ARTICLE IN PRESS
DTD 5
Available online at www.sciencedirect.com
SCIENCE DIRECT
Environmental Pollution ■■ (2005) ■■■–■■■
ENVIRONMENTAL POLLUTION
www.elsevier.com/locate/envpol

Synthetic fibers as an indicator of land application of sludge

Kimberly Ann V. Zubris, Brian K. Richards*

Department of Biological and Environmental Engineering, Cornell University, Riley-Robb Hall, Ithaca, NY 14853-5701, USA

Received 3 December 2004; accepted 5 April 2005

Synthetic fabric fibers present in wastewater sludge are a semi-quantitative long-term indicator of past sludge application in soils.

Abstract

Synthetic fabric fibers have been proposed as indicators of past spreading of wastewater sludge. Synthetic fiber detectability was examined in sludges (dewatered, pelletized, composted, alkaline-stabilized) and in soils from experimental columns and field sites applied with those sludge products. Fibers (isolated by water extraction and examined using polarized light microscopy) were detectable in sludge products and in soil columns over 5 years after application, retaining characteristics observed in the applied sludge. Concentrations mirrored (within a factor of 2) predictions based on soil dilution. Fibers were detectable in field site soils up to 15 years after application, again retaining the characteristics seen in sludge products. Concentrations correlated with residual sludge metal concentration gradients in a well-characterized field site. Fibers found along preferential flow paths and/or in horizons largely below the mixed layer suggest some potential for translocation. Synthetic fibers were shown to be rapid and semi-quantitative indicators of past sludge application.

Keywords: Synthetic fabric fibers; Sludge; Soil contamination; Biosolids; Soil analysis; Wastewater sludge

1. Introduction

Agricultural lands are increasingly being sought as an outlet for the sludge by-products of wastewater treatment. The organic matter and nutrients can be beneficial for crop growth, but concerns remain over the possible detrimental impacts of long-term sludge application (National Research Council, 2002) since sludge also contains a number synthetic chemicals and potentially toxic elements (McBride, 2003). Potential adverse impacts of these contaminants include leaching to groundwater, phytotoxicity and degradation of soil quality (Harrison et al., 1999). Although increasingly monitored, the history of sludge application on a given field can be uncertain. Analysis for the presence of trace metals or trace organics can be used to infer past sludge application but can be expensive and may be inconclusive, depending on application rates and other soil amendments used. A simple, rapid and unambiguous method to detect past sludge use on a field can be of utility.

Because synthetic fibers (including nylon, rayon, polyester, etc.) that detach from clothing during laundering are non-biodegradable, they persist both in the sludge that is produced by wastewater treatment plants as well as in the treated wastewater effluent. Habib et al. (1998) showed that synthetic fabric fibers were an easily detected indicator of sludge and sewage effluent in the environment using polarized light microscopy (PLM).

However, sludge is distributed in a variety of forms due to the range of sludge stabilization processes

* Corresponding author. Tel.: +1 607 255 2463; fax: +1 607 255 4080.
E-mail address: bkr2@cornell.edu (B.K. Richards).

0269-7491/$ - see front matter
doi:10.1016/j.envpol.2005.04.013

available. Such processes typically include digestion (stabilizes sludge and reduces pathogens), dewatering (reduces mass and volume), and may include composting, pelletizing/drying, and alkaline stabilization, all of which serve to reduce pathogens and make sludge more amenable for use.

Briefly addressed by Habib et al. (1998), the effects of various sludge processing modes on the detectability of synthetic fibers need to be determined to confirm the utility of the PLM approach for the variety of sludge products. It is also important to examine samples from long-term sites to determine the effectiveness of synthetic fiber identification over time.

The purpose of this study was thus to more widely test the PLM approach to synthetic fibers as a marker of sludge application by taking advantage of a broad and unique array of archived sludge and soil samples generated in long-term studies (Richards et al., 1997, 1999, 2000; Steenhuis et al., 1999; Smith et al., unpublished data). Specific objectives were to test the effects of different methods of sludge processing on fiber detectability, and to determine long-term fiber detectability – as affected by sludge processing and soil type – in soils from both experimental columns and field sites.

2. Materials and methods

2.1. General approach

Polarized light microscopy was used to differentiate between natural and synthetic fibers in sludge and soil, as first described by Habib et al. (1998). Under polarized light, natural fibers have non-distinct edges, with a fuzzy or blurred look. Because natural fibers (such as cotton) are spun when used in textiles, fibers exhibit twists or convolutions along their length. In contrast, synthetic fibers (such as nylon, polyester, and rayon) are made by extrusion, resulting in smooth and very distinct edges (Greaves and Saville, 1995) and appear highly illuminated under polarized light, making them efficient indicators.

2.2. Equipment and slide examination

The system employed to capture experimental images was a Bausch & Lomb XB6935 light microscope (Bausch and Lomb Optical Co., Rochester, NY) with 3.5, 10 and 43× objectives and a 10× eyepiece. The microscope stage was equipped with a vernier *x*-*y* slide mount for precise control of slide movement. A Cohu high performance color CCD camera (model 8295-1000/0000) was mounted above the eyepiece. A linear polarizing filter was attached to the eyepiece in a rotating mount, with a stationary polarizing filter placed beneath the microscope stage (Models R43785 (rotating) and R53343 (stationary), both with 30% transmission and 95% polarizing efficiency; Edmund Scientific, Barrington, NJ). Image signals were converted from NTSC to RGB using a Harmonic Research Model VC101A decoder. Images were captured using *Scion Image* capture board and software for Windows (Scion Corporation, Frederick, MD) and were saved as 901 kb TIFF-format files.

Slides prepared (as described below) with fibers extracted from sludge or soil samples were examined in a uniform manner. Initially, the both surface of the slide and microscope lens surfaces were wiped with a lens paper. The slide was then brought into focus and the polarizing eyepiece filter was rotated until near-complete light extinction was achieved. The entire specimen area of each slide was examined using a "snaking" tracking pattern, beginning at the upper left hand corner of the cover glass and traversing downward, then shifting right one frame width and traversing upwards. This method was continued until the entire slide had been viewed. Whenever a fiber appeared, images were captured at 35×, 100×, and/or 430× magnification, depending on the fiber size. The fiber was then determined to be either natural or synthetic, and its description and *x*,*y* coordinates on the slide were recorded.

In an effort to thwart contamination from dust and fiber particles, special consideration was taken during the handling of all materials and samples (although no special consideration for fibers were made when the samples were originally collected for other studies). Before use, all petri dishes and flasks were rinsed with distilled water to remove any sedimentary dust. While settling, each solution was kept covered, as were all filters in between the filtering and mounting stages. Once analyzed, slides were stored in an indexed case for protection and future reference.

2.3. Initial fabric fiber examination

Six fabric samples were obtained (JoAnn Fabrics, Ithaca, NY) to allow us to observe and catalogue the visual identity of natural and synthetic fibers under PLM. Pure (100%) fiber content fabrics examined included cotton, jute (representing natural fibers), nylon, rayon, polyester and acetate (representing synthetic fibers). Microscope slides were prepared by separating single fibers from the fabric and mounting them with two drops of Permount® mounting medium (Fisher Scientific, NJ) and a cover slip. A sample of dryer lint fibers collected from a home clothes dryer following a load of mixed fabrics was also examined in a similar manner.

2.4. Fiber extraction procedure: sludge

The fiber extraction process used was based on the work of Habib et al. (1998). Duplicate extractions and slides were used for all treatments examined. For digested-dewatered sludge, 2.0 g (dry weight) of sludge was placed in a glass petri dish. Approximately 25 mL of distilled water was then slowly added with constant mixing in order to separate the solids from the fibers. Once the sludge had been thoroughly mixed, the solution was covered and allowed to settle before the supernatant containing suspended fibers was removed with a pipette. The supernatant was then vacuum-filtered through a fiber-free membrane (0.45 μm GN-6 Metricel; Pall Corporation, Ann Arbor, MI). A distilled water rinse bottle was used to wash the fibers and other retained material during filtration. The extraction process for other sludge types was similar, however pelletized sludge and alkaline-stabilized (N-Viro™ process; Burnham et al., 1992) sludge particles were first crushed with a mortar and pestle before weighing. A single drop of surfactant (store brand dishwashing liquid containing anionic and non-ionic surfactants) was added to the solution to assist wetting of the sludge, if needed. Composted sludge had been coarsely screened (6 mm grid) when originally received (Richards et al., 1997) to simulate the full-scale process wherein large wood chips are removed from the final compost product for subsequent reuse. Nevertheless, the screened composted sludge still contained an abundance of small wood chips, making it necessary to ensure the solids were thoroughly wetted to aid extraction of fibers.

Once filtration was complete, any fibers and other particles on the filter membrane were scraped from the filter onto a clean microscope slide. Depending on the amount of other particles present on the slide, three to four drops of Permount mounting medium were applied to the slide and a cover slip was put in place. For the digested-dewatered sludge, slides were also prepared using a smearing technique. A small sample of sludge was first smeared between two slides. The top slide was then removed and the smear was fixed with Permount and a cover glass. Slides were allowed to air-dry and stabilize for 2 to 3 days so that the mounted specimens would not shift during slide handling. Additional methods of sludge mounting – including mounting dry samples of each sludge and mounting the extract supernatant without filtering wet or after it had air-dried – were tested and deemed unreliable.

2.5. Fiber extraction procedure: soil

The method for extracting fibers from test soils was similar to that used for sludge, with duplicate extractions and slides made for each sample tested. All soils had previously been air-dried, ground with a mortar and pestle and passed through a 2 mm screen. For each replicate, 3.0 grams of soil was weighed into a 50 mL Erlenmeyer flask and 25 mL of distilled water was added. The flask was then swirled and aggressively agitated, and more water was added until the flask was filled to a point 0.5 cm from the top. Once the solids had settled to the base of the flask, and any fibers that may have been in the soil had been allowed to surface along with other less dense particles, water was added drop-wise until the water surface level reached the very top of the flask. This supernatant was then pipetted and filtered as before. To ensure collection of any fibers present, more water was added to the flask, with the supernatant again removed and filtered after settling. After transfer of particles and any fibers from the filter membrane onto a slide, four to five drops of Permount mounting medium was applied to the slide and a cover glass was set in place, with slides allowed to dry before analysis. Initial confirming tests of this procedure (varying the soil mass from 2 to 4 g) using sludge applied-topsoil (from Site 1 as described later) yielded 0.82 ± 0.31 fibers per g soil ($n = 8$ samples) as compared to 0.0 ± 0.0 fibers per g from a control soil ($n = 3$), a statistically significant difference ($P < 0.0001$).

2.6. Statistical analysis

Concentrations (expressed as fiber counts per g of air-dried soil) were compared for significant differences using 1-way and 2-way ANOVA analyses and Duncan tests for differences among groups, as implemented in WinSTAR v1.71 (Anderson Bell Software, Arvada CO) and Corel Quattro Pro v 9.0 (Corel Corporation). Correlation analysis (r) was performed using Corel Quattro Pro.

2.7. Experiment 1: sludge processing effects

To gain insight on the potential effects of sludge processing on fiber detectability, four different sludge types were tested. A unique resource was the existence of an array of sludge products (Table 1) all derived from a single source, as detailed in Richards et al. (1997). A single day's production of dewatered anaerobically-digested sludge from a large municipal treatment plant (Onondaga County Department of Drainage and Sanitation, Syracuse, NY) was the sole feedstock to each of the processes which included drying & pelletizing, composting with wood chips, and advanced alkaline stabilization (N-Viro process). Processing was carried out in full-scale municipal facilities with the exception of pilot-scale pelletization, and large volumes were transferred to Cornell University for use in soil column studies (Richards et al., 2000) discussed below. Subsamples of these sludge products were air-dried when initially received and were stored in sealed plastic containers at room temperature.

It should be noted that the same mass of sludge product was used for each sample, however due to amendments added and other processes occurring during original sludge processing, each product's content of dewatered sludge feedstock differed, as shown in

Table 1
Sludge products tested for fiber detectability in Experiment 1 (Richards et al., 1997)

Sludge product	Comments	Final dewatered sludge content
Dewatered	Dewatered digested municipal wastewater sludge. Served as the feedstock for other processes	1.00
Pelletized	Dried and pelletized in a pilot-scale facility	1.00
Composted	Co-composted with wood chips in a full-scale facility; large wood chips subsequently screened out	0.91
Alkaline stabilized (N-Viro™ process)	Sludge mixed with CaO and ground limestone and allowed to cure; full-scale facility	0.33

Final content of dewatered sludge in other products expressed on fractional dry weight basis.

Table 1: each gram of composted sludge represented 0.91 g of initial dewatered sludge content, while each gram of N-Viro stabilized sludge represented 0.33 g of dewatered sludge (Richards et al., 2000).

2.8. Experiment 2: long-term soil column array

Soil samples examined were archived from a long-term controlled application greenhouse experiment (Richards et al., 2000; McBride et al., 2004) that investigated the effects of sludge processing (using the sludge products detailed for Experiment 1), soil type and initial soil pH on long-term mobility and uptake. The soils tested (Table 2) were Hudson silt loam (fine, illitic, mesic Glossaquic Hapludalf) and Arkport fine sandy loam (coarse-loamy, mixed, active, mesic Lamellic Hapludalf), each at initial pH levels of 5 and 7. Large undisturbed soil columns representing each soil were extracted from the field and applied with sludge over the course of four cropping cycles. The cumulative loading for columns applied with dewatered sludge was 215 dry T/ha, with loading rates for compost and N-Viro sludges normalized to represent the same dewatered sludge content (Table 2). This accelerated loading simulated about 30 years of agronomic applications. Once sludge loadings were completed, the columns continued to be regularly watered and cropped with red clover and romaine lettuce in alternating cycles, with the surface layer (10 cm deep) tilled at the beginning of each cropping cycle. The archived soil samples tested in this study were collected at the end of cropping cycle 12 in May 2001, over five years after the final application of sludge in cropping cycle 4 in January 1996.

Samples from 24 soil columns were analyzed, representing four sludge types (dewatered, pelletized, alkaline-stabilized, composted, plus a no-sludge control), two soil types (Hudson, Arkport) and two initial soil pH values (5, and 6.5–7); the total also included two "natural control" columns for each soil type, which served as null controls throughout the experiment, with no applications of sludge, pH adjustment or nutrients throughout the duration of the experiment. Replicate extractions were prepared for each soil sample tested.

Table 2
Soil column and field site soil samples examined for Experiments 2, 3 and 4

Soil type	Sludge type	Sludge loading rate (T/ha)	Time since sludge applied (y)	Sampling depth(s) (cm)
Experiment 2: Soil column array				
Hudson silt loam & Arkport sandy loam	Dewatered	215	5	0–10 cm (tilled surface layer)
	Pelletized	215		
	Composted	237*		
	N-Viro™	645*		
	Control	0		
		* equivalent loading of dewatered sludge content		
Experiment 3: Field site 1 (Cornell Orchards)				
Hudson silt loam	Digested	244	15	Depth & flowpath test: 0–10, 10–25, 25–50 cm
	Control	0		Concentration transect: 0–25 cm
Experiment 4: Additional field sites				
Site 2: Mohawk & Honeoye clay loam	N-Viro	338	2	0–10, 10–25, 25–50 cm
	Control	0		
Site 3: Tioga silt loam	Composted	~30	0–3	0–10, 10–25, 25–50 cm
	Control	0		
Site 4: Georgia silt loam	Digested	85	8–13	0–10, 10–25, 25–50 cm
	Control	0		

2.9. Experiment 3: Cornell Orchards field site

Several groups of archived samples were examined from an old sludge application site within the Cornell University Orchards (Ithaca, NY), designated here as Site 1. The site soil type was a Hudson silt loam as previously described for Experiment 2. (In fact, the Hudson soil columns for experiment 2 were extracted from a field immediately adjacent to the Orchards field site). The samples tested were generated in a series of previous experiments (Richards et al., 1998; McBride et al., 1997; Smith et al., unpublished data). In 1978, the site was treated with a single heavy loading of sludge from the Ley Creek Treatment Plant (Syracuse, NY), applied to a 24.5×24.5 m plot at a nominal dry matter application rate of 244 dry T/ha. A subsequent tillage operation at the site included the sludge-applied area, resulting in a gradient of residual sludge-borne trace metal concentrations, with the greatest concentrations found at Row 14 (designated based on the row numbers of apple trees that were subsequently planted at the site), near the center of the original application area.

Samples examined from this site (Table 2) included soil profile samples collected during a 1995 preferential flowpath study at Row 14.5. During the 1995 study, blue dye was applied in a 0.7-m ring infiltrometer in order to demarcate preferential percolate flowpaths in the sludge plot soil. This allowed discrete sampling and comparison of trace metal concentrations in the flowpath soils vs. the bulk soil beneath the infiltrometer (Richards et al., 1998). The samples tested for fiber detection included dyed and non-dyed 0–10 and 10–25 cm depths, along with dyed 25–50 cm. Several isolated flowpath samples from greater depth were also tested to determine if fibers translocate along flow paths. Control soils (0 to 25 cm depth) were collected from a non-sludge site immediately adjacent to the Orchard.

Also examined at Site 1 was a transect of surface layer samples from row 14.5 to row 19.5 (outside the application area) collected by Smith et al. (unpublished data) in a 2003 study assessing residual impacts of sludge application on earthworm populations. Prior research showed diminishing residual sludge-borne trace metals along this transect due to tillage operations subsequent to the original application. This in effect provided a test of fiber detectability as a function of soil sludge concentration by seeing if fiber detection would correlate to the previously observed gradient in residual metals.

2.10. Experiment 4: additional field sites

Archived soil profile samples from three additional field application sites – denoted Sites 2, 3 and 4 – were examined to confirm field detectability of sludge-derived fibers. All field site samples (summarized in Table 2) were collected in 1993 as part of a larger sludge application study, with samples collected from 0–10, 10–25 and 25–50 cm depths. Site 2 samples were from test plots installed at the State University of NY at Cobleskill (Cobleskill, NY) by Dr. Douglas Goodale. The predominant soil was the clay loam subsoil of Mohawk (fine-loamy, mixed, active, mesic Mollic Hapludalf) and Honeoye series silt loams (fine-loamy, mixed, active, mesic Glossic Hapludalf). The plots were loaded in 1991 with advanced alkaline-stabilized sludge (N-Viro process) at loading rates of 0 (control) and 338 dry T/ha. The N-Viro sludge was rototilled into the soil before planting.

Site 3 samples were extracted from turfgrass soil (Tioga silt loam soil; coarse-loamy, mixed, superactive, mesic Dystric Fluventic Eutrudept) applied with in-vessel composted sludge from Endicott, NY. The sludge was surface-applied to the soil surface during late fall and early spring from 1990 to 1993, with a cumulative loading near 30 dry T/ha. Site 4 was a large test field site in Rome, NY, on Georgia silt loam soil (coarse-loamy, mixed, semiactive, mesic Aquic Dystric Eutrudepts). Liquid (5 years) and dewatered (1 year) municipal sludges were applied at cumulative rates of 85 dry T/ha of sludge dry matter (Kresse and Naylor, 1987, 1991) between 8 and 13 years before soil samples were collected. Corn (*Zea mays*) was grown during the application period using conventional tillage. After applications ceased, the area was planted with alfalfa (*Medicago sativa*) used exclusively as a hay crop.

3. Results and discussion

3.1. Initial fiber examination

Fig. 1 shows several of the initial images of natural and synthetic fibers under polarized light. It is readily apparent whether a fiber is synthetic or natural based on distinction, form and luminescence. The cotton fiber (Fig. 1a) is an example of the non-distinct edges and blurred appearance of natural fibers. Also apparent are the convolutions that occur when natural fibers are spun for use in textiles. Jute had similar physical attributes as cotton but was much thicker. Nylon fiber (Fig. 1b) is an example of how highly illuminated synthetic fibers are under PLM. The edges of the nylon fiber are smooth and distinct due to its formation by the process of extrusion. Rayon and acetate were similar to nylon, although they are not as luminescent as nylon. Polyester also had high definition, showing the tiny pits typically found on its surface (Fig. 1c).

3.2. Experiment 1: effects of sludge processing

It is important to remember that the dewatered sludge served as the feedstock for the other treatment processes

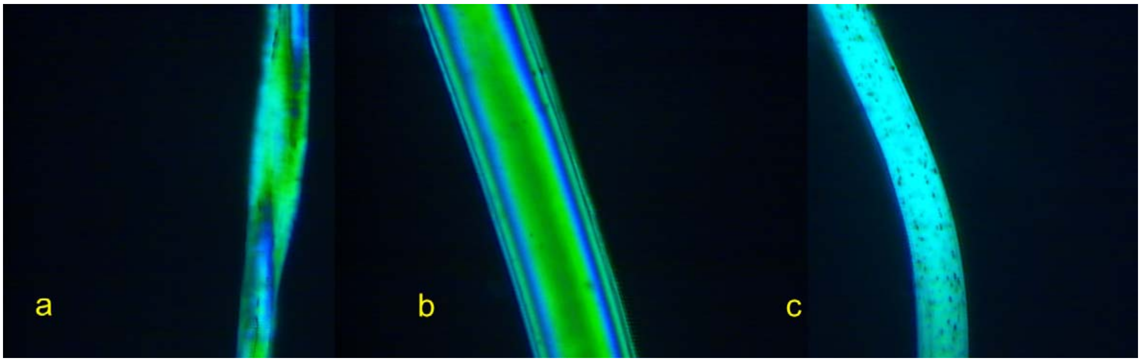

Fig. 1. Composite image showing (left to right) single fibers of a) cotton, b) nylon and c) polyester. Images captured under polarized light (at maximum extinction) at 430× magnification.

(Richards et al., 1997), so the observed effects were solely attributable to those processes. Detecting the presence of highly illuminated fibers by sequentially scanning the slide was an elementary task, even when small sludge particles were also on the slide or were partially encasing a fiber. (Even dewatered-digested sludge test slides that were prepared by a smearing process had readily visible fibers throughout.) Fig. 2 presents the mean number of fibers detected per gram of sludge dry weight. Also shown in the figure are the predicted fiber contents of the other sludge products based on the fractional dewatered sludge content (as discussed in the materials and methods section) of each section.

Synthetic fibers in the dewatered digested sludge tended to be the longest (Fig. 3a; length is described herein only qualitatively) and appeared in the greatest quantity (Fig. 2) in comparison to the other sludge products. Fibers in pelletized sludge were often found embedded in solid sludge particles (Fig. 3b). Fibers tended to be long and sometimes tangled, but showed little evidence of abrasion from processing. The fiber content of the pelletized sludge sample was somewhat lower than would be predicted from the dewatered sludge (noting that no amendments are added during pelletization), but this difference was not significant at $P = 0.05$.

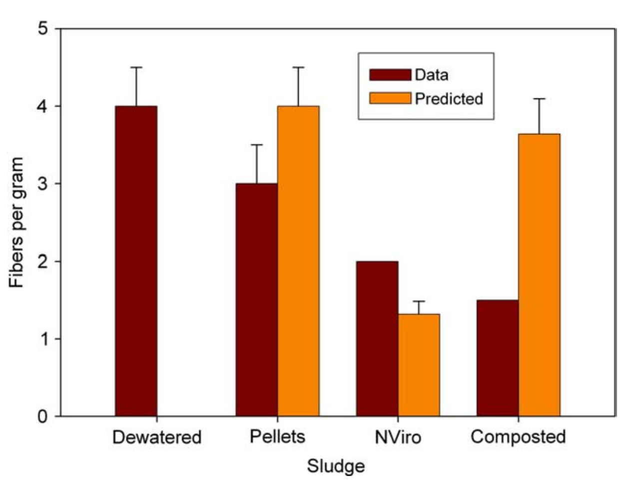

Fig. 2. Effects of sludge processing on sludge fiber concentrations (mean, with error bars representing standard deviation). Predicted values based on dewatered sludge concentration and amendment additions.

Composted sludge was the most difficult to extract due to the abundance of small floating or suspended wood chips. Fiber concentrations were significantly less than both dewatered and pelletized sludge ($P = 0.05$). Most captured fibers appeared embedded in solid particles (Fig. 3c). Fiber numbers were substantially smaller than would be predicted from the dewatered sludge content. Given the tendency of the fibers to adhere to particles, this is likely attributable to either incomplete extraction from larger particles, or disproportional losses of fibers due to adhesion to the larger wood chips that were screened out after the composting process was complete.

Fibers in the N-Viro treated sludge had a unique appearance. Almost all fibers found in this sludge product were short, exhibited high abrasion and/or erosion, and appeared very brittle (Fig. 3d). This was attributable to the elevated pH and temperature conditions and mechanical mixing that occur in the N-Viro process. Fiber breakage is thus the most likely reason for the detection of more numerous (but shorter) fibers than would be predicted from the dewatered sludge content and the substantial dilution due to amendments (Fig. 2). Resulting concentrations were statistically distinguishable only from the dewatered sludge ($P = 0.05$) An unexpected finding was the luminescence of numerous small particles present on the N-Viro slides. Examination of the amendments used in the alkaline-stabilization process indicated that particles of cement kiln dust, used in this case as a bulking agent as well as a supplemental source of process alkalinity (Richards et al., 1997), exhibited similar luminosity under polarized light.

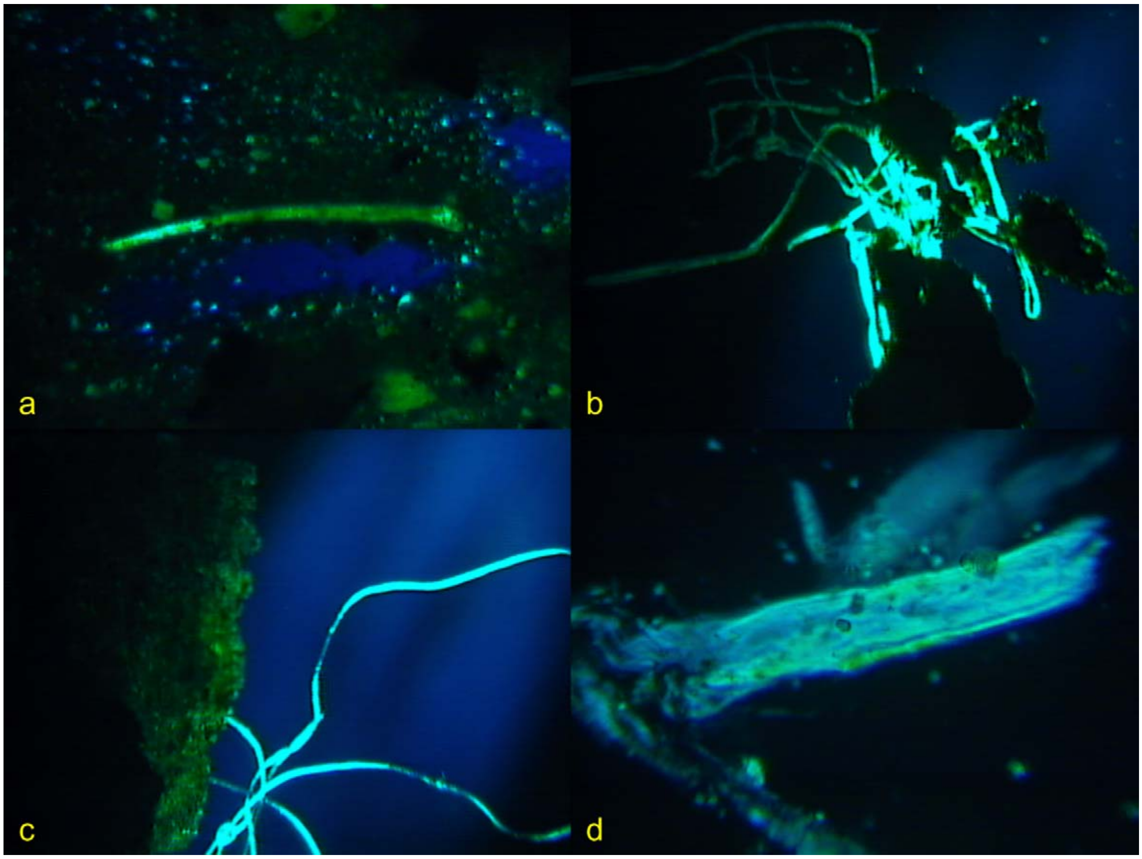

Fig. 3. Composite image of synthetic fibers extracted from sludge products; a) dewatered sludge (100×); b) pelletized sludge (35×); c) composted sludge (35×); and d) advanced alkaline-stabilized sludge (430×). Images captured under polarized light at maximum extinction.

3.3. Experiment 2: long-term soil columns array test

Soil samples tested from the long-term greenhouse soil columns array were collected five years after the final sludge application. Regular tillage, watering and cropping continued in the interim, making this a useful test of long-term fiber detectability. Fiber detection results are summarized by sludge treatment and soil type in Fig. 4. Fiber concentrations within each sludge type were statistically similar, showing no significant difference between soil type (Hudson vs. Arkport) and soil pH (5 vs. 7) treatments, the only exception being one anomalous compost treatment discussed below. No differences in fiber detection due to soil type or soil texture were observed. As a result, it was considered reasonable to pool the replicate results for the soil pH 5 and 7 soil treatments as shown in Fig. 4, as well as to pool all soil pH and soil types in order to compare soil fiber concentrations as a function solely of sludge products, as shown in Table 3.

It was reassuring that control and natural control samples showed little to no sign of fiber presence. It was in fact surprising that more fibers were not detected in the control soils, given both the large amount of "contact time" that research personnel have spent on manual tilling, harvesting, and weekly watering operations (which gave ample opportunity for clothing fibers to fall onto the columns) as well as the opportunity for cross-contamination due to columns being in close proximity in a greenhouse where air currents can be significant.

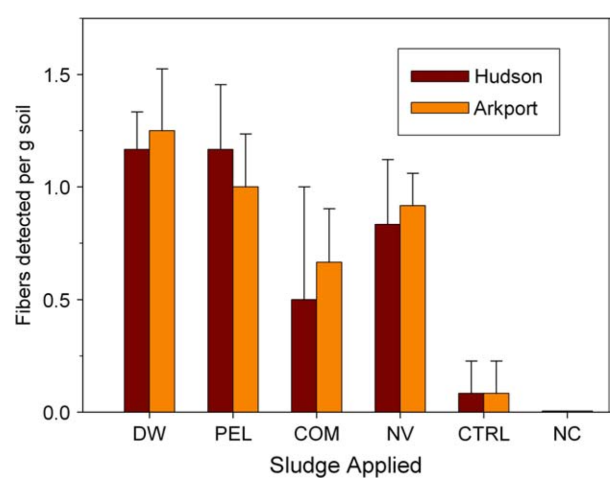

Fig. 4. Experimental soil column fiber concentrations (mean and standard deviation) more than 5 years following final sludge application, as a function of sludge type (DW dewatered, PEL pelletized, COM composted, NV N-Viro alkaline stabilized, CTRL control, and NC natural control) and soil type.

Table 3
Fiber concentrations (number per g soil) for soil column array samples pooled by sludge product type ($n=8$)

Applied sludge type		Mean fibers/g soil	Std. deviation
Dewatered	a	1.21	0.25
Pelletized	ab	1.08	0.30
N-Viro™	b	0.88	0.25
Composted	c	0.58	0.43
Control	d	0.08	0.15
Natural control	d	0.00	0.00

Products followed by similar letters were not significantly different at $P=0.05$.

Fibers in columns treated with dewatered sludge and pelletized sludge were typically observed to be long (as in the sludge products themselves) and were commonly partially embedded in soil or sludge particles. Fibers found in the compost-treated soil resembled those observed in the composted sludge, with substantial embedding in surrounding particles. They were similarly difficult to recover, resulting in one treatment (Hudson at pH 7) with no fibers detected in either replicate. As found with the sludge product itself, fibers in N-Viro treated soils exhibited the characteristic broken and worn appearance, and numerous luminescent particles assumed to be the cement kiln dust amendment were again apparent.

At the outset we assumed that the fiber analysis would simply be qualitative. An unexpected finding is shown in Fig. 5, a plot of actual vs. predicted soil fiber concentrations. Predicted soil concentrations in the soil column array samples were calculated from the actual sludge product fiber concentrations (determined in Experiment 1) adjusted by the soil dilution rate calculated from the known sludge application rates and the mixed soil layer mass, which was directly measured and recorded for each column (data not shown). As can be seen in Fig. 5, actual soil fiber concentrations generally mirrored the values predicted from the sludge fiber counts. Recoveries in fact tended to exceed the predicted values for all but N-Viro sludge, but were nevertheless within a factor of two. The reasons for detection exceeding predicted values for dewatered, pelletized and composted sludge are not known, but may be attributable to long-term sludge degradation that could allow more efficient analytical separation and recovery of fibers from the soil samples. Consistent with this is the fact that N-Viro sludge detection matched predicted values. This is unsurprising in view of the highly milled, soil-like nature of the N-Viro product coupled with apparent fiber embrittlement and breakage, which would lead to efficient recovery from the sludge product and hence little potential for an increase in recoverability over time. In any case, instead of merely being a qualitative test, the fiber detection approach can thus be safely viewed as being semi-quantitative, at the very least being able to distinguish relative rates of loading (i.e., low – medium – high).

3.4. Experiment 3: Cornell Orchards field site

The soil profile results from both Experiments 3 and 4 are summarized in Fig. 6. For the Cornell Orchards field site (Site 1), the results in Fig. 6 are separated into preferential flowpath (+f) and non-flowpath (–f), based on the presence of blue dye indicating flow. Fiber concentrations in the surface 10 cm were identical in flowpath and non-flowpath samples. Concentrations were significantly less in the 10–25 cm samples, with the flowpath sample concentrations significantly greater ($P=0.05$) than the non-flowpath samples. It is impossible

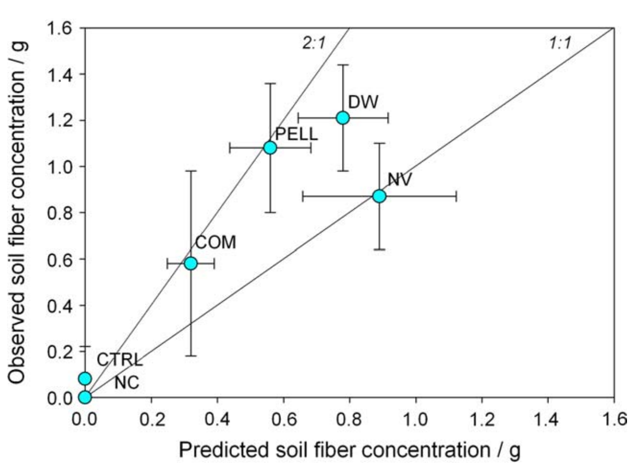

Fig. 5. Observed vs. predicted soil column fiber concentrations (mean and standard deviation) as a function of sludge type (DW dewatered, PEL, pelletized, COM composed, NV N-Viro alkaline stabilized, CTRL control, and NC natural control.

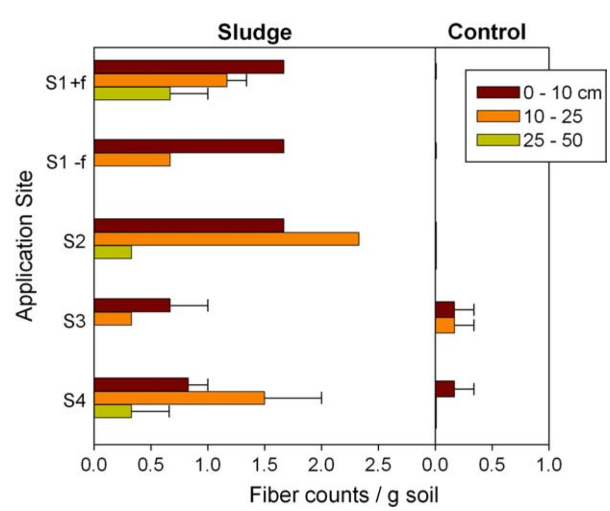

Fig. 6. Soil profiles at field sites S1, S2, S3 and S4; soil fiber concentration (mean and standard deviation) as a function of depth and (for site 1) presence (+f) or absence (–f) of preferential flowpaths.

to distinguish cause from effect at this stage – flowpaths may favor accumulation of fibers, or, conversely, the flowpath formation may have been favored by localized concentrations of sludge (and hence fibers). Although the upper fringe of the 25–50 cm soil layer at S1 may have been affected by a single deep tillage operation subsequent to sludge application, the substantial concentration of fibers in the 25–50 cm layer along the flowpath suggests that some fiber translocation may have taken place. (There was no non-flowpath 25–50 cm sample to compare to, but Richards et al. (1998) noted very little tillage-linked enhancement of trace metal concentrations in that layer.) Small fiber fragments were also observed along a dyed flowpath at 75–100 cm depth, but too few samples were tested to support any conclusions. No fibers were detected in the duplicate Hudson control samples, nor in the triplicate samples of the control soil used during initial method testing.

The results of Cornell Orchards transect samples are shown in Fig. 7. As noted previously, prior research has shown diminishing concentrations of residual sludge-borne trace metals along this transect, as illustrated by total Zn in Fig. 7 (Smith et al., unpublished data). This gradient presumably reflects the concentration of sludge in the soil and is a result of tillage (subsequent to the original sludge application study) that spread the sludge beyond the original application area. As is evident in the figure, fiber concentrations mirrored this trend, with a correlation coefficient of $r = 0.843$. Correlation with Cu (data not shown) was similar, with $r = 0.841$. Some differences are to be expected; McBride et al. (1999) estimated Zn and Cu losses of 23 and 38 percent, respectively, at Row 14 of the sludge plot, and it is not known whether losses were proportional across the transect. Nevertheless, the overall correlation between the residual soil metals and synthetic fiber counts reinforces the finding from Experiment 2 that synthetic fibers can provide at least a semi-quantitative assessment of relative sludge loadings.

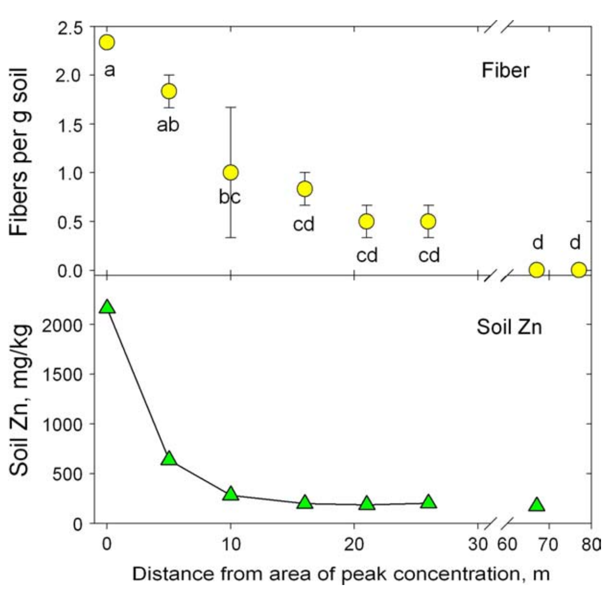

Fig. 7. Correlation of Field Site 1 fiber concentrations (mean and standard deviation) to residual sludge-borne Zn along transect. Fiber data points labeled with similar letters are not significantly different ($P = 0.05$).

3.5. Experiment 4: other long-term field sites

The results of investigations of long-term Sites 2, 3 and 4 were consistent with those reported above. Fiber concentrations in the experimental test plots at Site 2, treated with a heavy loading of N-Viro sludge, were significantly greater than controls and differed with depth. Fibers found in the sludge-treated soil were small and appeared abraded, similar to observations of fibers in N-Viro sludge products. Concentrations were greater at 10–25 cm than the 0–10 depth of the sludge plot, which may be due to the pattern of rototilling that followed application. Concentrations were much lower in the 25–50 cm layer, likely due to minimal mixing of the applied sludge into the upper portion of that layer. However, given the small size of the fiber fragments in this soil and the findings from Site 1, a degree of translocation is possible.

The light surface applications of composted sludge at Site 3 in Endicott, NY were far more difficult to detect (Fig. 6). Even though absolute fiber counts in the 0–10 cm layer of sludge plot (0.67/g soil) were greater than control (0.17), the variability in detection in the sludge plot and the finding of a single fiber in one of the control sample replicates prevented the difference from being statistically significant. Concentrations dropped by half in the 10–25 cm layer and were non-existent in the 25–50 cm layer. The results were unsurprising in view of the surface application regime, low dry matter loadings, and the dilution of sludge by wood chips in the product. Those fibers that were observed were typical of those in composted sludge in Experiment 2.

The repeated applications of liquid and dewatered sludge at the Site 4 test fields resulted in clearly detectable synthetic fibers nearly a decade after applications ceased (Fig. 6). Differences between sludge field and control field concentrations were significant ($P = 0.008$). Concentrations in the sludge-applied soil were greatest in the 10–25 cm layer, although not significantly greater than the 0–10 cm layer ($P < 0.05$). The interaction of annual applications coming from various treatment processes and tillage patterns could have contributed to this profile.

4. Conclusions and future considerations

The synthetic fiber assay developed by Habib et al. (1998) has been applied to a range of sludge treatment process and to long-term experimental/field site soils where sludge products have been applied. An array of sludge treatment processes (including dewatering, pelletization, composting, and advance alkaline stabilization) had unique effects on synthetic fiber appearance and concentrations. Most notably, composting reduced fiber numbers below levels predicted from amendment dilution due to fiber losses during wood chip separation and/or the extraction process, while advanced alkaline stabilization markedly altered fiber length and physical appearance.

Fibers from the array of sludge products were easily detectable in experimental soil columns more than five years after application, and retained the fiber characteristics observed in the applied sludge. The soil texture and soil pH treatments in Experiment 2 had no effect on fiber detectability. Fiber detection was found to be semi-quantitative, with observed soil concentrations corresponding (within a factor of two) to the concentrations predicted from applied sludge fiber concentrations and soil dilution.

Fibers were also found in soil samples from field application sites up to 15 years after application and similarly retained the characteristics of fibers in the applied sludge products. Fiber recovery was again found to be semi-quantitative, reflecting the concentration gradient of residual sludge-borne metals in a transect at Site 1. Fibers were found at depth along preferential flow paths in the strongly structured soil at Site 1 and in horizons below the mixed layer for Sites 1, 2 and 4, suggesting a potential for some downward translocation, although the extent and mechanisms are unknown at present.

Overall, the utility of synthetic fiber identification as a long-term indicator of sludge application has been confirmed for a range of sludge products. Potential applications of the test include corroboration of application records, confirmation of relative loadings among soils applied with the same sludge product, or even – given varied perceptions of risk (Beecher et al., 2005) – for confirmation that a given soil has not been applied with sludge. A planned use in our case is the determining the treatment layout of long-abandoned land application test plots (sludge, manure and control treatments) originally operated by a research group that left no known map of plot identities. Current soil trace element concentrations would allow some inferences to be made, but the fiber testing will in our case be regarded as determinative.

The relatively low number of sample replicates ($n=2$ in most cases) used in this study proved sufficient for statistically significant results in almost every case, but an increased number of replicates would be helpful where likely sludge loadings are low or unknown. It should be noted that cumulative sludge loading rates (aside from sites 3 and 4) were heavy, especially for the N-Viro treated soils. The 3 g soil samples used here yielded counts of up to 7 fibers per slide, thus larger samples would be recommended for soils with low cumulative sludge loadings. Especially in the case of composted sludge, more efficient procedures for fiber separation and recovery from samples would be useful. Although not tested here, application to other waste streams with containing fabric fibers such as septage seems reasonable. Finally, while potential interference from "false positives" was not examined in depth, no false positive conclusions were made in this study. Reasonable care will prevent contamination during sampling, processing and analysis, and in the field, only a few alternative fiber sources – such as close proximity to laundry facilities – can be envisioned.

Acknowledgements

The authors would like to acknowledge the expert imaging equipment assistance of Mr. James Throop, and the support of Drs. M. Todd Walter and Tammo S. Steenhuis, directors of the NSF-REU project in the Department of Biological & Environmental Engineering. This work was supported by the National Science Foundation – Research Experience for Undergraduates Program, grant number 0139529.

References

Beecher, N., Harrison, E., Goldstein, N., McDaniel, M., Field, P., Susskind, L., 2005. Risk perception, risk communication and stakeholder involvement for biosolids management and research. Journal of Environmental Quality 34, 122–128.

Burnham, J.C., Hatfield, N., Bennett, G.F., Logan, T.J., 1992. Use of kiln dust and quicklime for effective municipal sludge pasteurization and stabilization with the N-Viro soil process. In: Walker, D.D., Committee Cn Lime (Eds.), Innovations and Uses for Lime. ASTM Special Technical Publication No. 1135. ATSM, Philadelphia, PA, pp. 128–141.

Greaves, P.H., Saville, B.P., 1995. Microscopy of Textile Fibres. BIOS Scientific Publishers Ltd, Oxford, Great Britain, pp. 92.

Habib, D., Locke, D.C., Cannone, L.J., 1998. Synthetic fibers as indicators of municipal sewage sludge, sludge products, and sewage treatment plant effluents. Water, Air, & Soil Pollution 103, 1–8.

Harrison, E.Z., McBride, M.B., Bouldin, D.R., 1999. Land application of sewage sludge: an appraisal of the US regulations. International Journal of Environment and Pollution 11 (1), 1–36.

Kresse, E.J., Naylor, L.M., 1987. Municipal Sewage Sludge: A 7 Year Research Effort. Oneida County Cooperative Extension, Oriskany, NY.

Kresse, E.J., Naylor, L.M., 1991. Municipal Sewage Sludge: A 7 Year Research Effort Addendum Report. Oneida County Cooperative Extension, Oriskany, NY.

McBride, M.B., 2003. Toxic metals in sewage sludge-amended soils: has promotion of beneficial use discounted the risks? Advances in Environmental Research 8, 5–19.

McBride, M.B., Richards, B.K., Steenhuis, T.S., Russo, J.J., Suave, S., 1997. Mobility and solubility of toxic metals and nutrients in soil fifteen years after sludge application. Soil Science 162, 487–500.

McBride, M.B., Richards, B.K., Steenhuis, T.S., Spiers, G., 1999. Long-term leaching of trace elements in a heavily sludge-amended silty clay loam soil. Soil Science 164, 612–613.

McBride, M.B., Richards, B.K., Steenhuis, T.S., 2004. Bioavailability and crop uptake of trace elements in soil columns amended with sewage sludge products. Plant and Soil 262, 71–84.

National Research Council, 2002. Biosolids Applied to Land: Advancing Standards and Practices. National Academy of Science, Washington, DC.

Richards, B.K., Steenhuis, T.S., Peverly, J.H., Liebowitz, B.N., 1997. Effect of processing mode on trace elements in dewatered sludge products. Journal of Environmental Quality 26, 782–788.

Richards, B.K., Steenhuis, T.S., Peverly, J.H., McBride, M.B., 1998. Metal mobility at an old, heavily-loaded sludge application site. Environmental Pollution 99, 365–377.

Richards, B.K., Steenhuis, T.S., Peverly, J.H., McBride, M.B., 2000. Effect of sludge processing mode, soil texture and soil pH on metal mobility in undisturbed soil columns under accelerated loading. Environmental Pollution 109, 327–346.

Steenhuis, T.S., Peverly, J.H., Richards, B.K., 1999. Long-term fate of land-applied wastewater sludge. Final report for Project NYSERDA #1990-ERER-MW-93. New York State Energy Research and Development Authority, Albany, NY.