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Identification of Asbestos and Glass Fibers in Municipal Sewage Sludges

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It is estimated that about seven million tons of sewage sludge will be produced annually in the United States by 1985 (Walsh 1976). Sewage sludge is produced by removal of solids and pathogens in wastewater from industrial and domestic users in sewage treatment plants. Asbestos fibers in such water would expectedly become entrained in the remaining sludge solids. Asbestos fibers have been found in drinking water in United States cities. A review of the studies dealing with surveys of asbestos fibers in water supplies in the United States has been published (Millette et al. 1983).

It has been proposed that sewage sludge be utilized as a soil amendment both in the greenhouse and field since it contains essential plant nutrient elements and is high in organic matter. It may also contain a wide spectrum of toxic heavy metals and organics largely of industrial origin (Mumma et al. 1983, 1984; Babish et al. 1981). Studies of the possible presence of asbestos fibers in sewage sludge have not been reported. Many investigations have focused on possible root absorption of toxicants by plants and resultant harmful effects in foraging animals from its use in agriculture. Contamination of crops by soil debris through rain-splashing and inadvertant ingestion of soil by ruminants that tear out and consume plant roots during grazing are other mechanisms by which plants and animals can become contaminated (Fries 1982). Inhalation of dust during the preparation and bulk-mixing of dry growth media containing sludge in greenhouses or occupational exposure during preparation (grinding, mixing) of dry, bagged, commercial sludge products is a little-studied problem. While root absorption of asbestos by plants is improbable, these latter mechanisms (splashing, soil ingestion, dust inhalation) could cause exposure of animals and humans to asbestos.

About 800,000 tons of asbestos fiber were used annually in the United States between 1971 and 1975 (Levine 1978). It is used as a component in a wide variety of products including floor tile,

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shingles, gaskets and packings, friction products, coatings, reenforced plastics, cement pipe and sheet, textiles and paper materials.

This report provides the results of the analysis of municipal sewage sludges from five large American cities for the presence of asbestos fibers. The possible toxicologic significance of the findings for specific occupational groups is discussed.

MATERIALS AND METHODS

The sludge samples were taken by personnel at each of the municipal sewage treatment plants studied. Information pertaining to their origin and the respective wastewater treatment process is given in Table 1. The composition of sludge in a given treatment plant is constantly changing with time as determined by shifts in industrial activities. However, since wastewater solids typically undergo continuous mixing during anaerobic and aerobic digestion, a single sludge sample probably represents the average composition of wastes entering a plant over a period of several days. The samples were freeze-dried and examined by x-ray diffraction, high power light optical microscopy and electron microscopy.

The sludge samples were mounted as randomly oriented powders and placed in front-loading aluminum sample holders (Jackson 1969). Diffractograms were obtained using a Philips x-ray diffractometer, employing Cu-Kα radiation and equipped with a 2θ compensating slit. Scans were taken from 3 to 60 degrees of 2θ at instrument settings of 40 kilovolts and 20 milliamperes. Data obtained was referred to the standard reference values of Chao (1969) for asbestos minerals.

A small quantity of each of the sludge samples was placed on a glass slide and examined using an optical microscope with oil immersion. The resolution limit of the microscope was about 1 micron.

In order to further identify and quantify possible fibers in sludge, electron microscopic analysis was used. About 200 mg of the sludge sample was ashed in a beaker using a Dionex low temperature asher with a power setting of 250 watts. The ash was suspended in 300 ml of filtered water and agitated in an ultrasound bath for 5 min. The suspension was stirred automatically and after 8 seconds a sample was withdrawn using an automatic pipette at a depth of 2.5 cm. Three such pipetted samples ranging in volume from 0.75 to 10.0 ml were collected. The samples were diluted with filtered water and passed through a 47 mm diameter Nuclepore filter (0.1 μm pore size) using vacuum. The Nuclepore filter was coated with carbon and a thin layer of gold in a vacuum evaporator to enable electron diffraction standardization. Small pieces of the filter were cut out and placed over TEM grids above a modified Jaffe-Wick chloroform bath. The grids were then removed and analyzed using a Hitachi Model H 500-H transmission electron microscope.

Table 1. Data pertaining to municipal sludges analyzed.

City	Sewage plant	Wastewater volume % Industrial	Wastewater volume % Domestic	Wastewater treatment scheme	Sludge handling scheme	Chemicals added	Ultimate sludge disposal method
Baltimore,MD	Back River	15	85	Activated sludge	Thickening, aerobic digestion, vacuum filtration	Polymer	Landfill, reclamation & compost
Cincinnati,OH	Mill Creek	60	40	Activated sludge (UNOX process)	Thickening, heat drying, incineration	Calgon Ellll	Landfill
Dallas,TX	Dallas Central	12	88	Activated sludge	Anaerobic digestion, lagooning	Polymer	Land application
Los Angeles,CA	Hyperion	15	85	Activated sludge, chlorination	Anaerobic digestion	None	Ocean disposal
Philadelphia,PA	Northeast	16	84	Activated sludge	Anaerobic digestion, centrifugation, conposting	None	Land application & reclamation

The procedure used was the Environmental Protection Agency's presently recommended method for analysis of asbestos in water (Anderson and Long 1980) except that the identification procedures were enhanced and fewer fibers were counted. Random grid squares were selected and all fibers within the grid square were counted, measured and analyzed. A "fiber" in this procedure is a particle with parallel sides and a length to width ratio of 3:1 or greater. An energy dispersive spectroscopy spectrum (EDS) was obtained from each non-chrysotile fiber to provide information about its chemical composition. Where possible, the fibers were tilted in the microscope to obtain zone axis electron diffraction patterns. These provide information about the crystal structure of the fiber and help to classify the fibers into mineral categories.

The concentration for each fiber classification was computed from the number of fibers counted in a particular area on the grid, the volume of sample filtered and the mass of ashed sludge. Amphibole fibers were those which were positively identified as amphibole by chemical composition, electron diffraction pattern, or both. Chrysotile fibers were identified by electron diffraction pattern only. "Non-amphibole, non-chrysotile" fibers were those which were positively not in the first two groups. Ambiguous fibers were those where the information (diffraction patterns and EDS spectra) was inadequate to classify the fibers in the first three groups.

RESULTS AND DISCUSSION

No evidence for the presence of asbestos fibers was found in any of the sludge samples by x-ray diffraction analysis using the Philips x-ray diffractometer. The relatively higher limit of detection of this method probably prevented their detection. Optical (oil immersion) microscopy indicated the presence of asbestos (inorganic and displaying biaxial optical character, i.e. two optic axes and three indices of refraction) fibers as well as glass (optically isotropic) fibers in all of the sludge samples. The dimensions of both fiber types ranged from up to about 5 microns in diameter to 50 microns in length.

Only the sludge sample from Los Angeles was subjected to electron microscopic analysis. The results of this analysis performed on two replicate samples of the sludge are shown in Table 2. The lower and upper limit columns in Table 2 refer to the 95% confidence interval. This interval is based solely on counting statistics. It assumes the fibers are randomly distributed across the filter area (Poisson distribution). Under this assumption, the width of the interval is inversely proportional to the number of fibers counted. This interval does not include errors due to sampling, preparation or identification of fibers on a sample. However, it has been our experience based on analysis of replicate samples, that random counting error is the overriding source of variability on these analyses. This confidence interval applies to a perfect count of randomly distributed fibers. The variability is greater where significant amounts of clumping exist.

Table 2. Fiber type and concentration found in two replicate electron microscopic analyses of Los Angeles sludge.

Fiber type	Rep. no.	Concentr. fibers/gm sludge (dry wt)	95% confidence limit concentr.(fibers/gm) Lower	Upper	Effective blank level
Amphibole	1	3.61×10^7	4.37×10^6	1.30×10^8	3.23×10^6
Amphibole	2	0	0	7.36×10^7	1.95×10^6
Chrysotile	1	5.77×10^8	3.95×10^8	8.15×10^8	3.56×10^7
Chrysotile	2	5.78×10^8	3.87×10^8	8.30×10^8	2.15×10^7
Non-amphibole Non-chrysotile	1	1.26×10^8	5.07×10^7	2.60×10^8	
Non-amphibole Non-chrysotile	2	2.19×10^8	1.09×10^8	3.92×10^8	
Ambiguous	1	3.61×10^7	4.37×10^6	1.30×10^8	
Ambiguous	2	3.99×10^7	4.82×10^6	1.44×10^8	
Total fibers	1	7.76×10^8	5.62×10^8	1.05×10^9	
Total fibers	2	8.37×10^8	6.04×10^8	1.13×10^9	

The effective blank level column in Table 2 provides a way of comparing the contamination level on blank filters to the sample concentration. The Nuclepore filters which are used to filter the samples and glassware used in ashing often have small amounts of asbestos on their surfaces. The effective blank level gives the concentration which these fibers would represent if the mass of sludge ashed for the sample were ashed and filtered through a blank filter. In other words, if the sample mass of asbestos-free sludge were ashed and filtered through a Nuclepore filter, we would expect, on the average, to find concentrations of fibers equal to the effective blank level. These numbers are averages ("mean" for amphibole, "median" for chrysotile) derived from the analysis of several ashed empty beakers. The lower confidence limit for the chrysotile concentrations in fibers/gram of sludge is above the effective blank level. This is a good indication that the chrysotile fiber concentration is not due to laboratory contamination. The limit of detection of the method was the concentration represented by one fiber counted over the entire area examined in the analysis.

The preparation method used for these samples will not analyze the largest particles in the ashed sludge. They will settle out of the suspension before the sample is obtained with the pipette. Those particles would be extremely difficult to include in a TEM fiber count because their size would obscure other particles and their thickness would prevent penetration of the electron beam.

The two amphibole fibers identified in the first replicate of the sample were both tremolite (Ca, Mg amphibole). Tremolite fibers have never been found in blank preparations. Therefore, these fibers are probably a part of the sample. The fact that they were not observed in the second replicate may be a result of counting

statistics and the size of the area on the grid that was examined. The close agreement between the sample and its replicate shows that the analysis for asbestos fibers is repeatable. This is important since inter-laboratory comparisons reported in the 1970's (Brown et al. 1976; Bishop 1976) often showed results that differed by more than an order of magnitude. Current analytical efforts by competent laboratories following standardized techniques can be expected to yield results that are reproducible to ±25% (Chopra 1978).

To our knowledge, no reports of a sewage sludge sample analyzed for asbestos have been published. It is difficult to assess the health significance of the presence of asbestos or glass fibers in these single sludge samples. The presently allowed time-weighted-average (TWA) concentration of asbestos in air among those occupationally exposed is 100,000 fibers per cubic meter (Lewis and Tatken 1979). What the exposure level and health effects would be to workers employed in preparation of dry, bagged sludge products for sale or those applying such products in floricultural or horticultural work is unknown. This would depend on many factors including the concentration and types of asbestos fibers present in the air, the simultaneous presence of other toxicants and pathogens which may exacerbate asbestos toxicity, volume of air inhaled, times of exposure, the use of protective respiratory devices and the susceptibility of the individual.

Considering the actual toxic effects of specific fiber types, glass fibers have been reported to inhibit cell growth of bronchial epithelial cells in culture but far less than asbestos fibers (Haugen et al. 1982). The mechanisms through which asbestos is involved in the production of various forms of cancer or other lesions when inhaled or ingested are controversial (Mossman et al. 1983). The possible significance of the nickel and chromium contamination of asbestos fibers regarding cancer initiation has been discussed (Roy-Chowdhury et al. 1973). The length and particularly the diameter of asbestos fibers reportedly is a crucial factor in the production of mesotheliomas (Timbrell et al. 1969; Timbrell 1965; Cook et al. 1982). The hemolytic activity of asbestos has been correlated with its surface charge as indicated by its zeta potential (Light and Wei 1977). Carcinogenic polycyclic aromatic hydrocarbons adsorbed to asbestos have been studied and implicated as synergistic with asbestos in tumor production (Thomson et al. 1978; Mossman and Craighead 1981). The possible synergistic effects of heavy metals or toxic organics and asbestos is especially pertinent in the case of sludge exposure since a galaxy of such toxicants typically would exist in sludge with asbestos (Mumma et al. 1983, 1984; Babish et al. 1981).

The type of asbestos fiber to which workers are exposed has been reported to markedly affect the incidence of cancer. Respiratory cancer was reported to be 2.4 times the normal incidence among workers exposed to chrysotile asbestos but 5.3 times greater among those exposed to both chrysotile and crocidolite (Enterline et al. 1973). Furthermore, specific organ cell types respond differently to asbestos (Mossman et al. 1983).

Hand to mouth contamination by children frequenting sludge-treated soils in agricultural or recreational areas is of concern (DeCrosta 1981). Ingested asbestos fibers have been reported to pass through the intestinal wall and move throughout the body (Meek and Grasso 1983; Patel-Mandlik and Millette 1983; Cook and Olson 1979; Pontefract and Cunningham 1973). The incidence of cancer in various organs among workers exposed to asbestos has been reviewed (Newhouse 1981).

Additional studies should be conducted to determine the concentrations and types of fibers present in sludges of other cities especially where elevated fiber levels have been found in their drinking water. Where high concentrations of fibers occur, inhalation and ingestion studies with laboratory animals exposed to these sludges should also be conducted.

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