

Particulate matter composition and emission rates from the disk incorporation of class B biosolids into soil

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Abstract

Biosolids contain metal, synthetic organic compound, endotoxin, and pathogen concentrations that are greater than concentrations in the agricultural soils to which they are applied. Once applied, biosolids are incorporated into soils by disking and the aerosols produced during this process may pose an airborne toxicological and infectious health hazard to biosolids workers and nearby residents. Field studies at a Central Arizona biosolids land application site were conducted to characterize the physical, chemical, and biological content of the aerosols produced during biosolids disking and the content of bulk biosolids and soils from which the aerosols emanate. Arrayed samplers were used to estimate the vertical source aerosol concentration profile to enable plume height and associated source emission rate calculations. Source aerosol concentrations and calculated emission rates reveal that disking is a substantial source of biosolids-derived aerosols. The biosolids emission rate during disking ranged from 9.91 to 27.25 mg s⁻¹ and was greater than previously measured emission rates produced during the spreading of dewatered biosolids or the spraying of liquid biosolids. Adding biosolids to dry soils increased the moisture content and reduced the total PM₁₀ emissions produced during disking by at least three times. The combination of bulk biosolids and aerosol measurements along with PM₁₀ concentrations provides a framework for estimating aerosol concentrations and emission rates by reconstruction. This framework serves to eliminate the difficulty and inherent limitations associated with monitoring low aerosol concentrations of toxic compounds and pathogens, and can promote an increased understanding of the associated biosolids aerosol health risks to workers and nearby residents.

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1. Introduction

Each year greater than 5 million dry tons of class B biosolids (processed sewage sludge) are applied

onto US agricultural land to fertilize and condition soil (Goldstein, 2000). Biosolids application is typically accomplished in two stages. First, dewatered or liquid biosolids are spread onto the soil surface by a slinger or sprayer at a rate governed by agronomic nitrogen requirements (USEPA, 1994). Biosolids are then incorporated into the soil by disking the applied area. Disking provides a barrier

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to disease vectors and, by minimizing volatilization of organic and inorganic compounds, reduces odor and prevents biosolids nitrogen loss (USEPA, 1999).

Biosolids contain heavy metals, synthetic organic compounds, organic and inorganic odor-causing compounds, pathogenic protozoa, bacteria and viruses, and biotoxins in concentrations that are greater than the agricultural soils to which they are applied (Epstein, 2003; Sagic et al., 1980; USEPA, 1999, 2001). An incomplete understanding of the aerosols emitted during land application as well as anecdotal health complaints from residents near land application sites have prompted recent investigations into the emission and characterization of biosolids-derived aerosols (Lewis and Gattie, 2002; NRC, 2002). Thus far, the focus of these aerosol studies has been to characterize the aerosols emitted during the spreading process (Brooks et al., 2005; Paez-Rubio, 2006). However, the disking step also logically provides a large potential for aerosol generation (Clausnitzer and Singer, 2000). Given this potential, prudent investigation of the health risk posed by the land application processes must include a description of the aerosols generated during disking activities.

The objective of this research was to characterize the aerosols produced during the disk incorporation of class B biosolids into agricultural soils. A suite of biological (total bacteria and biosolids indicator microorganisms), chemical (metals and endotoxin), and physical (PM₁₀ and particle size distribution) aerosol measurements were performed at the aerosol source. Source emission rates were determined to support offsite aerosol modeling efforts and provide a basis for comparing aerosol production during spreading and disking. A relationship between bulk biosolids concentration and the aerosol emission rate was developed and is presented here as a framework for estimating source aerosol concentrations and emission rates of any chemical or microorganism based on the concentration of that constituent in the bulk biosolids and source PM₁₀ values.

2. Materials and methods

2.1. Field site description

Field experiments were conducted in an agricultural area located southwest of Phoenix, AZ. Soils in this area are typically classified as aridsols. Soils

are sandy loam in texture and soil moisture is commonly below 7% (AZMET, 2006). The biosolids applied onto the fields were class B and were all stabilized by mesophilic (30–35 °C) anaerobic digestion. Dewatered biosolids (20–30% solids content) were applied in all cases and the average application rate ranged from 0.8×10^4 to 1.6×10^4 kg ha⁻¹ depending on the field nutrient requirements. In all experiments, biosolids were disk incorporated into soils within 48 h after spreading.

2.2. Experimental description and aerosol sample collection

Two aerosol producing experimental conditions were considered. One termed biosolids disking corresponded to disking soils on which biosolids had been applied. The other was a control experiment in which soils were disked before biosolids had been applied. This latter experimental condition was termed control disking. All biosolids disking experiments were repeated four independent times and duplicate control soil disking experiments were performed. Each experiment was performed for approximately 10 min. In that duration, the disking equipment traveled along a line of four stands. Four passes were completed for each experiment, with each pass being 6 m further away from the samplers (Fig. 1). The line of aerosol samplers contained equipment for biological, PM₁₀, and metals measurement elevated to a breathing zone height of

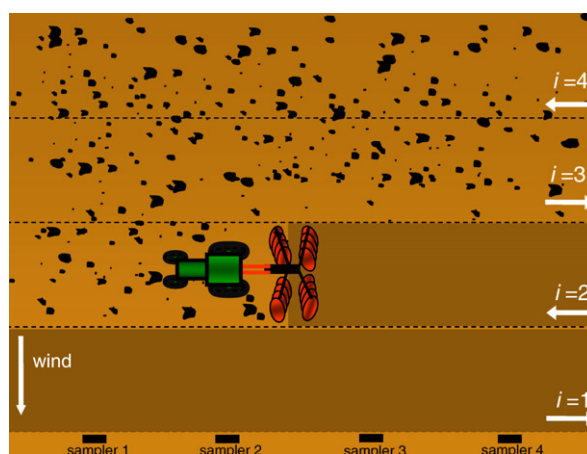


Fig. 1. Description of disking experiments. Disking equipment was located upwind of the aerosol samplers. Four passes ($i = 1-4$) were completed for each experiment and each pass increased the distance from the disking equipment to the samplers by 6 m. Average PM₁₀ concentrations in the fourth pass were approximately 80% of the PM₁₀ concentration in the first pass.

1.5 m. Source aerosol concentrations were calculated based on the period of time when the disking equipment passed each stationary sampler. This time was estimated as the duration that real-time PM_{10} concentrations were significantly higher (greater than two standard deviations above background concentrations) than ambient concentrations, and was approximately 2.7 min for each sampler in a 10 min disking experiment.

In addition to biosolids and control disking experiments, ambient concentrations of biological aerosol, PM_{10} , and airborne metals were determined. These ambient samples were taken a minimum of 100 m upwind of the disking activity and performed at the same time of the day as the disking experiments. Ambient samplers were operated for 45 min. To ensure a constant downwind flow direction and to control for wind aerosolization of land-applied biosolids, disking experiments were performed only if wind speeds were above 0.8 m s^{-1} and below 2.5 m s^{-1} . A weather station (Weather Monitor II, Davis instrument Corp., Hayward, CA) was used in each sampling event to measure and log wind speed and direction, temperature, and relative humidity. Aerosol sampling occurred only under neutral to slightly stable atmospheric conditions and experiments were conducted over smooth terrain.

For bioaerosol sampling, each aerosol experiment consisted of eight impingers total (four sampling stands each containing two impingers) (Fig. 1). Sterile liquid impingers (SKC Inc., Eighty Four, PA) collected aerosol samples for total bacteria, culturable heterotrophic bacteria (heterotrophic plate counts (HPC)), total coliforms, sulfite-reducing *Clostridia*, and endotoxin. Impingers were operated at a flow rate of 12.5 l min^{-1} in accordance with manufacture specifications and the flow rate was calibrated before each sampling event (Dry Cal DC-Lite, BIOS, Butler, NJ). The impingers were loaded with 20 ml of sterile phosphate buffer saline (PBS) solution (pH 7.2, 9.2 mM NaPO_4 , 125 mM NaCl). After sampling, the impinger contents were decanted into sterile 50 ml conical tubes and the volume recorded.

PM_{10} mass was measured at 1-s intervals using a real-time PM_{10} monitor (DustTrakTM Aerosol Monitor, Model 8520, TSI Inc., St. Paul, MN). For metal aerosol analysis, total suspended aerosol particles were collected onto a 47 mm diameter, $1 \mu\text{m}$ pore-size TeflonTM filter (Pall Corp., Ann Arbor, MI). The filter was attached to an open face

filter support and a flow rate of 31 l min^{-1} was used during collection. Finally, aerosol samples for particle size distribution measurements were collected onto 47 mm diameter, $0.4 \mu\text{m}$ pore size polycarbonate membranes (Whatman, Florham Park, NJ). These membranes were supported by polypropylene holders (Advantec MFS, Inc., Pleasanton, CA) and loaded at flow rates ranging from 11 to 15 l min^{-1} .

2.3. Bulk biosolids and soil sample collection

During each experiment, composite soil/biosolids mixture and soil samples from a minimum of five locations were collected. At least 150 g of bulk mixture were placed into sterile Whirl-Pak[®] bags (Nasco, Fort Atkinson, WI) and sealed for transportation. All aerosol, bulk soil/biosolids mixtures, and bulk soil samples were stored in the dark immediately after sampling was completed. Within 2 h of sampling, moisture content was determined gravimetrically by drying 10 g (wet weight) of soils or soil/biosolids mix for 18 h at 105°C . Soil texture was determined by sieve analysis to measure size distribution for the largest particles, and hydrometer analysis for particles smaller than $75 \mu\text{m}$ (Bardet, 1997).

2.4. Analytical methods

Culture assays for all aerosols, bulk soil/biosolids mix and bulk soil samples were started within 2 h after collection. Microorganisms were extracted from bulk soil/biosolids mixtures and soils using a $0.25 \times$ Ringer Solution (38 mM NaCl, 1.4 mM KCl, 1.1 mM CaCl_2 , 0.6 mM NaHCO_3) in accordance with previously described methods (Mocé-Llivina et al., 2003). The extract was then used for the bulk analysis of total bacteria, total coliforms, HPC, sulfite-reducing *Clostridia*, and endotoxin. For the microbial aerosol analyses, impinger contents from the same sampling event were pooled in order to obtain values above the limits of detection. The two impingers from each stand were pooled for HPC and total bacteria counts, and the contents of all eight impingers were pooled to determine total coliforms, sulfite-reducing *Clostridia*, and endotoxin concentrations.

Total bacteria were enumerated by staining nucleic acids with 4'6-diamidino-2-phenylindole (DAPI) (Pierce, Rockford, IL) at a final concentration of $20 \mu\text{g ml}^{-1}$. Stained cells were then filtered

onto a 25 mm diameter, 0.2 µm pore-size, black polycarbonate membrane (Osmonics, Inc., Minnetonka, MN) and directly counted using an epifluorescent microscope (Olympus BX51 microscope (Olympus America, Inc., Melville, NY) at 1000 × magnification (Kepner and Pratt, 1994). HPC and total coliform plate count analysis was performed in accordance with standard methods for water and wastewater samples (APHA et al., 1995). The enumeration of sulfite-reducing *Clostridia* was performed using a modified membrane filtration technique (Sartory et al., 1993). Briefly, aerosol collection liquid or solid mix extraction solutions were filtered through a 0.22 µm pore-size Durapore® membrane filter and the inoculated filter was placed on an egg yolk-free tryptose-sulphite-cycloserine (TSC) agar. Plates with filters were then incubated in an anaerobic environment at 37 °C for 48 h. Black colonies were counted as sulfite-reducing *Clostridia*. Endotoxin concentration analysis was conducted using the Limulus Amebocyte Lysate (LAL) Pyrochrome® Kit in accordance with manufacturer's instructions (ACCIUSA, Falmouth, MA). A colorimetric endpoint analysis was used by measuring adsorption at 405 nm on a 96-well V_{\max} microplate reader (Molecular Devices, Sunnyvale, CA). Endotoxin was quantified by comparing standard curves of adsorption to concentration.

To determine aerosol particle size distribution, particles collected on 0.4 µm polycarbonate filters (Whatman Inc, Florham Park, NJ) were analyzed with an automated JEOL Model JXA-8600 electron microprobe in accordance with previously describe methods (Anderson et al., 1996). Particle sizes are reported as the average geometric diameter, $(l+d)/2$ where l and d represent the length and the width of each particle counted, respectively. Particle sizes were arranged into bins of 0.1 µm increments and the percentage of particles within one bin was plotted against average geometric diameter. The geometric mean and standard deviation of the log normally distributed data as well as the percentage of particles under a specific size were calculated using statistical software (MINITAB® 14, Minitab Inc., State College, PA). Volumetric particle distributions were also calculated. The particle volume was estimated as the product of the particle area (measured during particle counting) and the particle width (d). The percent of total particle volume contained below a specified geometric diameter was calculated in accordance with previously published methods (De Nevers, 1999).

Aerosol metal concentrations were quantified using inductively coupled plasma mass spectrometry (ICP-MS) in accordance with methods for low-level aerosol particulate matter samples described by Lough et al. (2005). Briefly, filters were digested in a microwave-assisted acid bath prior to analysis. For bulk soils and bulk soil/biosolids mixtures, a representative portion of the biosolids sample (without drying) was weighed into a plastic test tube and digested with nitric acid and hydrogen peroxide in a hot block digester. The digestate was then refluxed with hydrochloric acid, cooled, and brought to a final volume of 50 ml. Metals were quantified using standard hot-plasma ICP-MS conditions. The bulk soil, bulk soil/biosolids mixture, and aerosol concentrations of the 10 metals (As, Cd, Cr, Cu, Pb, Hg, Mo, Ni, Se, Zn) that are regulated by the US EPA biosolids land application guidelines were measured.

2.5. Source emission rate calculation

Emission calculations were partially based on a previously described method for estimating PM_{10} flux produced during the tilling of agricultural soils (Holmén et al., 2001). These emission calculations utilize a continuous function to describe the PM_{10} concentration profile with height (Veranth et al., 2003). The aerosol PM_{10} emission factor, E ($mg\ m^{-2}$), was calculated as the product of the exposure time, t (s), the background corrected aerosol concentration $C(h)$ ($mg\ m^{-3}$), and the horizontal wind speed $U(h)$ ($m\ s^{-1}$) integrated from the soil roughness length, z_0 , to the height of the plume, H , and normalized by the upwind width of the disked soil:

$$E = \frac{1}{w} \int_{z_0}^H C(h)U(h)t\ dh. \quad (1)$$

Overall emission factors for each experiment were calculated as the average emission of the four passes. The vertical aerosol concentration profile was determined in triplicate independent experiments by vertically arraying (1.5, 2.7, 3.9, and 5.7 m) PM_{10} monitors at the emission source. Based on these profiles, H was defined as the height where source PM_{10} concentrations were equal to ambient PM_{10} concentrations. A first-order decay with height model provided a best fit to the vertical concentration profile measured data (Fig. 2). Using this model profile and the vertical wind profile

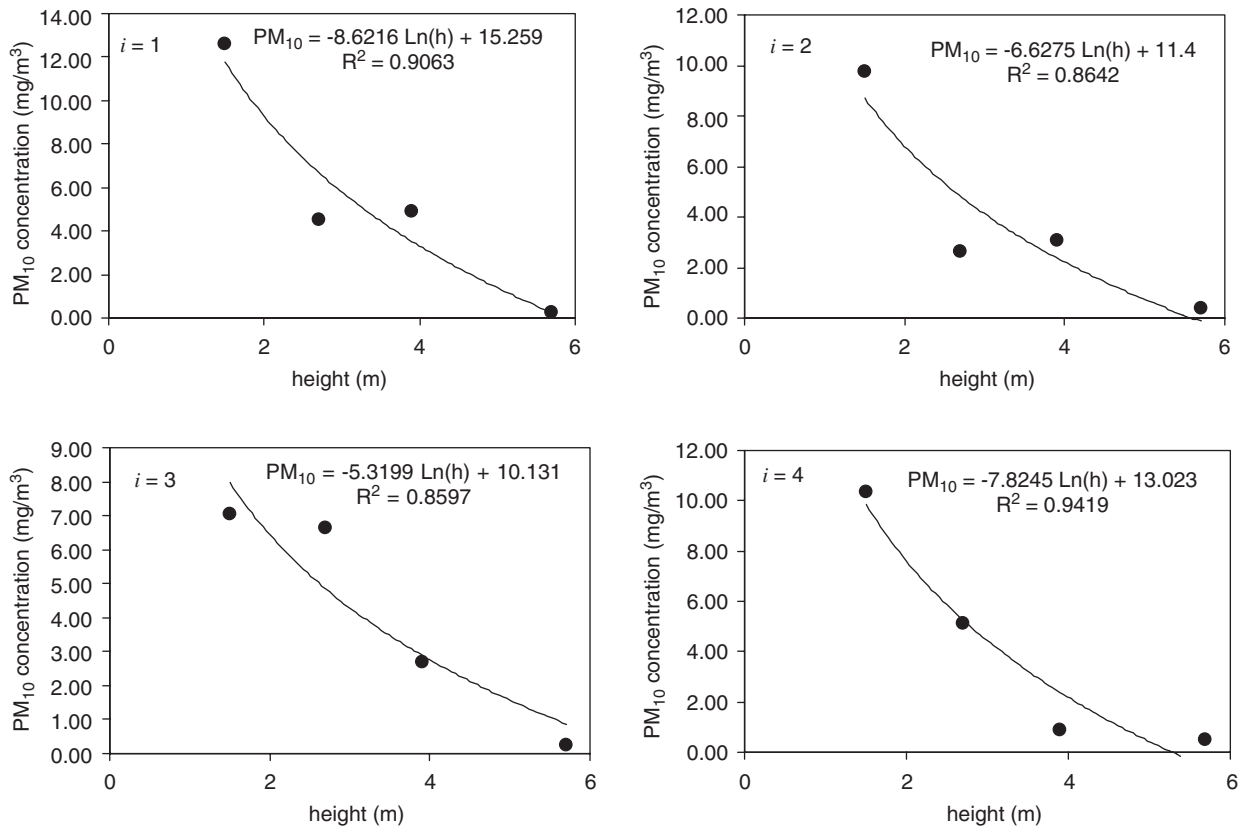


Fig. 2. Characteristic profiles describe PM₁₀ concentration decay with height at the aerosol source. The solid line represents best-fit first-order approximation. The first pass ($i = 1$) is 0–6 m from the PM₁₀ samplers, $i = 2$ is 6–12 m from the samplers, $i = 3$ is 12–18 m, and $i = 4$ is 18–24 m.

proposed by (Peterson et al., 1978) for smooth terrain, E can be expressed as

$$E = \frac{1}{w} \sum_i \int_{z_0}^{H_i} U_{H_0} \left(\frac{h}{H_0} \right)^p (a_i \ln(h) + b_i) t_i \cos \theta_i dh, \quad (2)$$

where i is each tractor's pass, H_0 is the wind speed measurement height, U_{H_0} is wind speed at H_0 , p is coefficient dependent on atmospheric stability class (Peterson et al., 1978), a and b are the slope and intercepts coefficients from the linearized first-order PM₁₀ concentration (Fig. 2), and θ is the angle between the wind direction and the plane perpendicular to the travel direction of the tractor. In order to compare emissions from biosolids disking to those of biosolids spreading, emission factors were converted to emission rates, ER (mg s⁻¹), by multiplying the emission factor by the area disked per second, A_d (m²).

PM₁₀ emissions were converted to chemical and biological emissions by first normalizing the PM₁₀

emission rates by the PM₁₀ concentration at 1.5 m and then multiplying by the average metal or biological concentration at 1.5 m (Eq. (3)). This method implicitly assumes that the vertical chemical or biological concentration profile is the same as the vertical PM₁₀ profile.

$$ER_{\text{chem/bio}} = \frac{ER_{\text{PM}_{10}}}{C_{\text{PM}_{10,1.5\text{m}}}} C_{\text{chem/bio},1.5\text{m}}. \quad (3)$$

To estimate the contribution of only biosolids to the source emission rate, the chemical and biological aerosol concentrations determined during the control disking experiments were subtracted from the biosolids disking concentrations. The aerosol concentrations produced during control disking were adjusted by multiplication with the ratio of biosolids disking PM₁₀ concentration to the control disking PM₁₀ concentration (1:3.2) to account for the aerosol production inhibition observed during biosolids disking. Error in source emission rate estimation was based on propagation of standard

deviations for each measurement through the emission calculations in accordance with accepted methods (Miller and Miller, 1993).

3. Results and discussion

3.1. Bulk sample and aerosol characterization

The mass fraction of biosolids in the soil/biosolids mixture was first estimated. Based on the dry mass of biosolids applied per soil area, volume of soil disked, and soil bulk density, biosolids composed between 4% and 11% of the dry mass of the soil/biosolids mixture. This mass fraction range was also confirmed by estimations based on the change of soil moisture content when soils were amended with biosolids.

Fig. 3a presents bulk soil and bulk soil/biosolids mixture concentrations of total coliforms, sulfite-reducing *Clostridia*, total bacteria, HPC, endotoxin, and total EPA regulated metals (As, Cd, Cr, Cu, Pb, Hg, Mo, Ni, Se, Zn). These concentrations demonstrate a slight enrichment in biological and metals content when biosolids are added to soils. Indeed, the concentrations of these microorganisms and metals in pure bulk biosolids are significantly ($p < 0.01$) greater than they are in bulk soils (Paez-Rubio, 2006). When soils are amended with biosolids, average soil/biosolids mixture concentrations were greater than average soil concentrations for all measurements; however, these increases were only significant for total coliforms ($p = 0.01$) and sulfite-reducing *Clostridia* ($p = 0.02$) due to the relatively small amount (4–11%) of biosolids added per mass soil. This increase in concentrations is listed in Table 1. For the aerosols concentrations (Fig. 3b), a concentration enrichment during the biosolids disking scenario was observed only for the biosolids indicator microorganisms (Fig. 3b). Total coliforms and sulfite-reducing *Clostridia* aerosol concentrations were 15 and 30 times greater, respectively. For total bacteria, HPC, endotoxin, and total regulated metals aerosol concentrations, no significant differences between biosolids disking and control disking were observed. This similar aerosol concentration, despite some enrichment in the bulk soil/biosolids mixture is partly a result of the lower amount of particulate matter that was aerosolized during the biosolids disking. Presumably caused by the increased moisture of the soil/biosolids mixture, the average PM₁₀ concentration measured during biosolids disking was 21% of the

concentration measured during control disking in the same soils.

Given the well-established respiratory health effects, endotoxin concentrations emitted during biosolids operations have been a particular concern. The average concentrations emitted during biosolids disking was not statistically greater than the concentration produced due to control disking and these concentrations were near 1000 EU m⁻³. These concentrations were higher than the 470 EU m⁻³ average concentrations previously measured at the source of tractor operations (no biosolids applied and level of PM₁₀ not provided) (Brooks et al., 2006). The aerosol endotoxin concentrations measured downwind of disking operations, regardless of the presence or absence of biosolids, were at least two times greater than average aerosol endotoxin concentrations produced during the loading and spreading of biosolids and greater than ten times the ambient aerosol concentrations measured at a feedyard operation (Brooks et al., 2006; Purdy et al., 2004).

Because both PM₁₀ and bulk soil/biosolids mixtures concentrations were characterized, chemical and biological aerosol concentrations could be estimated by reconstruction. Reconstructed chemical and biological source aerosol concentrations were estimated as the product of the concentrations of each chemical and biological constituent in the bulk soil/biosolids mixture and the source PM₁₀ concentrations. Comparison of reconstructed and measured aerosol concentrations reveals that they are linearly related ($r^2 = 0.78$) over the 10 order of magnitude region spanned by the biological and metal components considered. The reconstructed concentration was generally lower than measured concentrations. The average and standard deviation of reconstructed concentrations were $13 \pm 19\%$ of the measured values. Underestimation is in large part attributed to the efficiency associated with extracting chemical and biological constituents from the bulk soil/biosolids mixture. Extraction efficiencies from high organic matter matrices are well known and bacteria extraction from bulk biosolids has been documented at less than 10% efficiency (Rusin et al., 2003). These differences were less for metals (reconstructed concentrations were $18 \pm 23\%$ of measured values), where harsher extraction techniques can be used. Given that the extraction of microorganisms and chemical compounds from bulk biosolids is often less than 100%, inclusion of specific extraction efficiency in reconstruction

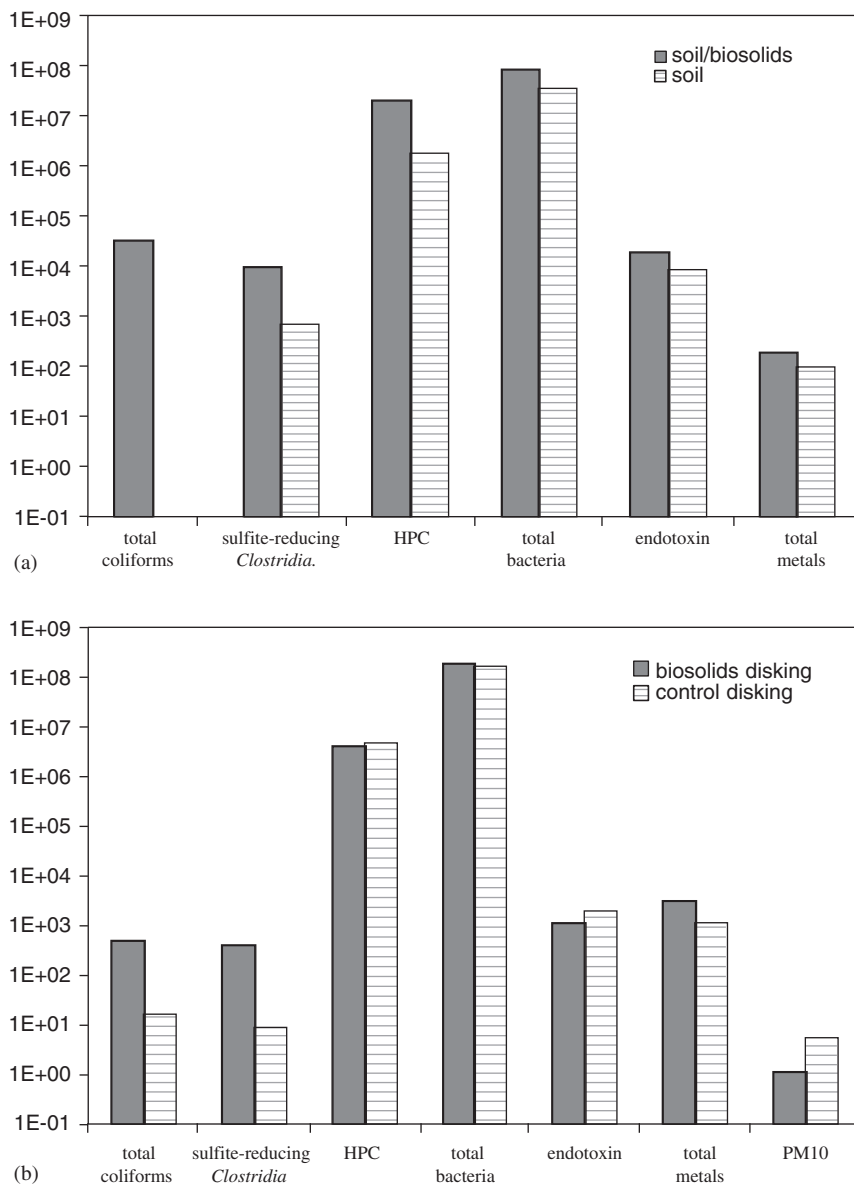


Fig. 3. (a) Average bulk soil/biosolids mixture and bulk soils concentrations for total coliforms (CFU dry g^{-1}), sulfite-reducing *Clostridia* (CFU dry g^{-1}), HPC (CFU dry g^{-1}), total bacteria (# dry g^{-1}), endotoxin (EU dry g^{-1}), and total metals (μg dry g^{-1}). (b) Average downwind source concentrations during biosolids disking and control disking for total coliforms (CFU m^{-3}), sulfite-reducing *Clostridia* (CFU m^{-3}), HPC (CFU m^{-3}), total bacteria (# m^{-3}), endotoxin (EU m^{-3}), total metals (μg m^{-3}), and PM₁₀ (mg m^{-3}).

calculations should serve to increase the agreement between reconstructed and measured values.

The experiments presented here were designed to control for soil moisture and texture in order to investigate the differences in aerosol emissions and aerosol characteristics caused by the addition of biosolids to soils. In contrast to other biosolids land application processes such as spreading and spraying, the emissions produced during biosolids disking

are a function of soil texture and soil moisture (Baker et al., 2005; Holmén et al., 2001; Smith and Lee, 2003). The addition of biosolids to soil increased the average moisture content from 4.9% to 8.0%. Correspondingly, the total average source PM₁₀ concentration during control disking was 5.12 mg m^{-3} and decreased to 1.58 mg m^{-3} during biosolids disking. Similar decreases in agriculturally produced particulate matter concentrations have

Table 1
Bulk biosolids concentrations, source emission factors and emission rates

| Parameter | Bulk soil/biosolids mixture concentration–bulk soil concentration | Aerosol source emission factor ^a | Aerosol source emission rate ^b |
|--|---|---|---|
| Total bacteria (number) | $1.55 \pm 19.7 \times 10^8$ | $1.37 \pm 2.15 \times 10^9$ | $1.09 \pm 1.72 \times 10^{10}$ |
| HPC (CFU) | $1.51 \pm 0.68 \times 10^7$ | $3.16 \pm 6.15 \times 10^7$ | $2.53 \pm 4.92 \times 10^8$ |
| Total coliforms (CFU) | $8.56 \pm 4.06 \times 10^4$ | $4.55 \pm 7.75 \times 10^3$ | $3.64 \pm 6.20 \times 10^4$ |
| Sulfite-reducing <i>Clostridia</i> (CFU) | $6.52 \pm 5.55 \times 10^3$ | $3.34 \pm 5.23 \times 10^3$ | $2.67 \pm 4.18 \times 10^4$ |
| Endotoxin (EU) | $5.7 \pm 13.8 \times 10^3$ | $1.66 \pm 2.76 \times 10^3$ | $1.33 \pm 2.21 \times 10^4$ |
| Cadmium (μg) | 0.15 ± 0.15 | $6.12 \pm 4.11 \times 10^{-2}$ | $4.90 \pm 3.28 \times 10^{-1}$ |
| Chromium (μg) | $3.61 \pm 3.49 \times 10^0$ | 2.72 ± 1.72 | $2.17 \pm 1.38 \times 10^1$ |
| Copper (μg) | $2.57 \pm 3.07 \times 10^1$ | $1.58 \pm 2.22 \times 10^1$ | $1.27 \pm 1.78 \times 10^1$ |
| Lead (μg) | $7.13 \pm 5.69 \times 10^0$ | 6.22 ± 8.07 | $4.97 \pm 6.46 \times 10^1$ |
| Mercury (μg) | $7.00 \pm 8.00 \times 10^{-2}$ | $1.64 \pm 2.25 \times 10^{-1}$ | 1.32 ± 1.8 |
| Molybdenum (μg) | 1.82 ± 1.82 | $1.57 \pm 3.56 \times 10^{-2}$ | $1.26 \pm 2.58 \times 10^{-1}$ |
| Nickel (μg) | $4.06 \pm 2.64 \times 10^1$ | 1.70 ± 1.11 | $1.36 \pm 0.89 \times 10^1$ |
| Zinc (μg) | $4.68 \pm 4.16 \times 10^1$ | $0.96 \pm 1.74 \times 10^1$ | $0.77 \pm 1.39 \times 10^2$ |
| Biosolids PM ₁₀ (mg) ^c | — | 1.24–3.41 | 9.91–27.25 |
| Total (soil and biosolids) PM ₁₀ (mg) | — | 31.0 | 247.75 |

^aAerosol source emission factor measured per m².

^bAerosol source emission rate measured per s.

^cRanges correspond to a soil biosolids mixture of 4–11% biosolids.

been observed due to an increase in soil moisture. In California's Central Valley, Clausnitzer and Singer (2000) evaluated respirable dust emissions during agricultural soil preparation at variable moisture levels and observed that the average respirable dust concentration emitted decreased five times when water content increased from 4.5% to 10%. For the disking emission results presented here, soil texture was classified as a sandy loam with 75% fine sand. We note that the addition of biosolids to soils caused small changes in soil texture and did not result in a change in textural class. Soil silt content, in particular, has been associated with higher PM₁₀ generation during soil preparation (Smith and Lee, 2003).

The particle size distribution of aerosols produced was also examined. Fig. 4 depicts the geometric diameter distribution frequency for aerosols downwind of biosolids disking and control disking operations. The log-normally distributed data demonstrate that the biosolids disking particles size distribution was similar to the distribution when control disking. The distribution's average particle geometric diameter and geometric standard deviation was $1.55 \pm 1.69 \mu\text{m}$. Greater than 99.0% of the particles emitted during disking were less than $10 \mu\text{m}$, and greater than 82% were less than $2.5 \mu\text{m}$. Greater than 82% of the particle volume

(mass) was less than $10 \mu\text{m}$. Based on methods for estimating aerodynamic diameters from geometric diameters (Chen and Fryrear, 2001), the small average geometric diameter and narrow size distribution of the disking aerosols suggest that the majority of these particles are less than $10 \mu\text{m}$ aerodynamic diameter. As a consequence of this size, particle settling at off-site setback distances of 100 m are negligible for the smooth terrain and unstable to neutral conditions tested (Etyemezian et al., 2004).

3.2. Source emission rates

The estimated biosolids source emission factors and emission rates with associated standard deviations are presented in Table 1 for total bacteria, HPC, sulfite-reducing *Clostridia*, total coliforms, endotoxin, EPA biosolids regulated metals, and PM₁₀. To address concerns over biosolids-derived aerosols, these emissions account for only the microorganisms and metals that were derived from the biosolids added to the soil, rather than the total emissions from the soil and biosolids. In Fig. 5, the log₁₀ values of emission rates are plotted versus the log₁₀ concentrations of these same microorganisms and metals that were added to the bulk soil when amended with biosolids (concentration_{soil/biosolids}

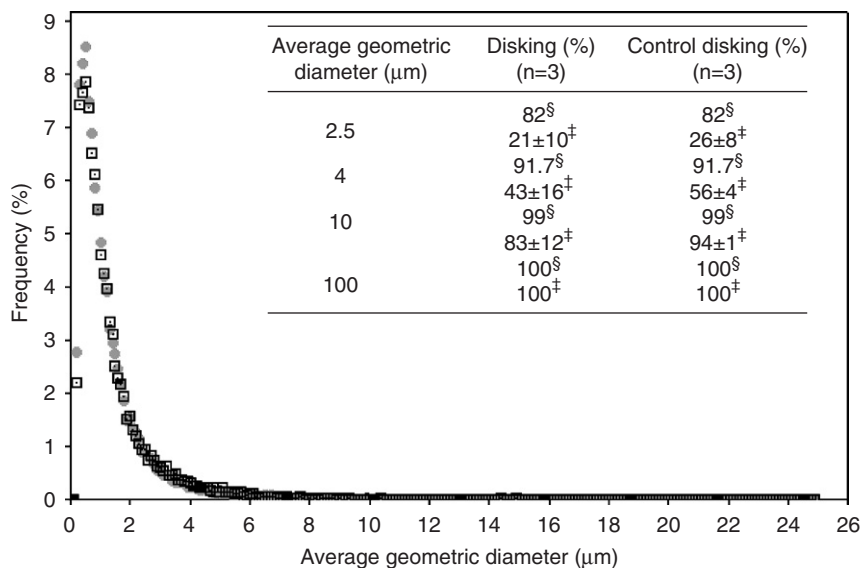


Fig. 4. Characteristic profile for the geometric diameter aerosol size frequency distribution (based on bins of 0.1 μm). Gray solid circles and black clear squares represent the particle size distribution frequency for samples taken during control disking and disking, respectively. The table presents the percentage of aerosol particles below a specific geometric diameter (§) and the percent (by volume) of aerosol particles below a specific geometric diameter (‡).

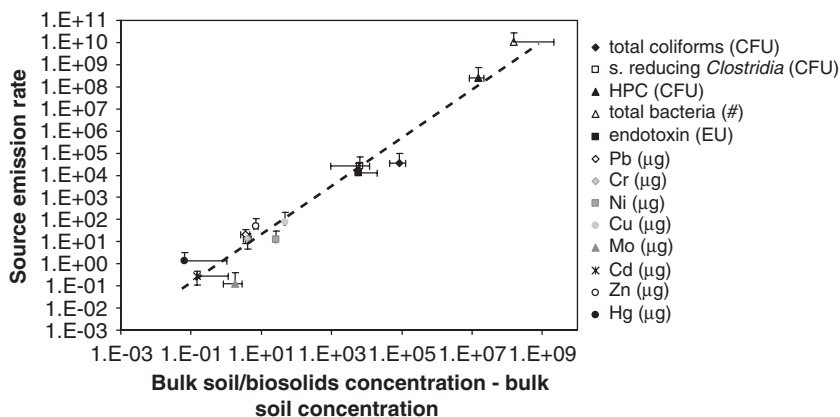


Fig. 5. Average aerosol emission rates produced during disking versus biosolids concentration. Source emission rates are units per m³, while bulk concentration are units per dry gram. Error bars represent standard deviation, $r^2 = 0.95$ and source emission rate = $10^{0.4779}(\text{biosolids concentration})^{1.0986}$.

mixture—concentration_{soil}). The plot demonstrates a correlation between the microbial and chemical concentrations emitted during disking and their content in the biosolids ($r^2 = 0.95$). The emission rates reported here, while specific to the biosolids application rate, soil moisture, and soil texture, reveal that disking can be a significant source of biosolids bioaerosols during the land application process. Although there are no other biosolids disking studies for comparison, direct measure-

ments for source emission rates during spreading of dewatered biosolids by side slinger (Paez-Rubio, 2006) and during spraying of liquid biosolids (Tanner et al., 2005) are available. Based on comparison of total coliform emission, disking biosolids emissions were approximately two times greater than spreading dewatered biosolids and at least two orders of magnitude greater than emission rates reported during liquid biosolids spraying. The emission rates reported during spraying were based

on detection limits as total coliforms were not detected in aerosol samples (Tanner et al., 2005).

Beyond the traditional scope of wastewater bioaerosol studies where typically only culturable wastewater indicators are measured and particulate matter is rarely monitored, the information obtained here provides a fundamental framework for extending biosolids and agricultural waste aerosolization research into understanding infectious and toxic characteristics of emitted aerosols. This framework would consist of estimating aerosol source concentrations and emission rates by both reconstruction and correlations between bulk soil/biosolids mixture content and emission rates, and thereby eliminate the need for direct pathogen or toxin aerosol measurement. From the standpoint of infectious agents, the aerosol emission of pathogenic microorganisms can be based on PM₁₀ measurements and pathogen concentrations in the bulk biosolids where kilograms of sample are easily obtainable. This method eliminates the expense, difficulty, and the extreme limitations of pathogen detection in aerosols. These limitations include bioaerosol sampler inefficiencies (Agranovski et al., 2004; Jensen et al., 1992), especially for viral particles less than 0.5 μm (Willeke et al., 1998), as well as the constraints associated with capturing aerosols from a mobile disking source where aerosol sampling time is limited to less than 3 min. In contrast, typical sampling durations in studies where disease-causing fungi, viruses, and bacteria have been isolated from air have ranged from 1.5 to 8 h (Chen and Li, 2005; Myatt et al., 2004; Schafer et al., 2003; Zeng et al., 2004).

Another advantage of estimating aerosol source concentrations and emission rates based on bulk biosolids concentration is that it allows for the more efficient aerosol monitoring of a diverse set of biosolids concentrations and application scenarios. Diversity in pathogen and chemical content of biosolids is caused by the variability in wastewater sources and the multitude of approved sludge stabilization methods such as anaerobic digestion, aerobic digestion, composting, air drying, and lime stabilization. Given the inherent complexity of the biosolids mixture, unanticipated changes in biosolids compositions, and the variety of in vivo responses to chemical toxins and biological pathogens, a recent National Research Council panel on biosolids safety suggested that conducting risk assessment that will produce adequate health protection may only be possible with some form

of ongoing monitoring of biosolids composition and surveillance of health effects in populations (NRC, 2002). The connection between aerosol emission and bulk biosolids concentration, as well as the possibility of reconstructing aerosol concentrations from bulk biosolids and PM₁₀ data will allow biosolids aerosol health studies to leverage the new information provided by bulk biosolids monitoring results as well as incorporate new and previously derived relationships between agricultural aerosol production and soil properties such as texture and moisture.

4. Conclusions

The sustainability of reusing biosolids by land application requires that this practice is not compromised of the health of workers and nearby residents. This study provides the first physical, biological, and chemical characterization of aerosols produced by disk incorporation of biosolids into agricultural soils. Results indicate that while biosolids depress the overall emission of particulate matter in dry soils, average concentrations of a soil/biosolids mixture greater than 1.5 mg m⁻³ can be produced. Emission rates have been estimated to enable future transport modeling in order to calculate human health risk and deposition of biosolids-derived pathogens and toxins at off-site locations. Based on emission rate comparisons, disking may represent the largest biosolids-derived aerosol-producing activity at land application sites. Correlating emission rate with bulk biosolids concentrations and reconstructing aerosol mass concentrations provides a framework for better estimating toxic or pathogenic aerosol emissions and source concentrations.

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References

- Agranovski, I.E., Safatov, A.S., Borodulin, A.I., Pyankov, O.V., Petrischchenko, V.A., Sergeev, V.A., Agafonov, A.P., Ignatiev, G.M., Sergeev, A.A., Agranovski, V., 2004. Inactivation of viruses in bubbling processes utilized for personal

- bioaerosol monitoring. *Applied and Environmental Microbiology* 70, 6963–6967.
- Anderson, J.R., Buseck, P.R., Patterson, T.L., Arimoto, R., 1996. Characterization of the Bermuda tropospheric aerosol by combined individual-particle and bulk-aerosol analysis. *Atmospheric Environment* 30, 319–338.
- APHA, AWWA, WEF, 1995. Standard methods for the examination of water and wastewater, Washington, DC.
- AZMET, 2006. The Arizona Meteorological Network. Arizona Board of Regents, <http://cals.arizona.edu/azmet/locate.html>.
- Baker, J.B., Southard, R.J., Mitchell, J.P., 2005. Agricultural dust production in standard and conservation tillage systems in the San Joaquin Valley. *Journal of Environmental Quality* 34, 1260–1269.
- Bardet, J., 1997. *Experimental Soil Mechanics*. Prentice Hall, Inc., Upper Saddle River, NJ.
- Brooks, J.R., Tanner, B.D., Josephson, K.L., Gerba, C., Haas, C.N., Pepper, I.L., 2005. A national survey on the residential impact of biological aerosols from the land application of biosolids. *Journal of Applied Microbiology* 99, 310–322.
- Brooks, J.P., Tanner, B.D., Gerba, C.P., Pepper, I.L., 2006. The measurement of aerosolized endotoxin from land application of class B biosolids in Southeast Arizona. *Canadian Journal of Microbiology* 52, 150–156.
- Chen, W.N., Fryrear, D.W., 2001. Aerodynamic and geometric diameters of airborne particles. *Journal of Sedimentary Research* 71, 365–371.
- Chen, P.-S., Li, C.-S., 2005. Quantification of airborne *Mycobacterium tuberculosis* in health care setting using real-time qPCR coupled to an air-sampling filter method. *Aerosol Science and Technology* 39, 371–376.
- Clausnitzer, H., Singer, M., 2000. Environmental influences on respirable dust production from agricultural operations in California. *Atmospheric Environment* 34, 1739–1745.
- De Nevers, N., 1999. *Air Pollution Control Engineering*. McGraw-Hill, Inc., New York.
- Epstein, E., 2003. *Land Application of Sewage Sludge and Biosolids*. Lewis, Washington, DC.
- Etyemezian, V., Ahonen, S., Nikolic, D., Gillies, J.A., Kuhns, H., Gillette, D., Veranth, J., 2004. Deposition and removal of fugitive dust in the arid Southwestern United States: measurement and model results. *Journal of the Air and Waste Management Association* 54, 1099–1111.
- Goldstein, N., 2000. Biocycle Nationwide Survey: the state of biosolids in America. *Biocycle*, 50–56.
- Holmén, B.A., James, T.A., Ashbaugh, L.L., Flocchini, R.G., 2001. Lidar-assisted measurement of PM₁₀ emissions from agricultural tilling in California's San Joaquin Valley II: emission factors. *Atmospheric Environment* 35, 3265–3277.
- Jensen, P., Todd, W., Davis, G., Scarpino, P., 1992. Evaluation of eight bioaerosol samplers challenged with aerosols of free bacteria. *American Industrial Hygiene Association Journal* 53, 660–667.
- Kepner, R.L., Pratt, J.R., 1994. Use of fluorochromes for direct enumeration of total bacteria in environmental samples: past and present. *Microbial Reviews* 58, 603–615.
- Lewis, D., Gattie, D.K., 2002. Pathogen risks from applying sewage sludge to land. *Environmental Science and Technology* 36, 286A–293A.
- Lough, G.C., Schauer, J.J., Park, J.S., Shafer, M.M., Deminter, J.T., Weinstein, J.P., 2005. Emissions of metals associated with motor vehicle roadways. *Environmental Science and Technology* 39, 826–836.
- Miller, J.C., Miller, J.N., 1993. *Statistics for Analytical Chemistry*. Horwood Limited, West Sussex, England.
- Mocé-Llivina, L., Muniesa, M., Pimenta-Vale, H., Lucena, F., Jofre, J., 2003. Survival of bacterial indicator species and bacteriophages after thermal treatment of sludge and sewage. *Applied and Environmental Microbiology* 69, 1452–1456.
- Myatt, T., Johnston, S.L., Zuo, Z., Wande, M., Kabadze, T., Rudnick, S., Milton, 2004. Detection of airborne Rhinovirus and its relation to outdoor air supply in office environments. *American Journal of Respiratory and Critical Care Medicine* 169, 1187–1190.
- NRC, 2002. *Committee on Toxins and Pathogens in Biosolids Applied to Land: Advancing Standards and Practices*. National Research Council, Washington, DC.
- Paez-Rubio, T., 2006. *Quantification of airborne biological and metals contaminants associated with land applied Class B biosolids*. Ph.D. Dissertation, Department of Civil and Environmental Engineering, Arizona State University.
- Peterson, E.W., Busch, N.E., Jensen, N.O., Hoejdyrup, J., Kridtensen, L., Peterson, E.L., 1978. The effect of local terrain irregularities on the mean wind and turbulence characteristics near the ground. *WMO boundary Layer Physics Applied to Specific Problems of Air Pollution*, International Organization, p. 45.
- Purdy, C.W., Straus, D.C., Parker, D.B., Wilson, S.C., Clark, R.N., 2004. Comparison of the type and number of microorganisms and concentration of endotoxin in the air of feedyards in the Southern High Plains. *American Journal of Veterinarian Research* 65, 45–52.
- Rusin, P., Maxwell, S., Brooks, J., Gerba, C., Pepper, I., 2003. Evidence for the absence of *Staphylococcus aureus* in land applied biosolids. *Environmental Science and Technology* 18, 4027–4030.
- Sagic, B., Duboise, S., Sorber, C., 1980. Health risks associated with microbial agents in municipal sludge. In: Bitton, G., Damron, B., Edds, G., Davidson, J. (Eds.), *Sludge-Health Risk of Land Application*. Ann Arbor Science Publisher, Ann Arbor, MI, pp. 15–46.
- Sartory, D.P., Prichard, D.P., Holmes, A.M., 1993. Enumeration of sulfite-reducing *Clostridia* from potable water supplies. *Water Science and Technology* 27, 279–282.
- Schafer, M.P., Martinez, K.F., Mathews, E.S., 2003. Rapid detection and determination of the aerodynamic size range of airborne *Mycobacteria* associated with whirlpools. *Applied Occupational and Environmental Hygiene* 18, 41–50.
- Smith, J.L., Lee, K.W., 2003. Soil as a source of dust and implications for human health. *Advances in Agronomy* 80, 1–32.
- Tanner, B.D., Brooks, J.P., Haas, C.N., Gerba, C.P., Pepper, I.L., 2005. Bioaerosol emission rate and plume characteristics during land application of liquid class B biosolids. *Environmental Science and Technology* 39, 1584–1590.
- USEPA, 1994. *Land application of sewage sludge. A guide for land appliers on the Requirements of the federal standard for the use or disposal of sewage sludge, 40 CFR part 503. EPA/831-B-93-002b*, Office of Enforcement and Compliance

- Assurance, US Environmental Protection Agency, Washington, DC.
- USEPA, 1999. Environmental regulations and technology: control of pathogens and vector attraction in sewage sludge. EPA/625/R-92-013, Office of Research and Development, US Environmental Protection Agency, Washington, DC.
- USEPA, 2001. Workshop on Emerging Infectious Disease Agents and Associated with Animal Manures, Biosolids and Other Similar By-products. USEPA National Risk Management Research Laboratory, Cincinnati, OH.
- Veranth, J.M., Seshadri, G., Pardyjak, E., 2003. Vehicle-generated fugitive dust transport: analytic models and field study. *Atmospheric Environment* 37, 2295–2303.
- Willeke, K., Lin, X.J., Grinshpun, S.A., 1998. Improved aerosol collection by combined impaction and centrifugal motion. *Aerosol Science and Technology* 28, 439–456.
- Zeng, Q., Westermark, S., Rasmuson-Lestander, A., Wang, X., 2004. Detection and quantification of *Wallemia sebi* in aerosols by real-time PCR, conventional PCR, and cultivation. *Applied and Environmental Microbiology* 70, 7295–7302.