Historical ambient airborne asbestos concentrations in the United States – an analysis of published and unpublished literature (1960s–2000s)

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Abstract
Outdoor concentrations of airborne asbestos have been measured throughout the US over time. However, a thorough review and analysis of these data has not been conducted. The purpose of this study is to characterize asbestos concentrations in ambient air by environment type (urban, rural) and by decade, using measurements collected in the absence of known asbestos emission sources. A total of 17 published and unpublished studies and datasets were identified that reported the results of 2058 samples collected from the 1960s through the 2000s across the US. Most studies did not report asbestos fiber type, and data based on different analytical methods (e.g. Phase Contrast Microscopy, Transmission Electron Microscopy, etc.) were combined in the dataset; however, only fibers ≥5 μm in length were considered. For a small subset of the measurements (n = 186, 9.0%), a conversion factor was used to convert mass-based data (e.g. ng/m³) to count-based values (i.e. f/cc ≥5 μm). The estimated overall mean and median ambient asbestos concentrations for the 1960s through 2000s were 0.00093 f/cc and 0.00022 f/cc, respectively. Concentrations generally increased from the 1960s through the early 1980s, after which they declined considerably. While asbestos use decreased throughout the 1970s, these results indicate that ambient concentrations peaked during the early 1980s, which suggests the possible contribution of abatement or demolition activities. Lastly, ambient asbestos concentrations were higher in urban than rural settings, which is consistent with the greater use of asbestos-containing materials in more densely populated areas.

Keywords
Ambient levels, asbestos, fibers, outdoor concentrations

Introduction
Between the 1960s and the late 2000s, outdoor airborne asbestos fiber concentrations have been measured in both rural and urban environments in the US. Although the published literature contains a reasonably large number of datasets containing information on ambient asbestos, a thorough review and analysis of these data has not been conducted. The purpose of this study was to characterize historical to present day measurements of asbestos in outdoor air throughout the US, using measurements that were collected in the absence of known or potential asbestos emission sources. The data were also evaluated for temporal and spatial trends.

Background
Asbestos is ubiquitous in the environment in the US (ATSDR, 2001). Sources of this asbestos are primarily related to anthropogenic activities, such as mining, milling, manufacturing and use of asbestos-containing materials (ACM), and transport and disposal of associated waste. Furthermore, in some areas of the US, concentrations of airborne asbestos are due to weathering of naturally occurring asbestos (NOA) in seams of exposed rock (ATSDR, 2001; IARC, 2012; NTP, 2014; U.S. EPA, 2010).

In the US, the use of asbestos has decreased dramatically from its peak in the mid-1970s (Figure 1) (Virta, 2002, 2006, 2010, 2014), mainly as a result of regulations promulgated as a response to concerns regarding health hazards associated with occupational exposures. As a consequence, emissions of asbestos to the environment have decreased; however, the presence of and activities involving ACM likely continue to affect the ambient air concentrations of asbestos to the present day (ATSDR, 2001; NTP, 2014; Wylie & Candela, 2015). For example, as noted by Wylie & Candela (2015), between 2003 and 2013, an average of 6500 tons of “friable” asbestos was disposed of or otherwise released to the environment annually in the US (U.S. EPA, 2015). Currently, there are no federal standards for outdoor air concentrations of asbestos to which the general public is exposed each day.

Naturally occurring asbestos
Weathering of NOA and anthropogenic activities, such as excavation, agriculture, mining and road construction can
result in the release of measurable concentrations of asbestos fibers in the environment (Gunter et al., 2007; Hendrickx, 2009; Van Gosen, 2007; Walton, 1982). At least 35 states in the US have reported findings of NOA, with the major areas of concern located along the Appalachian Mountains and in the Western Cordillera (Harper, 2008; U.S. EPA, 2008). Additionally, large areas of exposed ultramafic bedrock in northern California, some now densely populated by housing and infrastructure, have become the focus of attention after they were found to contain chrysotile and tremolite-actinolite asbestos (Churchill & Hill, 2000; Clinkenbeard et al., 2002; Lee et al., 2008; Ross & Nolan, 2003). Furthermore, Wylie & Candela (2015) recently estimated that in the US, 400 tons of amphibole asbestos is released annually through weathering of naturally occurring minerals.

**Mining and milling**

In the US, asbestos was mined commercially beginning in 1900, and mining occurred over time in 15 different states, with the largest production occurring in Arizona, California, North Carolina and Vermont (Virta, 2006). Asbestos production reached its maximum in 1973, after which it declined rapidly until the last mine closed in 2002 (Van Gosen, 2007; Virta, 2006). The vast majority of the total cumulative asbestos production in the US, as well as most imported materials, was chrysotile, whereas amphibole asbestos was imported to the US mainly from South Africa and Australia (Ross & Virta, 2001; Virta, 2002, 2006). However, some of the early production in the US was amphibole asbestos, and while such mines were most often short-lived, anthophyllite asbestos was mined in North Carolina from the early 1930s until 1979 (Virta, 2006). The control of emissions from mining and milling operations was reportedly often poor; however, very little quantitative data characterizing airborne asbestos concentrations in the ambient air near these operations exist (Gibbs & Du Toit, 1973; Harwood & Blaszak, 1974).

**Manufacturing operations**

According to the US Environmental Protection Agency (EPA), manufacture of ACM occurred in at least 22 states (U.S. EPA, 1974a,b). Predominant asbestos-containing products historically manufactured in the US include cement pipe and sheets, and flooring, friction and roofing products, which together made up about 60 to 70% of the country’s total annual demand for asbestos (USBM, 1976, 1985). Amid rising health concerns and associated regulations, asbestos use in the manufacture of commercial products in the US declined from a peak of about 800,000 metric tons in 1973 to 217,000 in 1983, and subsequently to 33,000 in 1992 (U.S. EPA, 1993b; USBM, 1976, 1985). Estimates of US asbestos consumption for recent years indicate that the decreasing trend has continued, with industry demand falling from 2,230 metric tons in 2006 to 772 in 2013 (Virta, 2008, 2010, 2014).

Emissions of asbestos fibers from manufacturing operations occurred due to inadequate controls, through accidental releases, and from handling, transporting, and storing waste materials (Bruckman & Rubino, 1975; Harwood & Blaszak, 1974; U.S. EPA, 1981). Specifically, prior to regulations instituted in the early 1970s waste piles were often located in or near densely populated areas and were frequently kept uncovered, resulting in uncontrolled emissions of asbestos to the ambient environment (Harwood & Blaszak, 1974).

**Use of asbestos-containing products**

Asbestos has been incorporated in various products because of its low cost and desirable qualities, such as heat and fire resistance, wear and friction characteristics, tensile strength, heat, electrical and sound insulation, adsorption capacity, and resistance to chemical and biological attack (ATSDR, 2001). Asbestos-containing products utilized in construction, industry, commerce and in the military can be found in encapsulated and friable forms. Encapsulated products, such as wallboard, ceiling tile, molded phenolic materials, roofing, piping and floor tile, contain asbestos fibers that are bound in a matrix such that they do not spontaneously release fibers unless the materials are disturbed (Lee & Van Orden, 2008; Michaels & Chissick, 1979; U.S. EPA, 1985). However, damage to some encapsulated ACM from severe weather, chemicals and mechanical forces (i.e. cutting, drilling, sanding or breaking) may result in release of fibers (U.S. EPA, 1985, 1990). Conversely, by definition, friable materials contain more than 1% asbestos and can be crumbled, pulverized or reduced to powder by hand pressures (U.S. EPA, 1985). Examples of friable products include sprayed or troweled-on materials on ceilings, walls and other surfaces (e.g. for decoration, fireproofing, or heat and sound insulation), as well as insulation on pipes, tanks, ducts and other equipment (Mangold, 1983). The application or use of such products was widespread from the 1950s to the mid-1970s (Mangold, 1983; U.S. EPA, 1987).
Friction materials

Beginning in the early 1900s, asbestos was used in the manufacture of automotive friction products (i.e. brakes and clutches); from 1965 to 2001, an estimated 1.4 million tons of chrysotile asbestos were used in friction products in the US (Paustenbach et al., 2003, 2004; Virta, 2006). Although wear debris from these products could be released into the environment during the high temperature and mechanical forces of the braking and clutching processes, asbestos fibers in friction materials degraded into forsterite, leaving only a very small percentage (<1%) of the chrysotile fibers intact. Therefore, the contribution to ambient concentrations of asbestos fiber from chrysotile-containing friction materials was limited. Forsterite has a chemical composition similar to chrysotile, but the material is amorphous and non-fibrous and does not pose the same threat to human health as asbestos (Anderson et al., 1973; Cha et al., 1983; Hickish & Knight, 1970; Jacko et al., 1973; Luxon, 1970; Lynch, 1968; Rowson, 1978; Sheehy et al., 1989; Williams & Muhlbaijer, 1982). Furthermore, as noted by Langer (2003) and others, any remaining chrysotile fibers in the brake wear debris (i.e. those not subjected to mechanical destruction or thermal transformation to forsterite) do not retain their natural properties or biological activity.

Shipyards

Until the late 1970s, asbestos-containing insulation was used extensively within naval ships, and was composed mainly of amosite (up to 86%), and to a much lesser extent, chrysotile (Franke & Paustenbach, 2011; Murbach et al., 2008). Amosite was principally used due to its thermal conductivity, resistance to spreading fires, physical and chemical stability, light weight, strength and refractoriness. Additionally, sections of molded, fragile, amosite-containing insulation were typically covered with a protective layer of chrysotile asbestos (Fleischer & Viles, 1946; Harries, 1971; Rushworth, 2005). Between the 1930s and the 1970s, 30 to 500 tons of asbestos insulation could be used aboard a single warship (Murbach et al., 2008; Rushworth, 2005). Because of increasing concerns related to asbestos health risks, by the 1970s the amount had been reduced to between 3 and 50 tons per ship (Murbach et al., 2008; Rushworth, 2005). Subsequently, most ACM (e.g. gaskets, packing and insulation) were replaced by asbestos-free alternatives in the mid- or late 1970s, and, by 1979, the US Navy ceased using asbestos aboard warships completely (Hollins et al., 2009; Murbach et al., 2008; Rushworth, 2005). During the time that ACM were used in shipbuilding, fiber releases to the ambient air could have occurred due to inadequate exposure control policies, such as the use of dilution ventilation or “open doors” instead of local exhaust ventilation (Marr, 1964). In addition, asbestos-containing insulation materials were also fabricated at the shipyards, which could have resulted in further emissions into the environment (Hollins et al., 2009).

Substantial releases of asbestos fibers can also occur during the overhaul, repair and disposal of ships (Andersen, 2001; Harries, 1971; U.S. EPA, 2000). The disposal of a ship (also known as dismantling) entails removal of equipment, components and consumables for reuse or resale, whereupon the ship is broken up and the material is recycled or disposed of (Andersen, 2001). Inadequate procedures for removing ACM, such as pipe insulation or blankets could result in incidental asbestos releases. Exposure and emissions data related to handling of ACM during ship dismantling operations are not readily available. However, it is likely that although the use of ACM in shipbuilding was discontinued over 30 years ago, the sustained use of historical vessels, as well as maintenance activities will continue to be a source of asbestos emissions into the ambient environment. In addition, given that the service life of a warship can be up to 35 years, decommissioning of ships containing ACM will likely continue far into the twenty-first century (Koenig et al., 2008).

Regulation of asbestos in ambient air

There are no current federal standards that limit the concentration of asbestos in ambient air in the US. Rather, federal regulations set forth restrictions on (1) emission levels from known point sources, (2) the manufacture, importation, processing and distribution of certain asbestos-containing products and “new uses” of asbestos, and (3) the use and handling of ACM during construction, demolition and renovation (U.S. EPA, 1988, 1993a, 1999a).

Starting in the early 1970s and driven by increasing concerns regarding health effects associated with exposure to asbestos, various restrictions and regulations related to ACM were instituted. However, it is interesting to note that demand for asbestos in the US did not peak until the late 1970s. US EPA’s first regulations related to airborne asbestos were proposed in 1971 and promulgated in 1973 under the Clean Air Act; National Emissions Standards for Hazardous Air Pollutants (NESHAP) were intended to protect the public by minimizing release of fibers into the atmosphere during activities that involved processing, handling and disposal of ACM (i.e. materials containing more than 1% asbestos) during building demolition activities (U.S. EPA, 1971, 1973). NESHAP prohibited “visible emissions” from asbestos milling and nine major manufacturing operations through the use of process controls, such as air cleaning equipment and during demolition of structures that contained asbestos (U.S. EPA, 1973).

Through NESHAP, the US EPA also prohibited sprayed-on application of friable ACM for fireproofing and insulation and established routine maintenance procedures (including adequate wetting of any friable fireproofing, insulation or asbestos-insulated pipe prior to removal from a building) for handling ACM on boilers, pipe or load-bearing structural members (U.S. EPA, 1973). Over the following years, the regulations were amended to include additional activities (e.g. fabrication and renovation) and materials (U.S. EPA, 1974c, 1977, 1999b). Notably, in 1978, the US EPA extended its original ban on spray-on asbestos insulation to include banning all uses of spray-on ACM for decorative purposes (U.S. EPA, 1977). In addition, in the late 1970s, the US EPA began to focus on friable ACM in schools by initiating technical assistance programs and publishing guidance documents, as well as enacting and expanding the Asbestos Hazard Emergency Response Act (AHERA) in
1982 and 1986, respectively (U.S. EPA, 1979a,b, 1987). In addition, several states have established their own emission levels or ambient air standards (e.g. State of Connecticut, 2006; State of Vermont, 2011).

Methods
Identification of data sources
A thorough search of the peer-reviewed literature and other publicly available documents was performed to identify asbestos air sampling data for ambient settings in the US using several database search engines (e.g. PubMed, NTIS, Medline, TOXNET, ScienceDirect, ProQuest). To locate additional studies, a systematic review was performed of the reference lists of all studies identified by the initial search, as well as of key review papers. Furthermore, relevant entities were contacted to acquire any supplementary data. If multiple studies presented results pertaining to the same dataset, the study with the original data and/or that contained the final results (as opposed to preliminary analyses) was selected for analysis.

During the literature search, an unpublished dataset was identified, consisting of data collected by the RJ Lee Group, Inc., in urban settings across the US between 1986 and 1998 (Personal Communication, Van Orden, RJ Lee Group, 2013); a summary was previously reported by Lee & Van Orden (2008). These data were reportedly collected as reference samples in relation to demolition and asbestos abatement work at various worksites. For purposes of the analysis presented in this manuscript, only samples collected prior to any work commencing were included. Similarly, all samples collected in locations with known sources of NOA were excluded.

Data for ambient settings were defined as samples collected outdoors in the absence of a known or potential emission source. Data collected near operations likely to be associated with emissions of asbestos fibers, such as a shipyard or asbestos mine, were excluded. In some studies, such as those related to asbestos remediation work, both source-related and ambient data were reported; for such instances, only reference samples collected prior to this type of work were included.

Whenever possible, the following elements were identified and abstracted for each measurement; detailed descriptions of several of these elements are provided below:
- Sample collection date (year)
- Sampling location (name of city or town, state)
- Population category (urban, rural, unknown)
- Sample identifier
- Sampling duration
- Analytical method
- Analytical limit of detection
- Fiber definition or counting protocol (length and/or aspect ratio)
- Concentration (including units of measurement)

Population category
Sampling locations were classified as urban or rural, as designated by the study authors. If such designations were not explicitly assigned, data from the US census conducted on or immediately prior to the sampling year were used to classify the location based on population size. Locations with populations exceeding 50,000 persons were considered urban, and populations less than or equal to 50,000 persons were considered rural (U.S. Census Bureau, 2013). If sampling location could not be determined, the population category was classified as “unknown.”

Analytical methods
Several analytical methods exist for analyzing airborne asbestos samples. In addition, fiber counting protocols (i.e. the definition of what was counted as an asbestos fiber) varied between studies regardless of the analytical method used. Below is a brief description of analytical methods that were identified in the studies that were included in this analysis. Asbestos fibers are typically characterized by their length and aspect ratio (i.e. ratio of fiber length to width). All count-based samples which counted only those fibers equal to or longer than 5 μm, regardless of the analytical method employed or the aspect ratio considered, were combined into one category (i.e. “≥5 μm”). Thus, any count-based measurements for which fiber length was reported as <5 μm, or where fibers of “all lengths” were counted, were excluded from this analysis.

- **Phase contrast microscopy (PCM):** PCM samples are analyzed using a standard optical microscope. In brief, any structure ≥5 μm and with a length to width ratio ≥3:1 is counted as a fiber; however, only fibers ≥0.2 μm are visible, and hence the presence of thinner fibers would not be detected. This method is often favored because it is relatively inexpensive and simple, and sample preparation is straightforward and does not require analysts to use specialized equipment (e.g. a complex electron microscope) (Perry, 2004). This method, however, cannot distinguish between asbestos and non-asbestos structures (e.g. asbestos versus fiberglass), or between asbestos fiber types (e.g. chrysotile versus amphiboles); therefore, in the presence of other fibers, the PCM method may overestimate the actual asbestos fiber concentration. Although in the past, researchers may incorrectly have referred to fiber concentrations based on PCM as “asbestos fiber concentrations”, for the current analysis, fibers were assumed to be asbestos if the original study reported them as such.

- **Transmission electron microscopy (TEM) and electron microscopy (EM):** The TEM technique relies on electron microscopy, rather than optical microscopy; thus, unlike PCM, it can be used to help distinguish between asbestiform and non-asbestiform structures, and also between different types of asbestos fibers based on their crystal structures (U.S. EPA, 1987). TEM also has much greater resolution than PCM, and can better detect fibers <5 μm in length and <0.2 μm in width (Kauffer et al., 1996; Mossman et al., 1990). Disadvantages of TEM include higher equipment costs and increased level of training required for operators (Stewart, 1988). Results from TEM analysis can be used to determine the percentage of asbestos fibers of all fibers in a sample.
For three additional studies included in this analysis, the analytical method was reported simply as “electron microscopy” (EM); for the purpose of this analysis, it was assumed that TEM was utilized for these samples. Additionally, the International Organization for Standardization (ISO) has developed a TEM method (i.e. ISO 10312) for determining concentration of asbestos fibers in ambient air (ISO, 1995). The main difference between the ISO TEM method and the TEM method most often employed in the US (i.e. NIOSH 7402) is that individual fibers are counted even when they are located inside higher-order structures when employing the ISO TEM protocol (ISO, 1995; NIOSH, 1994b). Data collected using the ISO TEM method were kept separate from other TEM and EM data in the analysis. Regardless of the protocol, only fibers reported as ≥5 μm in length were included in the analysis.

- **Scanning electron microscopy (SEM):** Like TEM, the SEM technique relies on electron microscopy rather than optical microscopy. SEM offers similar advantages as TEM, but has less sensitivity for smaller fibers (Burdett & Jaffrey, 1986). For purposes of the current analysis, only fibers with a reported length of ≥5 μm were included in the dataset.

### Analytical limits of detection

In a majority of studies included in this analysis, an analytical method-specific limit of detection (LOD) was provided. In three studies (Chesson et al., 1985; Nicholson et al., 1975; Sawyer, 1977), no LOD was provided, but one or more values of 0 f/cc (or equivalent) were reported. For the purposes of this analysis, it was assumed that these values were below the LOD, and a default value of 0.0010 f/cc was assigned based on the current LOD for PCM as described by OSHA and NIOSH; this is also the typical working minimum LOD for TEM (NIOSH, 1994a,b; OSHA, 1997).

As a result of the range of LODs reported across the studies included in the dataset, concentrations reported as detects in some data subsets were lower than the LODs for other samples. To assess the effect of this overlap, ranges of both detected data and data below the LOD are presented separately.

### Concentrations of asbestos in air and units of measurement

Because the data considered were generated using a variety of analytical techniques, the dataset contained asbestos concentrations that were reported in both count-based [e.g. f/cc, structures per cc (s/cc), etc.] and mass-based (e.g. ng/m³) units. To enable comparisons between these various units of measurement, a literature search was conducted to identify conversion factors to be used to convert mass-based concentrations to count-based (i.e. f/cc) concentrations. In 1986, the US EPA published a report, which included an evaluation of available mass-to-count concentration conversion factors based on samples collected in five studies conducted in occupational settings (principally manufacturing), as well as during one laboratory study (U.S. EPA, 1986). Based on these six studies, the agency suggested that 30 μg/m³ per f/cc be used as a general conversion factor when count-based data were not available. This value was calculated as the geometric mean of the reported conversion factors, which ranged from 5 to 150 μg/m³ per f/cc for fibers ≥5 μm. However, from a review of the studies included in US EPA’s analysis, as well as the scientific literature as a whole, it was found that the conversion factor derived from Davis et al. (1978) of 5 μg/m³ per f/cc may be more appropriate for use in the current analysis. This factor was based on the results of laboratory experiments that were conducted to determine the mass-to-fiber relationship using pure chrysotile asbestos (Davis et al., 1978). Thus, unlike conversion factors derived based on PCM measurements collected in occupational settings [such as those cited by the U.S. EPA (1986)], the mass-fiber ratios reported by Davis et al. (1978) would not have been influenced by the presence of non-asbestos fibers. This outcome was desirable since all mass-based concentrations included in this study were the result of TEM analysis, and were thus a measure of asbestos only (and not other agents or dusts). Moreover, given that chrysotile asbestos was the most widely used fiber type in the US, it would be expected that it would be the primary asbestiform fiber type found in the ambient air.

### Data analysis

Statistical analyses were performed using R software (V3.2.0, The R Foundation for Statistical Computing, Vienna, Austria) and the NADA package for R (Lee, 2013). Data were not normally or log-normally distributed, and as noted above a large fraction were below various LODs; thus, non-parametric methods were used for all statistical analyses. For the same reason, the median concentrations were estimated and were presented alongside the mean concentrations.

The reverse Kaplan-Meier (KM) estimator was used to estimate the mean and median asbestos concentrations, because this method tends to be insensitive to outliers and a good choice for analysis of relatively small datasets (Antweiler & Taylor, 2008; Gillespie et al., 2010). Mean asbestos concentrations were estimated by calculating the area between the reverse KM estimator’s cumulative density function (CDF) and 1 for data >0; which is equivalent to summing the products of each detected value and its corresponding probability (Gillespie et al., 2010). Similarly, the median asbestos concentration was determined to be the smallest detected value in any given data subset for which the respective CDF was ≥0.5. When the CDF exceeded 0.5 at the minimum detected concentration and the smallest value was < LOD, the software did not estimate a median. For these subsets, the minimum detected concentration was reported as the median, and this was denoted in the table.

A high percentage of data was below the LOD in some subsets of the data used in the study. Traditional statistical methods for testing differences between subsets of data would not be informative or even appropriate. Therefore, comparisons by environment type (urban versus rural) or decade were instead evaluated by comparing differences in the proportion of samples for which asbestos concentrations exceeded a pre-selected cut-off value. The cut-off value was representative of an upper bound for the LODs reported across all the data sets. Because of the wide range of LODs for data below the detection limits across the studies evaluated (a result of the
variety of sampling and analytical methods), it was not feasible to select a cut-off point above the highest reported LOD. Therefore, the cut-off point was chosen as the 95th percentile of all reported LOD values and was estimated to be 0.0046 f/cc. The concept of using the 95th percentile as the cut-off point is commonly used in statistical analyses. The proportions of measurements above the cut-off for different subsets were compared through statistical analysis: a two-sample test of proportions was used to compare two subsets, and the Holm-Bonferroni corrected pair-wise two-sample test of proportions was used to compare more than two subsets simultaneously (Holm, 1979).

Results
A total of 16 studies, either published in the scientific literature or as reports, were identified that described the results of 381 samples collected from the 1960s through the 1980s, and the 2000s in at least 30 states across the US (i.e. some studies only reported ‘‘US’’) (ATSDR, 2007; Baxter et al., 1983; Bruckman, 1978; Chesson et al., 1985; Heffelfinger et al., 1972; LeMoine, 1981; Mangold, 1982, 1983; Nicholson, 1971; Sawyer, 1977; U.S. EPA, 1974d, 1975, 2007, 2009; Wendlick, 1983, 1984). As noted above, a previously unpublished dataset was also identified, consisting of data (n = 1677) collected by the RJ Lee Group, Inc., between 1986 and 1998 in urban settings across at least 34 states in the US (Personal Communication, Van Orden, 2013); a summary was previously reported by Lee & Van Orden (2008). Thus, the total dataset contained 2058 data points collected in at least 40 states across the US between the 1960s and 2000s. A description of the studies included in this analysis is presented in Table 1.

The descriptive statistics for the overall dataset are presented in Table 2. The overall mean and median ambient asbestos concentrations based on all data were 0.00093 f/cc and 0.00022 f/cc, respectively. Histograms depicting the overall data distribution, as well as for subsets of data based on environment type and decade are included in Supplementary Appendix A.

Environment type
The mean ambient asbestos concentrations in urban (n = 1954) and rural (n = 102) settings were 0.0011 f/cc and 0.00039 f/cc, respectively (Table 2). The median ambient asbestos concentration was 0.00050 f/cc in urban settings and 0.000020 f/cc in rural environments. The percentages of urban and rural data that exceeded the 95th percentile of the reported LOD values (i.e. 0.0046 f/cc) were 8.1 and 3.9%, respectively. There was no statistically significant difference between these two proportions (p = 0.13).

Data for both urban and rural settings were only available for the 1970s. While the mean rural concentration (0.0018 f/cc) exceeded the mean urban concentration (0.0010 f/cc) in the 1970s, the median urban concentration (0.00060 f/cc) was considerably higher than the median rural concentration (0.000021 f/cc). Surprisingly, while 19% of samples collected in rural settings in the 1970s exceeded the 95th percentile of the reported LOD values, only 4.6% of urban samples collected during the 1970s exceeded this cut-off; these two proportions were found to be statistically significantly different (p = 0.017).

Temporal trends
As seen in Table 2, ambient air samples collected in the 1960s (n = 64), 1970s (n = 132), 1980s (n = 659), 1990s (n = 1122) and 2000s (n = 81) were identified. The mean ambient asbestos concentrations for these decades were 0.0012, 0.0011, 0.0022, 0.0016 and 0.000017 f/cc, respectively. The median asbestos concentrations were 0.00028, 0.00044, 0.00090, 0.0016 and 0.000014 f/cc. As shown in Table 2, the percentages of data that exceeded the 95th percentile of the reported LOD values varied, and increased from 4.7% in the 1960s to 6.8% in the 1970s and 20% in the 1980s. Then it decreased to 1.7% in the 1990s and 0% in the 2000s. The proportion of measurements exceeding the cut-off for the 1980s was statistically significantly different than for all other decades (p values ranging from <0.0001 to 0.029), and the proportion from the 1970s was statistically significantly different than the proportion from the 1990s (p = 0.0043).

Converted values
Approximately 9.0% (n = 186) of the samples included in this dataset were reported as mass-based concentrations (Table 3). These data were based on five studies (Bruckman, 1978; Heffelfinger et al., 1972; Nicholson, 1971; U.S. EPA, 1974d, 1975) and included samples collected in the 1960s and 1970s. The overall mean concentration for the converted data was 0.00096 f/cc, which is within a factor of 1.2 of the overall mean concentration for the non-converted data (0.00080 f/cc). However, the median concentration for the converted data (0.00032 f/cc) exceeded that for the non-converted data (0.000014 f/cc) by over 20-fold.

Methods of analysis
Most ambient air samples were analyzed using TEM or EM [n = 1943; range: <0.000031–0.019 f/cc (note: the highest LOD was 0.025 f/cc)], with the others analyzed by PCM (n = 105; range: <0.0010–0.050 f/cc), SEM (n = 9; range: <0.0000024–0.016 f/cc), and ISO (n = 1; <0.000010 f/cc). The mean and median PCM concentrations were higher than those reported for samples analyzed by TEM and SEM, which may be a result of date of sampling (1970s and early 1980s for all PCM, while a majority of TEM samples were collected in the 1990s), or the fact that PCM does not distinguish between asbestos and non-asbestos fibers.

Discussion
The task of identifying data relevant to describe ambient concentrations of asbestos in the US over time, as described above, was rather straightforward. Anderson et al. (2015) recently published a review of “ambient air asbestos concentrations”. There was some overlap between the data evaluated by Anderson et al. (2015) and those included in the current analysis; however, their dataset was smaller, the inclusion criteria appear to have been less rigorous (e.g. some data which could be attributed to point sources were

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| Reference | Objective | Ambient data selected | Analysis method | Sample size | Sampling location | Environmental type |
|-----------|-----------|-----------------------|-----------------|-------------|-------------------|-------------------|
| ATSDR (2007) | Evaluation of potential asbestos contamination of public beach | Reference samples | TEM | 7 | Zion, IL | Rural |
| Baxter et al. (1983) | Evaluation of ambient concentrations throughout California | 1983 | SEM | 9 | Various locations: CA | Urban, rural |
| Bruckman & Rubino (1978) | Study of airborne asbestos fiber concentrations in Connecticut | 1977 | TEM | 2 | CT | Urban |
| Chesson et al. (1985) | Project related to ACM removal in schools | Reference samples collected five months after completion project | 1985 | PCM | 6 | Various locations: WA, OR, TX | Urban |
| Heffelfinger et al. (1960s, 1970s) | Study to develop new sampling methods for ambient asbestos | Reference samples | 1972 | TEM | 33 | Various locations: CA, DC, KY, OH, PA, TX | Urban, rural |
| LeMoine (1981) | A survey of the asbestos levels in the ambient air of Seattle | 1980 | PCM | 36 | Seattle, WA | Urban |
| Mangold (1982) | Evaluation of contribution to ambient concentrations from certain products | Reference samples | 1982 | PCM | 18 | Various locations: WA, OR | Urban |
| Mangold (1983) | Evaluation of ambient concentrations in the Greater San Francisco area | 1983 | PCM | 25 | Various locations: CA | Urban |
| Nicholson (1960s, 1970s) | Evaluation of ambient concentrations in the U.S. | 1971 | NR | 117 | Various locations: CO, GA, IL, MA, MD, MI, OH, TX | Urban |
| Personal Communication, Van Orden, R.J. Lee Group (2013) | Unpublished data collected as reference samples | 1986–1998 | TEM | 1677 | Various locations and states | Urban |
| Sawyer (1977) | Evaluation of asbestos concentrations in buildings | Reference samples | 1977 | PCM | 1 | Various locations and states | Urban |
| U.S. EPA (1974d) | Evaluation of ambient concentrations in buildings | 1974 | EM | 33 | New Haven, CT | Urban |
| U.S. EPA (1975) | Evaluation of naturally occurring asbestos exposures to naturally occurring asbestos | 1975 | EM | 1 | Berkeley, CA | Urban |
| Wendlick (1983) | Evaluation of contribution to ambient concentrations from certain products | Reference samples | 1983 | TEM | 10 | Portsmouth, VA, Newport News, VA | Urban |
| Wendlick (1984) | Study of airborne asbestos fiber concentrations in the state of Pennsylvania | Reference samples | 1984 | PCM | 9 | Philadelphia, PA | Urban |

*Unless otherwise noted, any data reported which met the inclusion criteria of the current study were selected for analysis.
*NR = Not Reported.
*A summary of these data was previously reported by Lee & Van Orden (2008).*
was higher than in urban areas, due to the small sample size. The mean concentration for rural samples collected in the 1970s in urban environments was nearly 29-fold higher than the median concentration for samples collected in both urban and rural environments, whereby the median concentration for samples collected in rural environments was generally higher than in urban environments. Specifically, the median concentration in urban environments were generally higher than in rural environments. This relationship held true for the 1970s, the only decade for which samples were collected in both urban and rural environments, whereby the median concentration for samples collected in urban environments was nearly 29-fold higher than the median concentration for rural samples. While the mean concentration for rural samples collected in the 1970s was higher than in urban areas, due to the small sample size for rural environments, the mean was greatly influenced by a limited number of samples collected in Minnesota and California. It is possible that these concentrations were impacted by nearby (but not identified) sources of asbestos, such as NOA or mining operations. Because these samples skewed the distribution of this subset of data, the median concentration is likely a better indicator of the central tendency. The observation of higher ambient air asbestos concentrations in urban versus rural environments may be explained by factors, such as the greater usage of friable ACM in more densely populated areas, especially in the construction industry where spray insulation was common.

Overall, when considering both the estimated concentrations and the percentage of data above the cut-off value of 0.0046 f/cc, ambient asbestos concentrations generally increased from the 1960s through the 1980s, after which they declined considerably. The continued increase in ambient air concentrations of asbestos throughout the 1980s was somewhat unexpected given that use of ACM in construction decreased dramatically throughout the 1970s. However, this observation may be the result of new federal regulations promulgated by the US EPA that led to increased abatement and demolition activity, which possibly resulted in uncontrolled emissions of asbestos. This conclusion is further supported by a granular analysis of the 1980s data, which revealed that a majority of the higher concentrations were from samples collected in the early 1980s. The higher concentrations in the early 1980s may also be an artifact of the inclusion of PCM data, which, as previously noted, may overestimate the true asbestos fiber concentration compared...

| Data subset | Fraction < LOD (%) | Fraction of converted samples (%) | Range of values < Limit of detection (LOD) (f/cc) | Range of detected values (f/cc) | Median (f/cc) | Mean (f/cc) | Fraction > Cut-off (%) |
|-------------|---------------------|----------------------------------|-----------------------------------------------|--------------------------------|---------------|---------------|------------------------|
| Overall     | 2058 84.9 9.0       | <0.0000024 0.00025               | 0.000048 0.050                                 | 0.0022 0.00093                 | 7.9           |
| Environment | Urban 1954 85.3 8.8 | <0.00071 0.0025                 | 0.00004 0.050                                 | 0.0050 0.0011                  | 8.1           |
| Rural       | 102 76.5 14.7       | <0.0000024 0.00027              | 0.000048 0.013                                 | 0.00020 0.00039                | 3.9           |
| Unknown     | 2 100 0            | <0.0000024d                    | –                                              | –                              | –             |
| Decade      | 1960s 64 0 100      | –                               | 0.00004 0.019                                 | 0.0028 0.0012                  | 4.7           |
|             | 1970s 132 6.1 92.4 | <0.0000024 0.00020              | 0.000048 0.016                                 | 0.0044 0.0011                  | 6.8           |
|             | 1980s 659 83.6 0   | <0.000071 0.0025                | 0.00010 0.050                                 | 0.0090 0.0022                  | 20            |
|             | 1990s 1122 99.2 0  | <0.0017 0.0069                  | 0.016 0.0037                                  | 0.016 0.0016                   | 1.7           |
|             | 2000s 81 92.6 0    | <0.000031 0.00027              | 0.00014 0.000092                              | 0.00014 0.00017                | 0             |
| Urban       | 1960s 64 0 100      | –                               | 0.00004 0.019                                 | 0.0028 0.0012                  | 4.7           |
|             | 1970s 109 2.8 98.2 | <0.0010 0.0020                 | 0.00004 0.016                                 | 0.0060 0.0010                  | 4.6           |
|             | 1980s 659 83.6 0   | <0.000071 0.0025                | 0.00010 0.050                                 | 0.0090 0.0022                  | 20            |
|             | 1990s 1122 99.2 0  | <0.0017 0.0069                  | 0.016 0.0037                                  | 0.016 0.0016                   | 1.7           |
|             | 2000s 81 92.6 0    | –                               | –                                              | –                              | –             |
| Rural       | 1960s 0 – –         | –                               | –                                              | –                              | –             |
|             | 1970s 21 14.3 71.4 | <0.0000024 0.00020              | 0.000048 0.013                                 | 0.00021 0.0018                 | 19            |
|             | 1980s 0 – – –       | –                               | –                                              | –                              | –             |
|             | 1990s 0 – – –       | –                               | –                                              | –                              | –             |
|             | 2000s 81 92.6 0    | <0.000031 0.00027              | 0.00014 0.000092                              | 0.00014 0.00017                | 0             |

*aConversion factor derived from Davis et al. (1978) (i.e. 5 μg/m³ per f/cc).
*bCalculated using the reverse Kaplan-Meier estimator.
*cReported as percentage of samples with a concentration above 0.0046 f/cc (i.e. the 95th percentile of all LOD values in the dataset).
*dAll samples below the LOD.
–: Data not available/not applicable.
### Table 3. Descriptive statistics of ambient asbestos concentrations by conversion factor and analytical method.

| Data subset | Converted data | Data type | LOD (%) | n | Mean (f/cc) | Median (f/cc) |
|-------------|----------------|-----------|---------|---|-------------|--------------|
| All         | Overall        | Overall   | 1872    | 93.2 | 0.00080     | 0.000069     |
|             | Rural          | Overall   | 186    | 98.5 | 0.000048    | 0.000042     |
|             | Urban          | Overall   | 1943   | 98.6 | 0.000044    | 0.000020     |
|             | Rural          | Urban     | 171    | 92.7 | 0.000021    | 0.000060     |
|             | Urban          | Rural     | 104    | 94.3 | 0.000010    | 0.000016     |

**Fraction of converted samples (%)**

- **No conversion**
  - Overall: 1872, 93.2%
  - Rural: 186, 98.5%
  - Urban: 1943, 98.6%
- **Converted data**
  - Overall: 186, 92.7%
  - Rural: 171, 92.7%
  - Urban: 104, 94.3%

**Range of values (f/cc)**

- **No conversion**
  - LOD: 0
  - Range: <0.000024, <0.000027
  - <0.0000024, <0.000125
- **Converted data**
  - LOD: 0
  - Range: <0.000024, <0.000027
  - <0.000010, <0.00020

**Median (f/cc)**

- **No conversion**
  - LOD: 0
  - Median: 0.000014, 0.000014
- **Converted data**
  - LOD: 0
  - Median: 0.000010, 0.000010

**Analytical method**

- **TEM/EM**
  - Overall: 1943, 98.6%
  - Rural: 171, 92.7%
  - Urban: 104, 94.3%

**Conversion factor derived from Davis et al. (1978)**

- For mass-based data, the conversion factor is 0.00093 f/cc, which is 1.3 times lower than the estimated median for the converted mass-based data, as described in the "Methods" section. These results are shown in Tables B4 and B5 in Supplementary Appendix B. The overall mean ambient concentration using this conversion factor is 0.000069 f/cc, which is 1.3 times lower than the overall mean of 0.000093 f/cc from Table 2. Similarly, the overall median using this conversion factor is 0.000043 f/cc, which is 5.1 times lower than 0.000022 f/cc from Table 2. For converted data only, the overall mean concentrations using the Davis et al. (1978) and US EPA conversion factors were 0.000096 f/cc and 0.000016 f/cc, respectively; the corresponding median concentrations were 0.000032 f/cc and 0.0000053 f/cc, respectively.

The six-fold difference in concentrations determined using the two conversion factors corresponds exclusively to the relative magnitude of the two factors. Moreover, as a result of this six-fold difference, concentrations for subsets with high percentages of converted data (i.e. 1960s = 100%, 1970s = 92.4%) were found to be much lower when using the US EPA factor versus the Davis et al. (1978) factor, and when compared to other subsets of data with no converted measurements, such as the 1980s, these estimates appear to be lower than expected.

One limitation of the dataset was the combination of data collected using various analytical methods (i.e. PCM, TEM, SEM, ISO). Thus, data meeting either the simple criteria of 5 µm in length and a length-to-width ratio of >3:1 were combined, regardless of fiber type and diameter. Ideally, all data should have been converted to a single metric (e.g. PCME) prior to conducting any statistical analysis, but sufficient information for such conversions was not available for the vast majority of data points. Furthermore, as was noted by the ATSDR (2001, pp. 157–158) and the U.S. EPA (2001), the relationship between TEM and PCM fiber counts is too variable to allow for development of a conversion factor to be used across settings and fiber types. Nonetheless, the aggregation of data generated using different analytical methods may have had an impact on the results. For example, as noted in the "Methods" section, PCM does not distinguish between asbestos and other fibers, and the inclusion of PCM data may have inflated the concentrations for certain subsets of the data. Similarly, TEM analysis allows for detecting fibers with a diameter <0.2 µm, which could have inflated the concentrations for the very small fibers that were not detected by other methods. To evaluate the potential impact of the choice of conversion factor on the results, the dataset was analyzed using the conversion factor suggested by the US EPA (30 µg/m³ per f/cc; as described in the "Methods" section). These results are shown in Tables B4 and B5 in Supplementary Appendix B. The overall mean ambient concentration using this conversion factor is 0.000069 f/cc, which is 1.3 times lower than the overall mean of 0.000093 f/cc from Table 2. Similarly, the overall median using this conversion factor is 0.000043 f/cc, which is 5.1 times lower than 0.000022 f/cc from Table 2. For converted data only, the overall mean concentrations using the Davis et al. (1978) and US EPA conversion factors were 0.000096 f/cc and 0.000016 f/cc, respectively; the corresponding median concentrations were 0.000032 f/cc and 0.0000053 f/cc, respectively.

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resulting concentrations compared to if PCME data had been available. However, because certain subsets of the data heavily relied upon one analytical method (e.g. all early 1980s data were based on PCM, whereas all 1960s and almost all 1970s data were based on TEM or EM), it was deemed appropriate to include all metrics, to enable further statistical analyses (e.g. evaluations of concentration differences over time).

Considerable efforts were taken to identify all published and unpublished air sampling data for ambient asbestos; however, additional studies and reports containing relevant data may still exist. No attempt was made to differentially weight the studies in this analysis; however, there is likely some degree of variability in the quality of data collection and interpretation methods, particularly with respect to air sampling techniques and fiber counting protocols. One likely reason for the scarcity of data in some subsets is that, in general, sampling strategies for outdoor concentrations of asbestos may have entailed characterizing the highest potential “near source” concentrations (i.e. data not considered in this analysis). This strategy is analogous to that commonly employed in occupational settings, in which sampling generally is conducted in areas with high potential for exposure (Damiano & Mulhausen, 1998). Indeed, in several of the datasets used in the current analysis, only background or reference samples were identified as ambient data. Some of the studies included in this analysis were conducted in order to investigate the potential impact on ambient concentrations from certain point sources. For example, Mangold (1982, 1983) noted the likely contribution to ambient fiber concentrations in Bremerton, WA from a nearby shipyard. This is an important finding, although for purposes of the current analysis, per the inclusion criteria, samples specific to such settings (i.e., “near a known source”) were excluded from the dataset.

As shown in Tables 2 and 3, a limitation of this analysis was the high percentage of data below the LOD for certain subsets, especially for some of the later decades (e.g. 99.2% of samples collected in the 1990s and 92.6% of samples collected in the 2000s were < LOD). The statistical analysis attempted to account for these issues by comparing the proportions of samples below an expected upper-bound LOD for different data subsets, but lack of actual detected concentrations most likely has impacted the results nonetheless. It is also possible that any identified temporal trends are due to the decrease in the LODs over time instead of actual concentration decreases. For example, it is unknown whether a concentration reported as < LOD in the 1970s is higher or lower than a concentration reported as < LOD in the 2000s, yet due to the lower LOD in the 2000s, the resulting mean concentration may have been estimated to be lower than that in the 1970s solely due to the lower LOD. However, this is unlikely to account for the entire difference, given the drastic decline in asbestos use in the US between the 1970s and the present.

Moreover, for purposes of comparing the results from this analysis to those generated using other standard methods, an additional analysis was performed in which all measurements below the LOD were assumed to be equal to the LOD/2. This method is commonly used to deal with values below the LOD, but has limitations (e.g. there is a potential for bias, for data sets with multiple LODs, when some LODs are in the upper range of the actual data distribution) compared to the more appropriate methods employed in the current analysis (Gillespie et al., 2010). However, with few exceptions, the results of this additional analysis were consistent with those generated using the reverse KM method. There were a limited number of instances in which the reverse KM method generated a considerably lower estimate than the LOD/2 substitution method. For example, this was the case for the mean for the overall dataset and the 2000s data, and the median for the urban environment data. Nonetheless, this result was to be expected, as the reverse KM estimator considers the distribution of the data when calculating means (graphic representations of distributions are shown in Supplementary Appendix A), whereas the LOD/2 substitution method does not. In other words, for the overall dataset, the detected values tended to fall below the LOD/2 values; therefore, the mean concentration estimated using the reverse KM estimator was lower than the mean estimated using the LOD/2 method.

Lastly, the unpublished RJ Lee Group dataset represented a majority of the data included in this analysis, especially for urban environments in the 1990s. Given that these data were collected in a variety of sampling locations across the US (including many different cities and states), they were considered to be an important addition to the literature and this analysis. Nonetheless, in an effort to evaluate the relative impact of the RJ Lee Group data on the overall results, the statistical analysis was also performed without this dataset; the results are shown in Tables B6 and B7 in Supplementary Appendix B. Notably, the overall trends and conclusions did not change when evaluating the results of this alternative analysis.

Conclusions

In conclusion, the objective of this research was to provide a review and analysis of concentrations of asbestos in the ambient air in the US over time, an analysis that has not been published in the scientific literature. Based on the results of this study, it was found that ambient air concentrations of asbestos were higher in urban versus rural environments. In addition, ambient asbestos concentrations likely peaked sometime in the 1980s, and appear to have declined since then. These results are consistent with the patterns of use of asbestos in the US, and suggest that federal regulations introduced in the 1970s (such as asbestos abatement) aimed at decreasing asbestos exposure in the general public may have resulted in an unintended and transient increase in the ambient air concentration of asbestos. These data may also be useful in retrospectively assessing human exposures to asbestos present in ambient air.

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Declaration of interest

Cardno ChemRisk is a consulting firm which has been engaged by companies that have been, and are, involved in asbestos-related litigation, to provide general consulting and expert advice on scientific matters, as well as litigation support. This work was funded in its entirety by Cardno ChemRisk, and the preparation of the paper is the exclusive professional work of the authors. Drs Pierce and Paustenbach have served as experts in asbestos-related litigation, and they, along with others, may be called upon in the future to serve as expert witnesses in related litigation.

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