured contents of metals were standardized to the oven-dry mass of leaves tested.

The city boundary, major roads, hard-metal facility, and leaf-surface metal concentrations of Fallon were integrated into a multi-layer GIS using ArcMap (ESRI 2008). Road and city boundary data were obtained from TIGER files from the U.S. Census Bureau. All geographic data were projected into a common coordinate system for overlaying. Tree species and leaf chemistry were categorized using symbology to create map legends. Maps were produced in ArcMap (ESRI 2008).

RESULTS

The dominant types of trees sampled are evenly dispersed across Fallon. Considering the two main highways of Fallon as lines dividing the town into quadrants, each of the dominant types of trees was found and sampled in at least three quadrants (Figure 3B–E). When considered as a single group, the other types of trees sampled were found in all four quadrants (Figure 3F). Consequently, spatial patterns in metal concentrations on these leaf surfaces should not be biased by the sampling of different tree types in some areas of Fallon versus not in other areas.

Most of the elements measured in leaf-surface particulates show moderate variability across sample points within and around Fallon. For most elements, modified coefficients of variation (standard deviation standardized to the mean, Sokal and Rohlf 1981, modified by using the median in place of the mean) are less than or equal to 200% (Figure 4). This establishes an expected level of spatial variability in atmospheric

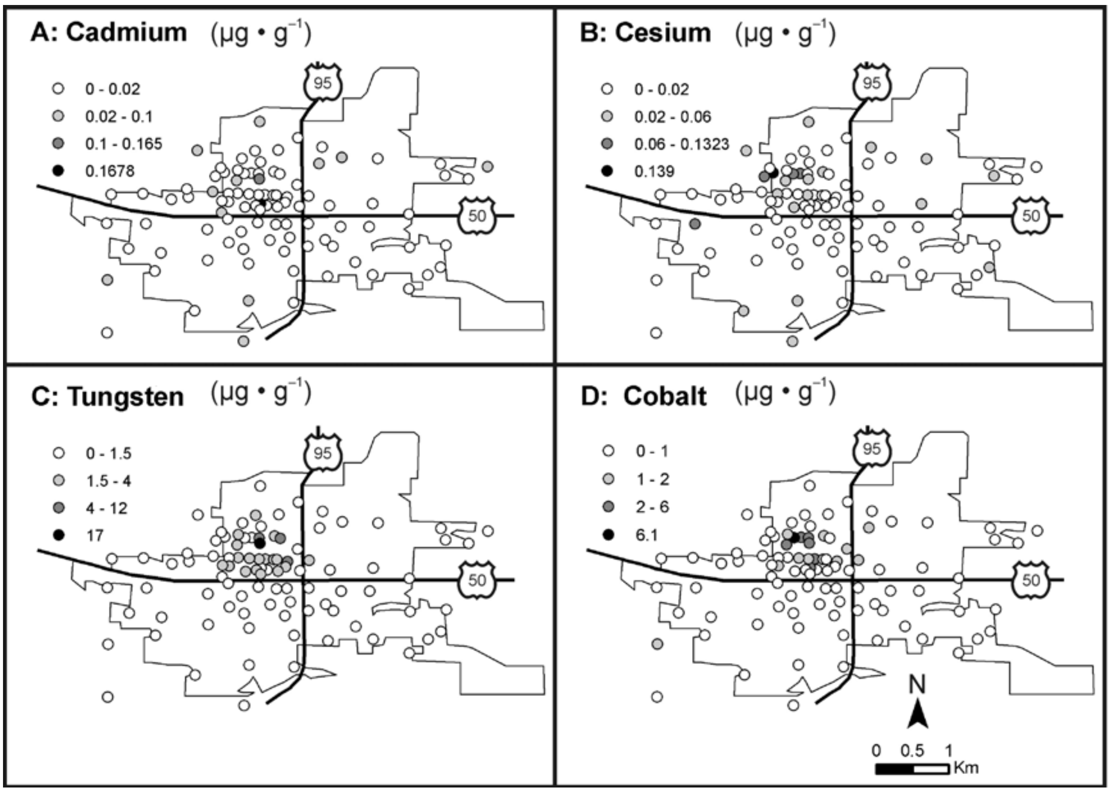

Figure 5. Maps of concentrations of cadmium (A), cesium (B), tungsten (C) and cobalt (D) in leaf-surface particulates of Fallon. In all maps, the irregular polygon is the outline of Fallon.

loading of particulate metals across Fallon. Compared to most elements, cadmium, cesium, tungsten, and cobalt show higher spatial variability, with modified coefficients of variation of 1092%, 577%, 390%, and 290%, respectively. These elements merit additional scrutiny by mapping their concentrations from all trees sampled. Cadmium and cesium have very low values (sub ppm) with only one or two high values causing their high variability. However, these two elements do not otherwise show notable spatial patterns (Figure 5A and B).

On the other hand, tungsten and cobalt have several sampling points with high accumulations that do show a notable spatial pattern in Fallon. The tungsten maximum, 17 μg g^{-1}, is located northwest of the town center, near the location of the hard-metal facility (Figure 5C). This maximum level is 30 times higher than the median of about 0.5 μg g^{-1}, which can be considered the background level of tungsten on leaf surfaces

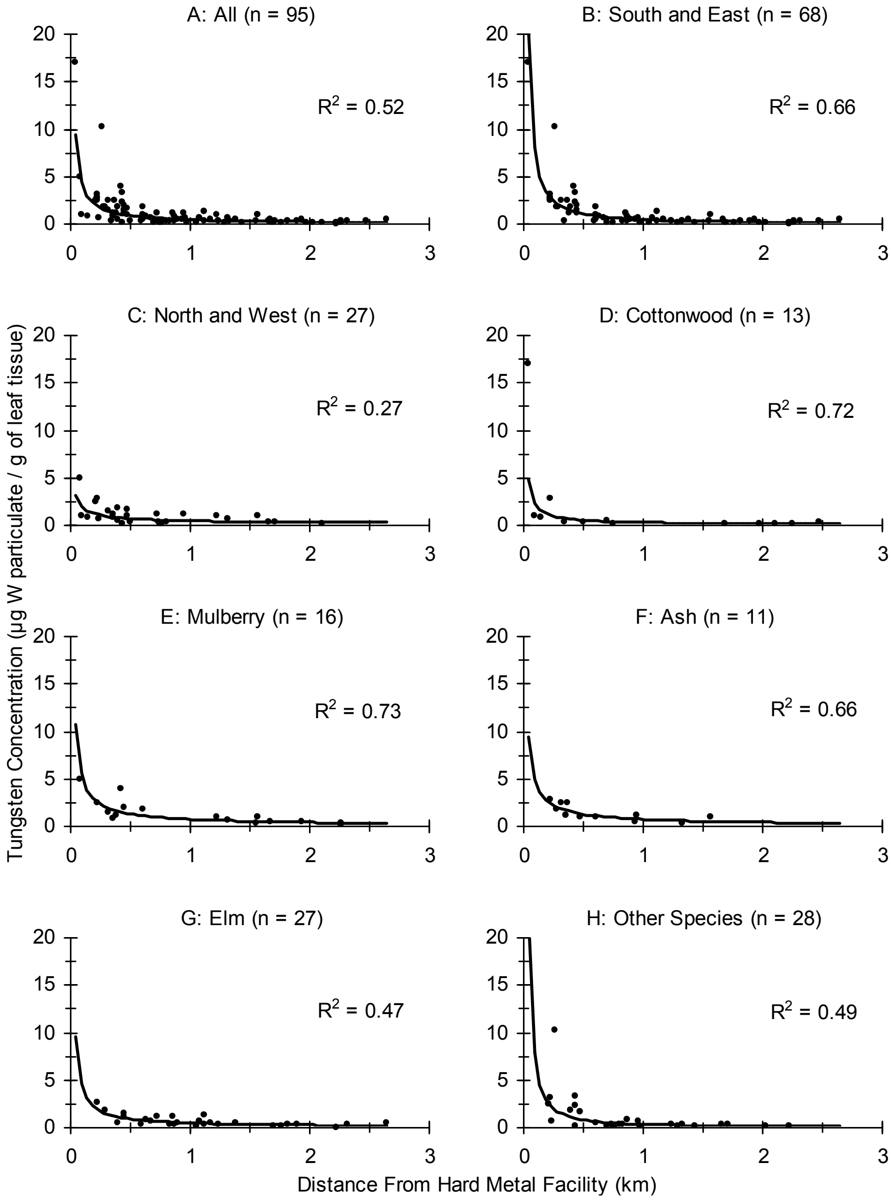

Figure 6. Bivariate scatter plots of tungsten concentrations in leaf-surface particulates as a function of distance from the hard-metal facility of Fallon. Power-model fit lines are overlain with R^2 values given.

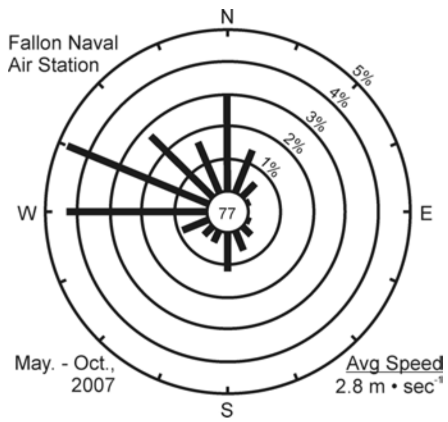

Figure 7. Wind rose diagram indicating the directions from which strong winds in Fallon came during May through October 2007. The number inside the center circle is per cent calm winds (less than 4 m sec^{-1}). Hourly data for the Fallon Naval Air Station were obtained from NCDC, NOAA; the wind rose diagram was generated using software from the Western Regional Climate Center (*http://www.wrcc.dri.edu*).

at Fallon. Several other samples show intermediate levels of tungsten, all of which came from trees located within about 0.5 km of the tree with the maximum tungsten level.

Considering all 95 data points, tungsten on leaf surfaces declines exponentially with increasing distance from the hard-metal facility (Figure 6A). This model is strong, with an R^2 of 0.52. The power model of a subset of samples located east and/or south of the hard-metal facility (i.e. at directions between 46° and 225° from the facility) is especially strong, with an R^2 of 0.66 (Figure 6B). The model of the subset of samples located west and/or north of the hard-metal facility is less strong, with an R^2 of 0.27 (Figure 6C). This difference in model strength depending on direction from the hard-metal facility reflects prevailing wind patterns during the warm season of 2007 (Figure 7). Most warm-season winds (77%) were weak (less than 4 m sec^{-1}), but strong winds (greater than or equal to 4 m sec^{-1}) were mostly from the west, northwest, or north.

The power-decline model for tungsten on leaf surfaces is consistent across tree species. The four dominant species show R^2 values ranging from 0.47 to 0.73 (Figure 6D–G), and all other species combined show an R^2 of 0.49 (Figure 6H). Given that tree species are well represented across Fallon and that they show similar power declines in tungsten on leaf surfaces with distance from the hard-metal facility, tree species is not a strong determinant of spatial variability of tungsten on leaf surfaces. Rather, the principal determinants of tungsten on leaf surfaces in Fallon are distance and direction from the hard-metal facility.

Cobalt shows a similar pattern on leaf surfaces in Fallon. The maximum level of cobalt, 6 µg g^{-1}, is located northwest of the town center, near the location of the hard-metal facility (Figure 5D). This maximum level is 16 times higher than the median value of 0.37 µg g^{-1}, which can be considered the background level of cobalt on leaf surfaces of Fallon. Several other samples show intermediate levels of cobalt, all of which came from trees located within about 0.5 km of the tree with the maximum cobalt level.

Considering all 95 data points, cobalt on leaf surfaces declines exponentially with increasing distance from the hard-metal facility (Figure 8A). This model is strong, with an R^2 of 0.40. The two power models of subsets of samples located east/south versus west/north of the hard-metal facility are equally strong, with R^2 values of 0.41 and 0.47, respectively (Figure 8B and C). The power-decline model for cobalt on leaf surfaces is also consistent across tree species. The four dominant species show R^2 values ranging from 0.44 to 0.85 (Figure 8D–G), and all other species combined show an R^2 of 0.37 (Figure 8H). Given that tree species are well represented across Fallon and that they show similar power declines in cobalt on leaf surfaces with distance from the hard-metal facility, tree species again is not a strong determinant of spatial variability of cobalt on leaf surfaces. As with tungsten, the principal

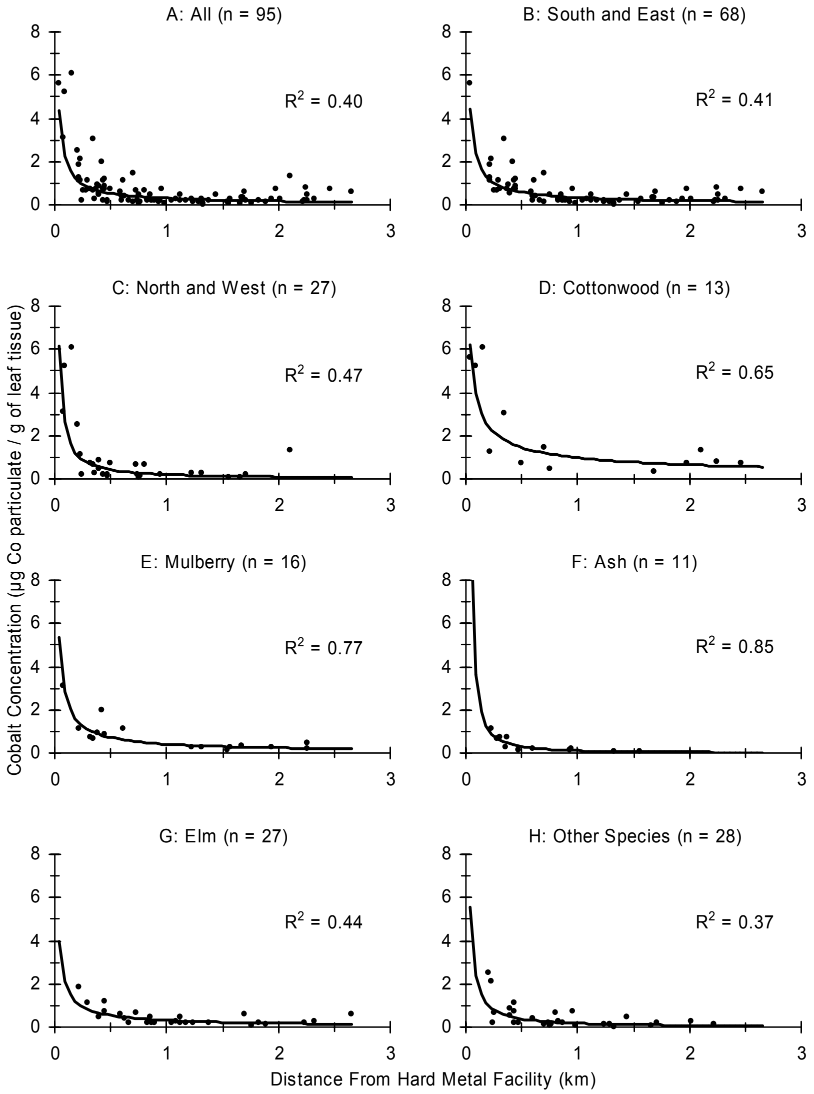

Figure 8. Bivariate scatter plots of cobalt concentrations in leaf-surface particulates as a function of distance from the hard-metal facility of Fallon. Power-model fit lines are overlain with R^2 values given.

determinants of cobalt on leaf surfaces in Fallon are distance and direction from the hard-metal facility.

DISCUSSION

Leaf-surface chemistry confirms prior environmental research in Fallon. Airborne tungsten and cobalt are elevated in Fallon relative to other towns of west central Nevada (Sheppard *et al*. 2006*a*) and to outlying desert areas (Sheppard *et al*. 2007*b*). More precisely, airborne tungsten and cobalt are especially elevated near the center of Fallon relative to the outskirts of Fallon, with notable maxima located northwest of the crossroads of the two main highways (Sheppard *et al.* 2007*a*; this study).

The question of the source of airborne tungsten and cobalt in Fallon remains open. Airborne tungsten in Fallon is anthropogenic in origin, not natural (Sheppard *et al*. 2007*d*), and the hard-metal facility in Fallon has been identified as a candidate source (Mullen 2003). However, the company has claimed that the facility no longer has any external emissions (McMillin Goodale 2005), implying that it is not the source of airborne tungsten and cobalt in Fallon. More research is needed to answer this question. For example, an environmental analysis using tracer particles (Heiken 1986) is warranted.

As an update of environmental research in Fallon, this leaf-surface chemistry study adds to years of studies showing elevated airborne tungsten and cobalt in Fallon. Earlier indications of elevated airborne tungsten and cobalt in Fallon came from total suspended particulate samples collected in March 2004 (Sheppard *et al*. 2006*a*) and then from surface dust collected in March 2005 (Sheppard *et al*. 2007*a*). Now, as of October 2007, tungsten and cobalt continue to be elevated in airborne particulates in Fallon, especially in the center of town, as indicated by leaf-surface chemistry. Consequently, pertinent public health and/or environmental protection agencies should continue monitoring airborne tungsten and cobalt in Fallon and trying to determine their source.

CONCLUSIONS

This study updates spatial patterns of airborne tungsten and cobalt in Fallon using leaf-surface chemistry. Fallon is distinguished from nearby towns in Nevada, as well as from outlying desert areas, by its elevated levels of tungsten and cobalt in airborne particulates. Elevated levels of these two metals that are otherwise unrelated geologically in Nevada suggest a local point source instead of widespread co-occurrence. Given the extent of sampling for leaf-surface chemistry, the source area of these two airborne metals is confirmed as north of Highway 50 and west of Highway 95, where the hard-metal facility is also located. This area merits direct monitoring to determine the source of airborne tungsten and cobalt particulates.

It cannot be concluded from environmental data alone that elevated airborne tungsten and/or cobalt cause childhood leukemia. Such a connection requires direct biomedical testing. Nonetheless, given that childhood leukemia in Fallon is the ‘most unique cluster ever reported’ (Steinmaus *et al*. 2004) and that Fallon is distinguished by its elevated airborne tungsten and cobalt particulates, it stands to reason that additional biomedical research is warranted to assess the leukogenicity of airborne tungsten and cobalt (e.g. Miller *et al*. 2001; Sun *et al*. 2003; Kalinich *et al*. 2005; Steinberg *et al*. 2007).

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