

Comment

Comment on “Elevated tungsten and cobalt in airborne particulates in Fallon, Nevada: Possible implications for the childhood leukemia cluster” by P.R. Sheppard, G. Ridenour, R.J. Speakman and M.L. Witten

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Sheppard et al. (2006) measured W and Co loading in air samples from Fallon and other neighboring towns in northern Nevada. They observed that W and Co loading was higher in Fallon, the location of a childhood leukemia cluster, than in the neighboring towns, and that W and Co loading decreased with distance from a hard-metal processing facility in Fallon. They concluded the temporal covariance of W and Co indicated a single source for the two, and that the heavy-metal processing facility was the likely source because no specific deposits of W and Co are known near Fallon. They hypothesized that emissions from the facility could be causally linked to the leukemia cluster because Co and W-carbide together are probably carcinogenic. It is this author's opinion that the article contained a significant error in how data were interpreted, at least two alternative hypotheses to explain their results were not evaluated, and the authors did not collect essential data necessary to test their hypothesis.

The fundamental error in interpretation made by Sheppard et al. (2006) is that comparisons are

made as W and Co loadings without evaluating concentrations or normalizing the loadings to a conservative trace element. The authors indicated that mass data was obtained by pre- and post-weighing of the collection filters; however, the loading data were not corrected to concentrations. Their results, therefore, cannot be parsimoniously interpreted to mean there is more W and Co than would be expected in Fallon dust. An alternative explanation of their results is simply that Fallon is dustier than the comparison towns. Fallon is located in the center of the Carson Desert, a large desert formed when Pleistocene Lake Lahontan dried about 10 ka ago and left large barren playas north and south of Fallon (Morrison, 1964). Winds from any direction can carry dust from surrounding desert into Fallon, whereas, all of the comparison towns are located near the bases of mountain ranges and only winds from certain directions could carry dust from nearby deserts into them. The relation between W and Co loading and distance from the facility shown by Sheppard et al. (2006; Fig. 7) could also be an artifact related to the dustiness of their sampling locations. The authors deleted two data points near the facility because they were located SW of the facility; the critical variable, however, could have been dustiness

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of the individual sampling locations and the authors do not present critical information about how, or whether, they controlled for this.

Even if the Fallon area is not dustier than the comparison towns, Sheppard et al. (2006) sampled only in towns and did not analyze local soil sources of dust for W and Co to test the alternative hypothesis that Fallon soil and dust contains naturally elevated concentrations of W and Co as a result of its unique geohydrologic history. Seiler et al. (2005) concluded that W in Fallon ground water had natural sources, one of which could be upwelling water in several geothermal areas beneath the Carson Desert. In the Fallon area numerous volcanic eruptions into Lake Lahontan occurred during the Pleistocene (Morrison, 1964), forming the basalt aquifer which is now the principal water supply for the town of Fallon (Seiler et al., 2005). Tungsten concentrations ranging from 1 to more than 1000 mg/L are readily attainable in geothermal fluids (Wood and Samson, 2000) and, thus, volcanic activity and upwelling geothermal water into Lake Lahontan during the hundreds of thousands of years it covered the Carson Desert could have enriched Fallon sediments in W, and widely distributed them in the areas surrounding Fallon. Furthermore, Co is enriched in basalts (Carr and Turekian, 1961), and Sheppard et al. (2006) did not consider contributions from these natural sources near Fallon. Volcanic activity during the Quaternary created a basalt cone that has eroded and is now exposed as a large hill in the center of Fallon approximately 2.5 km east of the facility. Large Quaternary-age basalt units are mapped 15 km north and 10 km west of Fallon (Willden and Speed, 1974) and erosion of the basalts during the Quaternary could have enriched Lake Lahontan sediment in Co near the facility and in the entire Fallon area.

Sheppard et al. (2006) imply that their observations are significant to the leukemia cluster by making the observation that Co and W-carbide appear

to have a synergistic carcinogenic effect. This is an unwarranted extrapolation of their findings because they did not test whether W-carbide is even present in the dust. SEM or electron microprobe analysis of the dust particles should readily distinguish between natural W minerals and W-carbide millings or other metallurgic W from the facility.

This author agrees that it is possible that Fallon dust is enriched in W and Co near the facility because of emissions from the facility. Nonetheless, Sheppard et al. (2006) have not demonstrated emissions from the facility are the source of the W and Co because they failed to consider alternative sources of W and Co and they failed to collect the necessary data to evaluate their hypothesis. The consequences of this paper's conclusions, if in error, are a serious matter, particularly since the facility is being sued for wrongful death on behalf of two families whose children died of leukemia (Mullen, 2003).

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Comment

Reply to comment on “Elevated tungsten and cobalt in airborne particulates in Fallon, Nevada: Possible implications for the childhood leukemia cluster”, by R. Seiler

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1. Introduction

Four criticisms of Sheppard et al. (2006) are raised in this comment. One, the data were expressed as mass per volume instead of mass per mass, and therefore they cannot be reasonably interpreted. This criticism was made both for the regional scale between towns of west central Nevada and for the local scale within Fallon. Two, the data were not normalized to a conservative trace element. Three, natural soil chemistry was not considered as an alternative explanation for elevated airborne W and Co in Fallon. Four, the exact species of W in Fallon's air was not identified and therefore cannot be associated with the assessment of the International Agency for Research on Cancer that tungsten carbide specifically might be carcinogenic in conjunction with Co (IARC, 2003). The authors respond to these criticisms as well as to a couple additional points that were raised.

2. Total suspended particulates: mass per volume or mass per mass

A common way to express contents of airborne particulates is “mass concentration”, ρ (Friedlander, 2000, p. 162), which the authors referred to as “loading” (Sheppard et al., 2006). Mass concentration is calculated by dividing the mass of particulates collected by the volume of air that was filtered (Baron and Willeke, 2001, p. 46), and it can be calculated for total suspended particulates as well as for individual constituents within total particulates, such as metals that have been quantified chemically. Because the denominator, volume of air sampled, is not an environmental variable but instead is a function of the air collection flow rate and length of time of collecting, air volume serves to standardize quantities of particulates so that they can be compared through time and space. Accordingly, total suspended particulates and their individual constituents are reported as mass per volume in order to facilitate comparisons and assessments against standards (Watson et al., 1989; US EPA, 1971, 1984). Because of these advantages, the authors opted to express the data as mass per volume.

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For individual constituents within particulates, an alternative to mass concentration is “instantaneous concentration”, c_i (Friedlander, 2000, p. 20), which is calculated as follows:

$$c_i = \rho_i / \rho$$

where ρ_i is the mass concentration of the i th chemical species of interest per unit volume of air. In this calculation, the volume of air is the same for both ρ_i and ρ , thereby canceling out and reducing instantaneous concentration to a dimensionless ratio of the mass of the i th chemical species to the total mass of particulates. Because this unit is a ratio of two environmental variables, variations in airborne metal values expressed as mass per mass could be due to changes in either the mass of the metal (numerator) or the total mass of particulates (denominator). This makes it potentially difficult to interpret mass per mass unambiguously: Is variation in mass per mass a consequence of true differences in the metal(s), or of extraneous variation in the total mass of particulates? Presenting the Nevada data as mass per mass could have introduced this uncertainty in the inter-town comparisons in the event of local dust events. Consequently, the authors opted against expressing the data as mass per mass.

An indication of the acceptance of each unit is their use in the scientific literature. Although precedence does not necessarily mean correctness, a consistent use of one unit over the other could be taken as a sign that a unit has been considered correct in a majority of cases. To get an indication of the dominant unit used in aerosol chemistry, peer-reviewed publications were found in a search of Science Citation Index using “total suspended particulates” and “metals” as keywords. Selected articles were similar in scope to the research in Fallon, namely, quantifying metals in total suspended particulates in urban or industrial settings. More than 90% of these articles presented their data as mass per volume (Table 1). Only one article used mass per mass exclusively, and one article used both units. Assuming that this sample of articles is representative of the population of research about metals in total suspended particulates, the dominant unit of expressing airborne metals is mass per volume, which the authors conformed with.

Nonetheless, the question has been raised: What would the Fallon results have looked like using mass per mass? At the regional scale, there is no difference between mass per volume versus mass per mass. For example, for the March collection, both

Table 1

Representative articles on metals in total suspended particulates, segregated by what unit was expressed

<i>Unit of mass per volume</i>	
1	Kan and Tanner (2005)
2	Rajput et al. (2005)
3	Shaheen et al. (2005)
4	Abdul-Wahab (2004)
5	Fang et al. (2004)
6	Gómez et al. (2004)
7	Guo et al. (2004)
8	Vanhoof et al. (2003)
9	Yaghi and Abdul-Wahab (2003)
10	García et al. (2002)
11	Ragosta et al. (2002)
12	Hršak et al. (2001)
13	Moreno-Grau et al. (2000)
14	Pfeifer et al. (1999)
15	Pirrone et al. (1995)
16	Pastuszka et al. (1993)
17	Foner and Ganor (1992)
18	Karue et al. (1992)
19	Trindade et al. (1981)
20	Morrow and Brief (1971)
<i>Unit of mass per mass concentration</i>	
1	de Kok et al. (2005)
<i>Both units</i>	
1	Fang et al. (2003)

Articles are sorted by year of publication.

data types show elevated W and Co in Fallon during the Lovelock, Fernley, and Reno sessions (Fig. 1). Both data types show low W and Co during the Yerington session. When merged together to increase sample depth, both data types show significantly higher W and Co within Fallon than all other comparison towns combined. Similarly for the November collection, both data types show elevated W and Co in Fallon during the Fernley, Yerington, and Reno sessions (Fig. 2). Especially notable, Fallon had very high values during the Fernley session, again shown by both data types. Both data types show very low W and Co during the Lovelock session. When merged together, both data types show significantly higher W and Co within Fallon than all other comparison towns combined.

This similarity in results across these two data types has precedence. For example, the article of Table 1 that presented both data types (Fang et al., 2003) also resulted in the same patterns of airborne metals for each data type.

In the case of Fallon, this similarity across data types is probably due to the absence of dust events

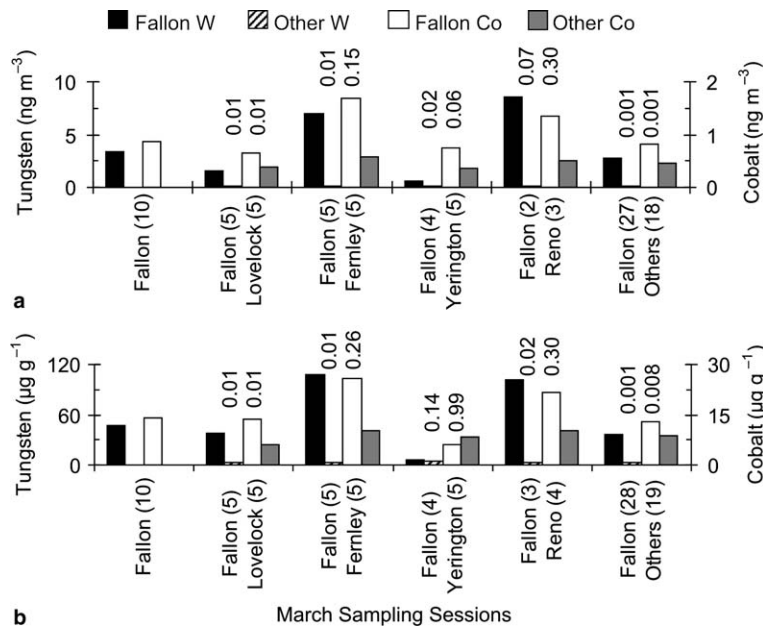


Fig. 1. Airborne W and Co concentrations within Fallon and other comparison towns for the March collection period: (a) results expressed in mass per volume units (ng m^{-3}), the same as Fig. 2a of Sheppard et al. (2006); (b) results expressed in mass per mass units ($\mu\text{g g}^{-1}$). In both graphs, sample sizes for each session-town are given in parentheses and p values of significance are given above the bars. Statistical analysis was the non-parametric Mann–Whitney test of no difference between median values, done as one-tailed tests with the alternative hypothesis that Fallon medians were higher than the comparison towns (Sokal and Rohlf, 1981).

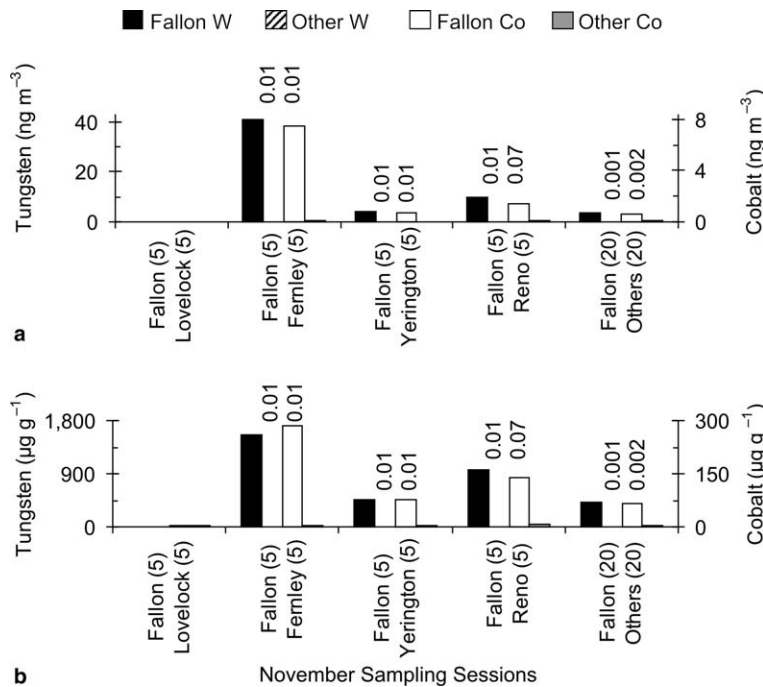


Fig. 2. Airborne W and Co concentrations within Fallon and other comparison towns for the November collection period: (a) results expressed in mass per volume units (ng m^{-3}), the same as Fig. 5a of Sheppard et al. (2006); (b) results expressed in mass per mass units ($\mu\text{g g}^{-1}$). In both graphs, sample sizes for each session-town are given in parentheses and p values of significance are given above the bars. Statistical analysis was the non-parametric Mann–Whitney test of no difference between median values, done as one-tailed tests with the alternative hypothesis that Fallon medians were higher than the comparison towns (Sokal and Rohlf, 1981).

that were big enough to have significantly affected one town but too small to have affected both towns of any given sampling session. The strategy of collecting total suspended particulates in two towns at the same time accounted for regional weather that could affect dust levels across west central Nevada. As expected, median total suspended particulate loadings rarely differed significantly between Fallon and comparison towns for any particular sampling session (Fig. 3). The only difference between towns occurred during the March Yerington session, when Fallon had significantly more total suspended particulates. This is curious because Fallon had low W and Co during that session (Fig. 1), which counters the intuition that simply more dust might lead to higher airborne W and Co. In short, correcting the W and Co data in mass per volume with total suspended particulate in mass per mass would not have changed the results at the regional scale.

At the local scale, total suspended particulates are even less likely to have varied significantly across sampling locations within Fallon. Dust can be kicked up at very local spatial scales due to things like pets and dirt roads (US EPA, 1999), but extraneous factors such as these were avoided as best as pos-

sible during the fieldwork of this research. Consequently, there was little variation in total suspended particulates across sampling sites within Fallon (Fig. 4a). Yes, sampling locations closest to the hard-metal facility did have higher total suspended particulates than locations more distant, but this difference was slight. Relative to the strong distance models for W and Co (Sheppard et al., 2006, Fig. 7, redone here as Fig. 4b), the same model for total suspended particulates is subtle, with a weak curve and a low R^2 indicating a low amount of shared variation. The sampling locations closest to the hard-metal facility also happen to be in the center of Fallon, and the spatial pattern of total particulates conforms to the typical pattern of urban areas being dustier than rural areas, largely because of automobile traffic and/or industrial activity (e.g., Gildemeister et al., 2005; Rösli et al., 2000; Zheng et al., 1997; Smith and Harrison, 1996; Monn et al., 1995; Menichini, 1992). By contrast, the strong falloff of W and Co with distance from the hard-metal facility is typical of patterns of air pollution from a point source (e.g., Sensen and Richardson, 2002; Makhholm and Bennett, 1998; Maserti and Ferrara, 1991; Bargagli et al., 1987; Lodenius and Laaksovirta, 1979; Steinnes and Krog, 1977).

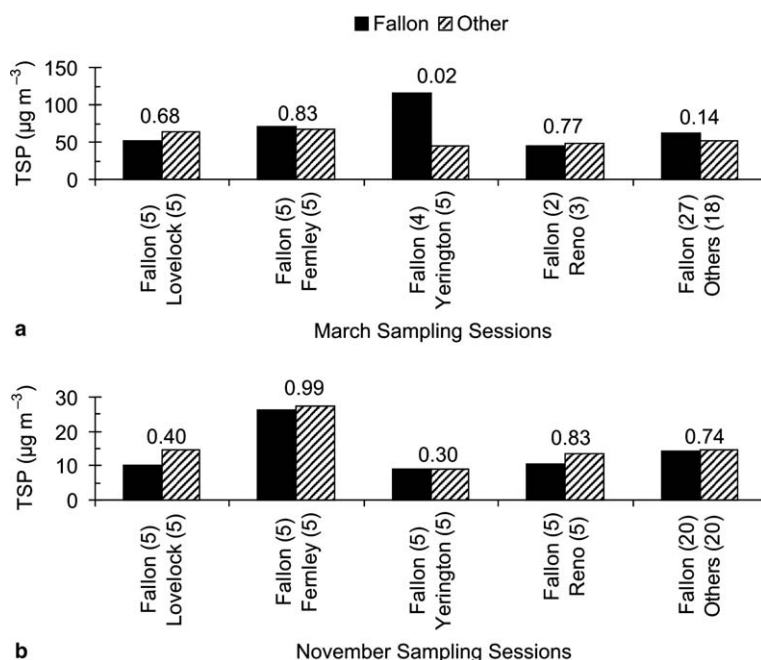


Fig. 3. Airborne total suspended particulates ($\mu\text{g m}^{-3}$) within Fallon and other comparison towns: (a) the March sampling sessions; (b) the November sampling sessions. In both graphs, sample sizes for each session-town are given in parentheses and p values of significance are given above the bars. Statistical analysis was the non-parametric Mann–Whitney test of no difference between median values, done as two-tailed tests with the alternative hypothesis that Fallon medians were different from the comparison towns (Sokal and Rohlf, 1981).

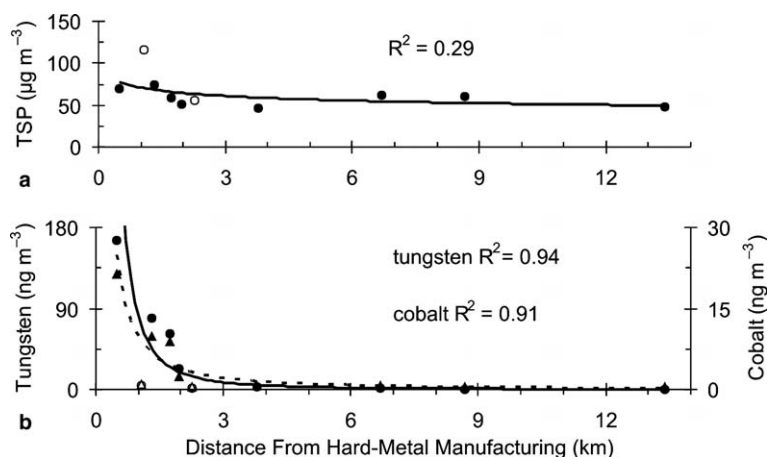


Fig. 4. Airborne loadings within Fallon as a function of distance from the hard-metal facility: (a) tungsten (circles) and cobalt (triangles) loadings (ng m^{-3}) with fit lines calculated excluding the two outlying low values (open symbols), which are from sample locations southwest of the hard-metal facility, the same as Fig. 7 of Sheppard et al. (2006); (b) total suspended particulates ($\mu\text{g m}^{-3}$) with a fit line calculated using all values, including the two outlying values of (a) (open circles).

Additionally, one of the W–Co negative outliers (Fig. 4b) is also an outlier for total suspended particulates (Fig. 4a), but curiously it is a positive outlier for total suspended particulates. This further counters the intuition that simply more dust might lead to higher W and Co, and it also exemplifies the ambiguity that can arise from expressing these data as mass per mass. The authors stand by the use of mass per volume as the correct unit of expression for the data.

3. Normalizing total suspended particulate data to a conservative trace element

Many other elements were measured besides W and Co, but none of them showed consistent differences or patterns between Fallon and comparison towns (Sheppard et al., 2006, p. 155 for the March collection and p. 156 for the November collection). These other elements combined to establish the background expectation of no unusual or consistent difference in airborne metals of Fallon compared to nearby towns, and as such they served the function of a conservative trace element for this project.

The only other notable element besides W and Co was high Cu in Yerington during the November collection period. This finding is explainable as a result of Yerington's location at the base of a Cu mine pit and its tailings pile. This finding did not necessarily perform any normalizing role, but it did provide a proof-of-concept that high-volume sampling of total suspended particulates could suc-

cessfully identify unusual airborne metals and accurately identify at least a candidate source for them.

4. Natural soil chemistry as an alternative hypothesis

Prior to the initiation of this research on total suspended particulates of towns of west central Nevada, the US Agency for Toxic Substances and Disease Registry had evaluated soils research done previously in and around Fallon (US ATSDR, 2003a). Analysis of 79 residential soil samples collected in Fallon in 2001 by the Nevada Division of Environmental Protection showed Co to be below the screening value and therefore not a contaminant of concern; W was not mentioned. Analysis of 431 surface soil samples collected throughout Churchill County in support of various US Geological Survey programs also showed typical levels for all analyzed metals to be below screening values; neither W nor Co was emphasized specifically, but presumably they were among the metals measured. The ultimate conclusion was that contaminant levels found in residential and non-residential soils in and around Fallon are not a public health concern (US ATSDR, 2003a, p. 20). This can be considered as rejecting, at least for the moment, the proposed alternative hypothesis that Nevada's natural soil chemistry is causing Fallon's unique airborne W and Co.

Rather than duplicating these soils studies, the authors chose to study total suspended particulates because prior work on this environmental topic in and around Fallon was not extensive. A total

suspended particulate study during 2002 used two high-volume samplers to collect at a site in Fallon and a site just outside of Fallon for seven 1-day sampling sessions (US ATSDR, 2003b). Tungsten was not measured in those particulate samples, as W had not yet emerged by that time as a contaminant of concern. Cobalt was measured, and the data show that Co (presented with the same units used by the authors, ng m^{-3}) was on average 5.1 times higher within Fallon than outside of Fallon (US ATSDR, 2003b, p. 59). In spite of that, Co was not emphasized in the text, and the ultimate conclusion was that no association between air pollutants and leukemia had been identified (US ATSDR, 2003b, p. 25). No further recommendations were made, but it was felt that more extensive research on total suspended particulate chemistry was justified. To accomplish this, 10 high-volume samplers were used to collect at five locations in Fallon and five locations in multiple comparison towns, and then the entire project was repeated during a different time of year using completely different field equipment (Sheppard et al., 2006).

5. Identifying tungsten in Fallon's air

For other metals with known health effects, it has not been imperative to know the exact species of exposure to raise concern. For example, Hg exists in the environment in its elemental form and as various salts (Bidstrup, 1964), and all of these forms can become available for methylation into organic methylmercury (MeHg), the common form of Hg poisoning (US ATSDR, 1992b). Similarly, Pb exists in the environment in its elemental form, as various salts, and in organic forms (Lansdown and Yule, 1986; US ATSDR, 1992a), but reference levels of

airborne Pb do not specify particular species of Pb (US EPA, 1984). By extension, it would seem worthwhile and useful to first quantify airborne W regardless of what specific kind of W it might be.

Nonetheless, the question has been raised: Is there any additional information about the particular kind of W collected in the filters? As suggested, scanning electron microscopy can speak to this question, and the authors did have an SEM image and associating X-ray spectrum of a W particle from one of the Fallon filters, specifically from the sampling location closest to the hard-metal facility. Physically, this particle is small (average diameter of $1.1 \mu\text{m}$), circular (a long-to-short axis ratio of $1.2\text{--}1 \mu\text{m}$), and smooth (Fig. 5a). Chemically, this particle exhibits a strong peak for W along with minor peaks for Co, which is used as a binder for tungsten carbide (Fig. 5b). Missing from this X-ray spectrum are peaks for Ca and Mn, the cations associated with scheelite (CaWO_4) and heubnerite (MnWO_4), which are the common natural minerals of W in Nevada (Stager and Tingley, 1988).

For comparison purposes, reference SEM images of representative scheelite and heubnerite particles show them to be bigger and more angular, and their X-ray spectra contain peaks for Ca or Mn (McCrone and Delly, 1973). An image of generic metal grinding dust includes small, spherical particles (McCrone et al., 1967). Thus, physical and chemical characteristics of the one Fallon particle suggest that it probably is not scheelite or heubnerite and that it could be a metal grinding.

This particle analysis was not presented in Sheppard et al. (2006) because it is not conclusive, being of only one particle. A more extensive SEM analysis of W particles from all of the sampling locations is clearly merited, and that is currently

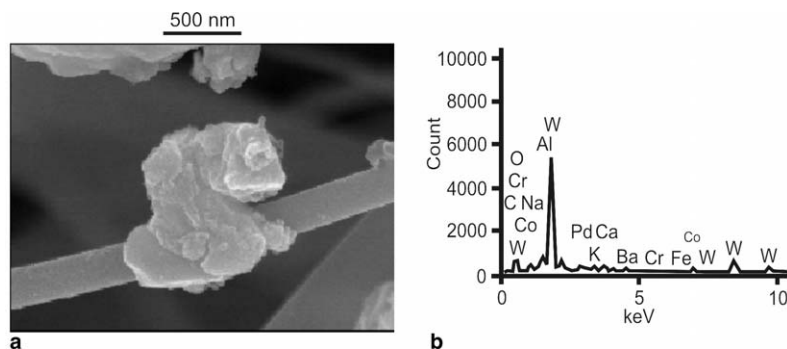


Fig. 5. (a) Scanning electron microscope image of a particle from one of the filters within Fallon; (b) X-ray spectrum of that particle showing a clear peak in counts at the characteristic emission for W.

being done. This would shed light on the association of the finding of elevated W and Co in Fallon with the assessment that Co and tungsten carbide are possibly carcinogenic (IARC, 2003).

6. Additional responses

All environmental research in Fallon is a serious matter. To reiterate, the cluster of childhood leukemia in Fallon has been dubbed, “one of the most unique clusters of childhood cancer ever reported” (Steinmaus et al., 2004, p. 768 and repeated on p. 770). Although causes of health clusters have been notoriously difficult to determine (Rothman, 1990), a reasonable response to this extraordinary public health event is to search for a cause, including possibly an environmental cause, and to that end a great deal of environmental research has been done in Fallon. The main objective of Sheppard et al., 2006, p. 153 was to test for something distinctive in outdoor airborne dust of Fallon, something that could become the focus of subsequent biomedical research related to leukemia. That objective was met: Fallon, or more precisely parts of Fallon, can have unusually elevated airborne W and Co, and this finding is equally true with either mass per volume or mass per mass as the unit of expression.

The secondary objective of Sheppard et al., 2006, p. 153 was to identify a candidate source of the distinctive dust chemistry, and this objective was also met. To clarify, the authors pointedly did not conclude definitively that the hard-metal facility in Fallon is the source of Fallon's airborne W and Co (Sheppard et al., 2006, p. 162). Instead, it was concluded that the hard-metal facility could tentatively be considered as a candidate source (Sheppard et al., 2006, p. 161). Sheppard et al. (2006) was not first to make this suggestion, as the Nevada Division of Environmental Protection intuited as much in 2003 (Reno Gazette-Journal, 5 February 2003). More than intuition, the current research shows spatial patterns of airborne W and Co that point to at least the location of the hard-metal facility as a potential candidate source area.

7. Conclusion

The final conclusion of Sheppard et al. (2006) was the ever-familiar call for more research, namely to continue testing for elevated airborne W and Co within Fallon and to definitively identify their source (Sheppard et al., 2006, p. 163). Curiously,

the Nevada Division of Environmental Protection has since announced that it has no plans to investigate in Fallon (Lahontan Valley News, 3 December 2005). In spite of this, the authors still call for more research, and the hard-metal facility has also welcomed more research in Fallon (Reno Gazette-Journal, 25 November 2005). In particular, new research should explore different types of environmental evidence besides what has been tried so far, as analysis of multiple, independent lines of evidence is a powerful strategy in environmental science (Reid and Thompson, 1996; Reimann and de Caritat, 2005). Upon accumulating multiple and corroborating lines of evidence, each one well replicated, environmental features found to be distinctive in Fallon might then be tested biomedically for possible linkages to leukemia. In this fashion, progress might be made in better understanding Fallon's cluster of childhood leukemia.

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