Chapter 5

# MULTIPLE TECHNIQUES FOR RESEARCHING AIRBORNE PARTICULATES: A COMPREHENSIVE CASE STUDY OF FALLON, NEVADA

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#### ABSTRACT

Airborne particulates are an important public health concern. Because of toxic effects of airborne particulates, they are monitored regularly for total loading and chemical constituents. Monitoring includes various techniques to measure airborne particulates. The primary objective of this paper is to describe, compare, and contrast airborne particulate techniques, both theoretically and in practice. Many of these techniques have been applied recently in a single location–Fallon, Nevada–where a cluster of childhood leukemia has been ongoing since 1997. This offers an opportunity to assess advantages and disadvantages of airborne particulate techniques.

Techniques: Total suspended particulate (TSP) chemistry is the filtering of air to capture airborne dust for measurement and interpretation of elements. Surface dust chemistry is the measurement and interpretation of elements in fine sediments that accumulate on outdoor surfaces. Lichen chemistry is the measurement and interpretation of elements in or on lichens. Leaf chemistry is the measurement and interpretation of elements in or on leaves of trees and other plants. Bark chemistry is the measurement and interpretation of elements on the surface of bark of trees and other plants. Dendrochemistry is the measurement and interpretation of chemical elements in tree rings.

Fallon: TSP chemistry showed elevated tungsten and cobalt in airborne particulates of Fallon compared to other towns of west central Nevada. Morphological and chemical

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testing of Fallon airborne tungsten particles showed them to be anthropogenic in origin, not natural. Surface dust chemistry and leaf surface chemistry showed peaks of tungsten and cobalt just northwest of the center of town. Lichen tissue chemistry showed elevated airborne tungsten and cobalt within Fallon compared to outlying desert. Dendrochemistry showed environmental availability of tungsten increasing in Fallon during the mid 1990s, coinciding roughly with the onset of the cluster of childhood leukemia in that town.

No single technique for measuring airborne particulates is perfect. Consequently, it would be best to apply multiple techniques in order to capitalize on advantages and offset disadvantages, as illustrated by this comprehensive study of airborne particulates in Fallon.

#### INTRODUCTION

Airborne particulates are an important public health concern. Outdoor airborne particulates are omnipresent, even in rural areas where air might appear to be clean relative to urban settings (Holmes, 2001). Inhalation of airborne particulates can have many toxicological effects on humans. For example, excessive inhalation of dust can lead to various respiratory ailments (Lippman 2000). Additionally, inhalation of trace elements contained in airborne particulates potentially leads to other illnesses (Costa 2000). A notable example of this is inhalation of lead resulting in increased body burdens of lead (Nriagu 1980), causing a litany of health effects (Singhal and Thomas 1980). Because of toxic effects of airborne particulates, major programs exist to regularly monitor total loading of airborne particulates and their chemical constituents (e.g., U.S. EPA 2008).

As part of these monitoring programs, various techniques exist to measure airborne particulates. The most obvious technique is filtering dust directly from air using regulated vacuum devices (Baron and Willeke 2001). Many other techniques employ less direct means to establish at least relative loadings of elements in airborne dust. The primary objective of this paper is to describe, compare, and contrast airborne particulate techniques, both theoretically and in practice. Many of these techniques have been applied recently in a single location–Fallon, Nevada (Figure 1a)–offering an opportunity to assess their relative advantages and disadvantages for studying airborne particulates.

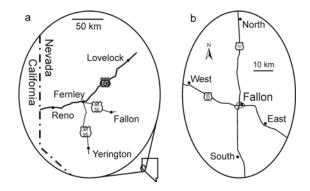


Figure 1.Maps of (a) west central Nevada showing Fallon and comparison towns where TSP samples were collected, and (b) Fallon and surrounding desert where lichens were collected. In (a), other towns besides Fallon were comparison towns for TSP and tree rings. In (b), locations marked outside of Fallon were sample sites for lichens in outlying desert.

Technique	Field Work	Field Expense	Lab Procedures	Indication of Particulates	Temporal Resolution	Multiple Time Steps	Spatial Resolution	Characterize Particles
total suspended particulates	difficult	high	detailed	direct	hourly to daily	no	low	yes
surface dust	very easy	very low	detailed	direct	daily to weekly	no	high	yes
lichen tissues	moderate	moderate	moderate	indirect	annual to multidecadal	possibly	potentially high	no
lichen surfaces	moderate	moderate	moderate	intermediate	weekly to monthly	no	mixed	possibly
leaf tissues	easy	low	easy	indirect	weekly to seasonal	no	high	no
leaf surfaces	easy	low	easy	intermediate	weekly to seasonal	no	high	possibly
bark surfaces	moderate	moderate	moderate	intermediate	annual to multiannual	no	high	possibly
tree rings	difficult	moderate	very detailed	indirect	annual to centennial	yes	low	no

Table 1. Characteristics of techniques for researching airborne particulates	s.
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## **AIRBORNE PARTICULATE TECHNIQUES (TABLE 1)**

#### **Total Suspended Particulates**

Total suspended particulate (TSP) chemistry is the filtering of air to capture airborne dust for measurement and interpretation of elements (Solomon et al. 2001). Advantages of TSP are (1) particulate chemistry is determined directly, i.e., without intermediate incorporation of elements by indirect monitors, which makes interpretation of results straightforward, and (2) collected particles can be removed from filters and characterized morphologically and chemically. Disadvantages are (1) field collectors are expensive and cumbersome to deploy, (2) laboratory procedures are detailed and include many steps, (3) individual filters represent only one time interval, and (4) spatial resolution is typically low, largely due to the expense of collectors. Temporal resolution of TSP ranges from hourly to daily, which is on the highfrequency end of the spectrum for airborne particulate techniques. Case studies exist worldwide using TSP chemistry to quantify atmospheric loading of heavy metals (e.g., Trindade et al. 1981; Foner and Ganor 1992; Karue et al. 1992; Pastuszka et al. 1993; Pirrone et al. 1995; Moreno-Grau et al. 2000; Ragosta et al. 2002; Guo et al. 2004; Rajput et al. 2005).

#### **Surface Dust**

Surface dust chemistry is the measurement and interpretation of elements in fine sediments that accumulate on outdoor surfaces (Duggan and Inskip 1985). Advantages of surface dust are (1) particulate chemistry is determined directly, (2) collected particles can be isolated and characterized morphologically and chemically, (3) field work is very easy and inexpensive, and (4) spatial resolution can be high, largely due to the ease of field collection. Disadvantages are (1) laboratory procedures are detailed and include many steps, and (2) individual samples represent only one time interval. Temporal resolution of surface dust ranges from daily to weekly depending on frequency and intensity of rain storms. Case studies exist worldwide using surface dust chemistry to quantify atmospheric loading of heavy metals and/or identify spatial patterns of deposition of airborne particulates (e.g., 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).

#### Lichens

Lichen chemistry is the measurement and interpretation of elements in or on lichens (Garty 1993), which are symbiotic associations of fungi and algae (Baron 1999). Lichens lack true roots, thereby diminishing the influence of growth substrates on their chemical composition (Szczepaniak and Biziuk 2003; Wolterbeek 2002). Lichens also lack a cuticle, allowing easy incorporation of atmospheric constituents directly into their tissues (Conti and Cecchetti 2001; Falla et al. 2000). An advantage of lichen chemistry is its potentially high

spatial resolution: Lichens are widespread geographically (Richardson 1992, 1995), colonizing tree bark or rock surfaces practically everywhere (Wolterbeek and Bode 1995). Even if lichens happen not to be growing in a study site of interest, they can be transplanted to serve as biomonitors of airborne particulates (Conti et al. 2004). It is also theoretically possible to measure multiple times steps from a single lichen, as inner parts of lichens are older than outer edges (Hale 1983), though this feature of lichen growth is not exploited much in lichen chemistry. Disadvantages of lichens are that field and laboratory work is moderately detailed and/or expensive. The indication of airborne particulates from lichen tissue is indirect, but rinsing tissues and analyzing rinse solutions is a more direct version of lichen surface chemistry. Similarly, particles are not easily characterized morphologically and chemically from lichen tissue, but characterization might be possible from lichen surface rinse samples. Temporal resolution of lichen tissue chemistry ranges from annual to multidecadal (Sensen and Richardson 2002), as lichens can live for decades (Bull et al. 1994). Temporal resolution of lichen surface chemistry is weekly to monthly depending on frequency and intensity of rainfall. Case studies exist worldwide using lichen chemistry to monitor or assess airborne elements (e.g., Jeran et al. 1996; Scerbo et al. 1999; Chiarenzelli et al. 2001; Zhang et al. 2002; Zschau et al. 2003; Bennett and Wetmore 2003; Loppi and Pirintsos 2003; Nash et al. 2003; Yun et al. 2003; Yenisoy-Karakaş and Tuncel 2004; Cuny et al. 2004; Helena et al. 2004; Freitas and Pacheco 2004).

#### Leaves

Leaf chemistry is the measurement and interpretation of elements in or on leaves of trees and other plants (Bargagli 1993). Advantages of leaf chemistry are (1) spatial resolution is high, especially in urban areas where trees are typically heavily planted for landscaping, and (2) field and laboratory work is easy and inexpensive. A disadvantage is that samples represent only one time interval. The indication of airborne particulates from leaf tissue is indirect, but rinsing leaves and analyzing rinse solutions is a more direct version of leaf surface chemistry. Particles are not easily characterized morphologically and chemically from leaf tissue, but characterization might be possible from leaf surface rinse samples. Temporal resolution of leaf samples ranges from weekly to seasonal to perhaps multiannual, depending on rainfall frequency and whether the tree species studied are deciduous or evergreen. Many case studies exist worldwide using leaf chemistry to quantify atmospheric loading of heavy metals and/or identify their spatial patterns (e.g., Ward et al. 1977; Dasch 1987; Aksoy and Öztürk 1997; Alfani et al. 1996; Aksoy et al. 1999; Gupta et al. 2004; Salve et al. 2006; Rossini Oliva and Mingorance 2006).

#### Bark

Bark chemistry is the measurement and interpretation of elements on the surface of bark of trees and other plants (Walkenhorst et al. 1993). Because of the nature of bark growth, airborne particulates are found mostly on bark surfaces, less so in bark tissue itself (Hampp and Höll 1974). An advantage of bark chemistry is high spatial resolution, especially in urban areas that are landscaped. Disadvantages are (1) field and laboratory work is moderately difficult and expensive, and (2) bark samples represent only one time interval. Temporal resolution of bark surface chemistry ranges from annual to multiannual depending on how fast bark sloughs off from the tree species studied. Case studies exist worldwide using bark chemistry to quantify airborne particulates (e.g., Lötschert and Köhm 1978; Kakulu 2003; Mandiwana et al. 2006; Ward et al. 2006).

#### **Tree Rings**

Dendrochemistry is a subdiscipline of dendrochronology, the dating and interpretation of tree rings to reconstruct and analyze past environmental conditions (Fritts 1976). In dendrochemistry, the measurement of primary interest is chemical elements, or suites of multiple elements, contained in individual growth rings or groups of multiple rings (Amato 1988; Smith and Shortle 1996). The principal advantage of dendrochemistry is that relative environmental availability of elements can be reconstructed for multiple intervals back in time, making tree rings unique amongst techniques for studying airborne particulates. Temporal resolution can range from annual to centennial, depending on ages of trees sampled. Disadvantages are (1) fieldwork is difficult and expensive, (2) laboratory preparations are very detailed, including overcoming potential contamination from metal sampling tools (Sheppard and Witten 2005), (3) determination of airborne particulate chemistry is indirect, (4) characterizing particle morphology and chemistry is not possible, and (5) spatial resolution is usually low, largely due to the difficulty of field sampling. Even with these disadvantages, dendrochemistry has been done in a wide array of applications in environmental science, including for heavy metals such as Pb (e.g., Hagemeyer and Weinand 1996), Ni (e.g., Yanosky and Vroblesky 1992), Cd (e.g., Guyette et al. 1991), and Hg (e.g., Zhang et al. 1995).

## **FALLON, NEVADA**

Fallon is a small, rural, farming community (Greater Fallon Area Chamber of Commerce 2008) located in west central Nevada (Figure 1a). 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).

A cluster of childhood leukemia has been ongoing in Fallon 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 ~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 2003a), outdoor air (U.S. ATSDR 2003b), surface soil and indoor dust (U.S. ATSDR 2003c), potential lingering effects of underground nuclear bomb testing in the area (Seiler 2004), and groundwater (Seiler et al. 2005).

Inhalation of dust (airborne particulates) can be a particular health concern for children, who often come in close contact with dust and soil when playing (U.S. ATSDR 2003c). Consequently, TSP chemistry of outdoor air has also been tested in and around Fallon (U.S. ATSDR 2003b). However, early air testing deployed just two high-volume samplers, one in the center of town and one located several kilometers west of town, exemplifying one of the disadvantages of TSP, i.e., low spatial resolution. That was not enough sampling to determine spatial patterns of airborne particulates, and ultimately no association was found between airborne particulate chemistry in Fallon and the childhood leukemia cluster (U.S. ATSDR 2003b, page 25). Because that sampling regimen was sparse, outdoor airborne particulates of Fallon were studied again with a more comprehensive, multi-technique approach. To date, TSP, surface dust, lichens, leaf surfaces, and tree rings have been analyzed in Fallon as well as in nearby comparison towns and from surrounding outlying desert. The following section compares results from these airborne particulate techniques used in Fallon.

Technique	Number of Samples	Number of Days Sampling	Unit of Measurement	Range of Values W	Range of Values Co	Source
total suspended particulates	83	29	ng metal m <sup>3</sup> air	<1 – ~40	<1 – ~8	Sheppard et al. 2006
surface dust	125	3	<u>μg metal</u> g dust	<1 – 934	<1 – 98	Sheppard et al. 2007a
lichen tissues	30	3	<u>μg metal</u> g lichen	<1 – ~40	<1 – ~9	Sheppard et al. 2007b
leaf surfaces	95	2	<u>μg metal</u> g leaf	<1 – ~17	<1 – ~6	Sheppard et al., in press
tree rings	100s	10	<u>ng metal</u> g wood	~40 - ~170	~80 - ~120	Sheppard et al. 2007c

#### Table 2. Summary of results from airborne particulate techniques used in Fallon, Nevada.

## **AIRBORNE PARTICULATES IN FALLON (TABLE 2)**

#### **Total Suspended Particulates**

Detailed methodology of our TSP study in Fallon is described elsewhere (Sheppard et al. 2006). Based on an extensive collection of 83 samples spanning 29 sampling days in Fallon as well as in nearby comparison towns (Figure 1a), tungsten and cobalt can be elevated in airborne particulates of Fallon compared to other towns of west central Nevada. Tungsten and cobalt varied in Fallon airborne particulates both temporally and spatially, with maximum values many times higher than minimum values. Capitalizing on the advantage of TSP that particles can be removed from filters and characterized morphologically and chemically, airborne tungsten particles of Fallon were mostly small (< 5  $\mu$ m in diameter) and evenly sized, and they were composed mostly of pure tungsten or a mixture of tungsten with cobalt and/or other metals (Sheppard et al. 2007d). As such, airborne tungsten particles of Fallon were anthropogenic in origin, not natural.

The high variability in TSP chemistry results provoked the use of other techniques for studying airborne particulates to better understand airborne tungsten and cobalt in Fallon.

#### **Surface Dust**

Detailed methodology of our surface dust study in Fallon is described elsewhere (Sheppard et al. 2007a). Based on an extensive collection of 125 samples located throughout and around Fallon, tungsten and cobalt showed major peaks in surface dust just northwest of the center of town. Peak tungsten and cobalt values were much higher than background values measured from the outskirts of Fallon. Based on this technique, the source area of airborne tungsten and cobalt was fine-tuned to just northwest of the center of town.

#### Lichen Tissues

Detailed methodology of our lichen tissue study in Fallon is described elsewhere (Sheppard et al. 2007b). Based on a collection of 10 separate lichens growing at one site in Fallon and 20 separate lichens growing in four different locations outside of town (Figure 1b), airborne tungsten and cobalt were elevated within Fallon compared to outlying desert. Tungsten and cobalt were not elevated in the rock substrates of the sampled lichens, so the source of these two metals to lichens within Fallon must have been airborne. Furthermore, natural desert of west central Nevada did not seem to be the source of elevated airborne tungsten and cobalt within Fallon.

### Leaf Surfaces

Detailed methodology of our leaf surface study in Fallon is described elsewhere (Sheppard et al., in press). Based on an extensive collection of 95 samples located throughout and around Fallon, airborne tungsten and cobalt concentrations showed major peaks just northwest of the center of town. Peak tungsten and cobalt values were much higher than background values measured from the outskirts of Fallon. Based on this technique, the source area of airborne tungsten and cobalt was confirmed to just northwest of the center of town.

#### **Tree Rings**

Detailed methodology of our tree-ring study in Fallon is described elsewhere (Sheppard et al. 2007c). Based on an extensive collection of many trees sampled in Fallon as well as in nearby comparison towns, with each tree having tens of growth rings, environmental availability of tungsten began increasing in Fallon during the mid 1990s, coinciding roughly with the onset of the cluster of childhood leukemia in that town. Cobalt has been elevated in tree rings of Fallon relative to trees in nearby comparison towns throughout this time period.

## CONCLUSION

No single technique for measuring airborne particulates is perfect. Each technique has advantages and disadvantages. Because of this, applying only one technique in research of a particular location might not be fully indicative of and/or convincing about airborne particulates in the study area. Consequently, it would be best to apply multiple techniques in order to capitalize on advantages and offset disadvantages. In general, corroboration across multiple techniques would be more convincing than relying on results of any one technique (Reid and Thompson 1996; Reimann and de Caritat 2005).

All five airborne particulate techniques used so far in Fallon have corroborated one another. Tungsten and cobalt were elevated in airborne particulates of Fallon compared to nearby towns (total suspended particulates) and to outlying desert (lichen tissues). Airborne tungsten and cobalt showed peaks near the center of Fallon compared to the outskirts of town (surface dust and leaf surfaces). Environmental availability of tungsten increased in Fallon during the mid 1990s, coinciding roughly with the onset of the cluster of childhood leukemia (tree rings). Morphological analysis of actual tungsten particles from TSP filters showed them to be anthropogenic in origin, not natural (particle analysis).

This comprehensive study of airborne particulates in Fallon capitalized on the advantages of each technique used. TSP gave a direct loading of airborne tungsten and cobalt (mass of metal per volume of air sample), and it allowed for morphological and chemical characterization of airborne tungsten particles. Lichen chemistry allowed for comparing airborne particulates between Fallon and outlying desert. Surface dust and leaf surface chemistry allowed for fine-scale spatial mapping of airborne tungsten and cobalt within Fallon. Dendrochemistry gave a time series of relative environmental availability of tungsten and cobalt. As has been stated in other published environmental research of ours pertaining to Fallon, it cannot be concluded from only environmental data 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. 2003) and that Fallon is distinctive environmentally by its elevated airborne tungsten and cobalt as determined using multiple airborne particulate techniques, 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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