Identifying Fly Ash at a Distance from Fossil Fuel Power Stations

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A method has been developed to identify fly ash originating at fossil fuel power stations, even at a distance where the ash level is lower by a factor of 1000 from that close to a source. Until now such detection has been difficult and uncertain. The technique combines collection of particles, measurement of magnetization and coercive field, and microscopy. The analysis depends on the fact that ash from iron sulfide in fossil fuels is in the form of spherical magnetite. These particles have a relatively high coercive field H_{c_i} near 135 Oe, compared with airborne particulates from soil erosion which have an H_c of \sim 35 Oe. The coercive field of any sample therefore gives an indication for the percentage of fly ash relative to the total amount of magnetic material that is airborne. The concentration of ash from a large, isolated coal burning power station is found to fall off with the distance from the source, approximately as D^{-1} . As D increases there is a drop in H_{c_1} associated with the reduced amount of fly ash relative to the airborne particulates from soil erosion.

I. Introduction

Airborne particulates have been shown to create significant respiratory problems (1). One of their sources is fossil fuel burned at power stations. This study has been undertaken to help answer questions about ash identification and the amount of ash to be found at a distance from its source. The method is based on the magnetic properties of ash from iron sulfide in fossil fuel when it is burned at high temperature (2)

II. Spherical Magnetic Fly Ash

On average, 16% of coal burned in a power station becomes ash (3) and approximately 16% of this ash is magnetic (4). At high temperature, FeS $_2$ in the pulverized coal is transformed into molten iron spheres and sulfur; these oxidize in the stack to form magnetite (Fe $_3$ O $_4$) and sulfur dioxide. The iron oxide ash particles become highly stressed as they rapidly solidify. The particle size distribution of the fly ash is determined mainly by the size of the pulverized coal and the temperature of combustion (5).

III. Collection

Airborne particles may be collected after they settle out on a deposition surface. If this surface is rigid and macroscopically flat, dust particles can be removed from a defined area by wiping it with a paper tissue or towel that is then rinsed in water. The material that is magnetic is withdrawn from the rinse water with a powerful permanent magnet. Pieces of plastic window screening may also be used to acquire particles and are placed directly into the rinse water for their removal of the particles.

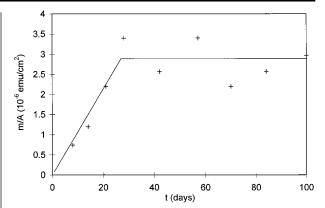


FIGURE 1. Magnetization of magnetic particles removed from pieces of plastic window screen as a function of time in Moorestown, NJ.

TABLE 1.

(a) Generator Capacity, Equilibrium Magnetic Concentration, Coercive Field and Maximum Sphere Diameter, for Particles Removed by Wiping Tree Bark 1 km Downwind from Coal Fired Power Plants

location	name	capacity (MW)	(mr A) _{eq} (µ emu/ cm²)	<i>Н</i> _с (0е)	d _{max} (μm)
Conesville, OH	Ohio Electric	2175	93	120	25
Wilsonville, AL	Gaston	2304	73	120	24
Gorgas, AL	Gorgas	1417	53	115	22
Eddystone, PA	Eddystone	1569	49	109	26
Trenton, NJ	Mercer	768	37	99	29
W. Jefferson, AL	Millera	2822	17	102	25
Demopolis, AL	Green Co.a	569	1.7	97	
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(b) (m/A)_{eq}, Coercive Field and the Percentage of Airborne Magnetic Material Relative to That in Philadelphia, PA, for Particles That Are Removed by Wiping 18-Mesh Window Screens at a Distance of More Than 10 km from a Coal Fired Power Station

location	(<i>m/A</i>) _{eq} (µemu/cm²)	to Phila., PA (%)	<i>H</i> _c (0e)	
East Coast				
Philadelphia, PA	7.4	100	82	
Baltimore, MD	2.9	40	86	
Boston, MA	2.1	30	69	
Orlando, FL	1.5	20		
Rochester, NH	0.17	2.5	77	
^a Initial operation da	ate 1965 or later.			

IV. Abundance and Deposition Rate

The amount of deposited magnetic material may be found by measuring its magnetization, its weight, or the diameter of a disk that is formed by concentrating all of the acquired particles on the bottom of a container with a standard magnet (6). In this paper we report only the magnetization m measured by the latter technique. After about 4 weeks, the amount of ash that can be removed from a surface does not significantly increase (Figure 1).

The deposition of magnetic material is expressed as the limiting magnetization m per unit acquisition area A (Table 1), $(m/A)_{\rm eq}$, and the rate as m/At (Table 2). The initial deposition rate is determined by the acquisition on a piece of 18-mesh plastic window screening as a function of time. Particles from each side of a framed window screen can be removed independently. If only one side of a vertical screen is exposed, as for an enclosed porch or a window in a building, approximately two-thirds of the particles are found on the inside surface. This relationship holds regardless of which

TABLE 2. Initial Accumulation Rate and Coercive Field for Particles Removed from Pieces of Plastic Window Screening That Are Rinsed in Water

location	m/At (µemu/cm²-day)	<i>Н</i> _с (0е)
Tuscaloosa, AL ^a	0.93 ^b	103
Lamar University, Beaumont, TX ^c	0.44	129
Philadelphia, PA	0.21	68
Seattle, WA	0.13	46
Boston, MA	0.08	75
Beaumont, TX ^d	0.06	133
Reigate, Surrey, U.K.	0.056	72
Birmingham, U.K.	0.054	56
Rosthwaite, Cumbria, U.K.	0.05	54
Oviedo, FL	0.016	71

 a 0.5 km from a coal fired steam plant that has no pollution controls and burns coal at a rate of 30–50 tons/day in the winter. b 0.24 $\mu emu/$ cm²-day when the facility is shut down in the summer. c Adjacent to petroleum processing. d 16 km from petroleum processing.

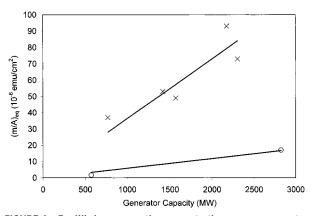


FIGURE 2. Equilibrium magnetic concentration versus generator capacity for particles removed by wiping tree bark 1 km downwind from coal fired power plants that were put into operation before (\times) and after (\bigcirc) 1965.

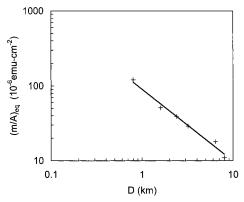


FIGURE 3. Log plot of $(m/A)_{\rm eq}$ vs the downwind distance D for ash removed by wiping trees near a coal fired power station at Conesville, OH.

side is wiped first. Two-thirds of the particles are also found on the underside of a horizontal screen that has both sides exposed. This distribution of particles probably occurs since the water droplets that accumulate following a rain shower or heavy dew are found inside an exposed vertical screen or on the underside of one that is horizontal.

The amount of material per unit area removed from a flat rigid vertical surface after a long exposure time can be correlated to that from pieces of window screening over a 28 day span. Rain washoff, adhesion, and electrostatic forces may play a role in setting this limit on particle accumulation.

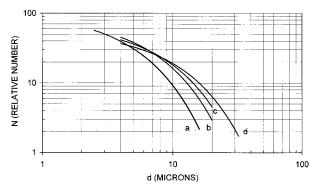


FIGURE 4. Log plot for the relative number *N* of fly ash spheres vs their diameter *d* when removed by wiping trees near (a) Tuscaloosa, AL, (b) Miller and Gorgas coal fired power stations in Alabama, (c) a plant at Gaston, AL, and (d) within a coal fired power plant at Eddystone, PA.

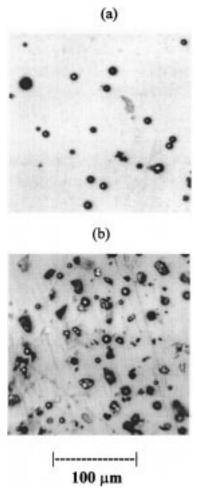


FIGURE 5. Optical micrographs of magnetic particles removed by wiping trees (a) 2 km from a coal fired utility at Gaston, AL, and (b) at Moorestown, NJ.

Varying with the strength and proximity of a source, the amount of deposited material that can be easily detected spans more than 3 orders of magnitude. Near a comparatively small Tuscaloosa, AL coal fired steam plant that has no pollution control, $(m/A)_{eq}$ on nearby trees and plastic window screens is relatively high, $36~\mu emu/cm^2$, and m/At is $0.93~\mu emu/cm^2$ -day. Approximately 75% of this dust is found to come from the local steam plant since m/At drops to $0.24~\mu emu/cm^2$ -day when the plant is idle. In the vicinity of much larger coal fired power stations that have particulate pollution controls, dust levels can be somewhat greater [Table 1a].

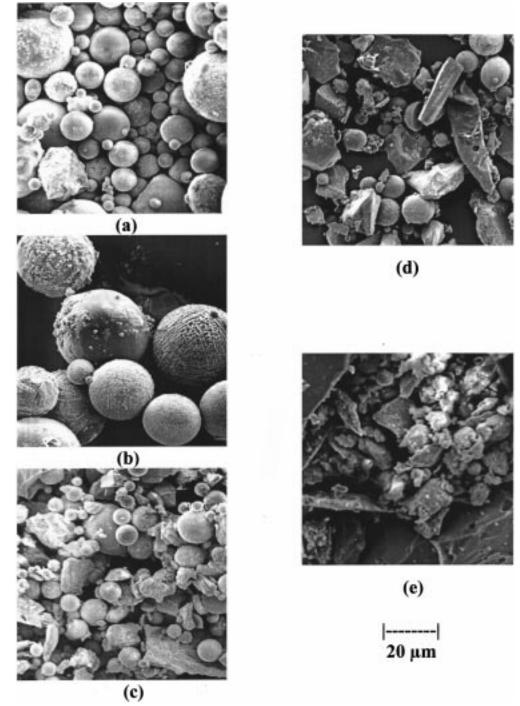


FIGURE 6. Scanning electron micrographs of magnetic particles removed from screens and by wiping trees (a) 0.8 km from Gaston, AL ($H_c = 127$ Oe), (b) 1.6 km NE of Conesville, OH ($H_c = 99$ Oe), (c) at Easthampton, NJ ($H_c = 81$ Oe), (d) at Rancocas, NJ ($H_c = 67$ Oe), and (e) at Rosthwaite, Cumbria, UK ($H_c = 54$ Oe).

Based on the age of the power stations that we checked, $(m/A)_{\rm eq}$ for each of two groups that were put into operation before and after 1965, scales with the generator capacity (Figure 2).

V. Geographic Distribution

Based on measurements made at least 10 km from a local source [Table 1b], we find that near Philadelphia, PA, many hundreds of miles downwind from midwestern coal fired plants, the magnetic particle level is comparatively high, 7.4 μ emu/cm². This relatively high level is attributed to the wind patterns from the plants located further west. North and

south of Philadelphia, along the east coast, the level drops off. Three hundred miles north, near Boston, MA, it is down to 30% of the level in Philadelphia. One hundred miles further north, near Rochester, NH it drops to only 2.5% of the Philadelphia level. One hundred miles south of Philadelphia, near Baltimore, MD, the relative level is 40%, and near Orlando, FL it is 20%.

The dependence of $(m/A)_{eq}$ on the distance (within 10 km) from an isolated coal fired power plant in Conesville, OH varies approximately as 1/D, where D is the downwind distance from the plant (Figure 3). This would occur if a small fixed percentage of the wind-blown airborne particles was deposited downwind per unit time within a given sector

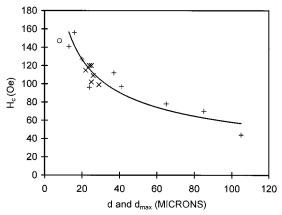


FIGURE 7. Coercive field vs the diameter of spherical particles from a stack at a coal fired plant at Eddystone, PA (+). H_c vs the maximum diameter of spherical particles, d_{max} , for samples collected 1 km downwind from coal fired power stations (×), [Table 1a], and for airborne particles near a petroleum processing plant in Beaumont, TX (\bigcirc).

angle. A similar spatial distribution has been reported for other airborne materials (7).

An estimate has been made for the average deposition rate of magnetic ash from all of the coal fired utilities in the U.S. It is based on the premise that each year in the continental U.S., electric utilities burn 830 million tons of coal (8), from which 0.25 million tons of ash particulate matter emissions less than 10 μ m (PM-10) are reported to escape into the atmosphere (9, 10). If 16% of the coal becomes ash, then only $\sim 0.2\%$ of the ash escapes from the stack. If 16% of the escaping particles have the magnetic moment of magnetite, 93 emu/g, the magnetization from coal ash would accumulate over the continental U.S. at an average rate of $0.14 \,\mu\text{emu/cm}^2\text{-day}$. This is in reasonable agreement with measurements of m/At in many places located between 10 and 100 km downwind from a coal fired utility. Typical rates (Table 2) measured near Philadelphia, PA, and Boston, MA, are 0.2 and 0.08 μ emu/cm²-day, respectively.

VI. Particle Size and Shape

Earlier reports have shown that magnetic material collected over the ocean on nylon and terylene mesh sails includes airborne particles from soil erosion in addition to fly ash (11).

To qualitatively examine how the relative amounts of these two materials vary with location, optical micrographs were taken at ${\sim}300\times$ and scanning electron microscope pictures at $1000\times$ to identify by shape the spherical ash particles and the nonspherical airborne material in the form of soil erosion minerals.

Size distributions for spherical particles are shown for magnetic ash acquired in and near several coal fired power stations (Figure 4). Maximum particle diameters are about $20-30~\mu m$, being somewhat larger in and near the plant. Diameters below $\sim 2~\mu m$ are not shown since they are below the resolution of the optical microscope used for these measurements.

Although virtually all of the collected magnetic particles that we find near coal fired utilities are spherical [Figures 5a and 6a,b], some additional irregular shaped particles are observed in samples collected at a distance from utility plants [Figures 5b and 6c,d]. In a region that is far from a coal fired utility, almost all of the magnetic particles have an irregular shape, [Figure 6e]. Progressing in order from Figure 6a—e, the coercive field is shown to decrease. The significance of this behavior is discussed in the next section.

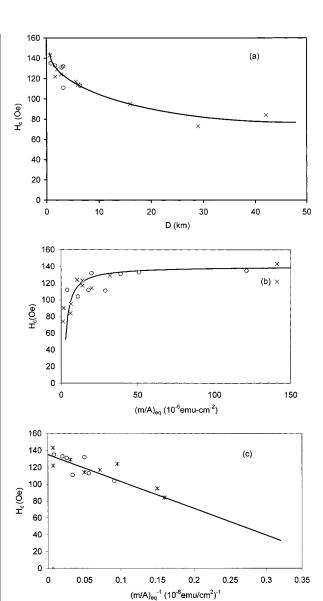


FIGURE 8. (a) H_c vs D and (b) H_c vs $(m/A)_{eq}$, for magnetic particles removed by wiping tree bark near coal fired power stations at Conesville, OH (\bigcirc) and Gaston, AL (\times), and (c) H_c vs $(m/A)_{eq}^{-1}$ using the data in Figure 8b.

VII. Coercive Field

Because of the high stress developed in spherical coal ash as it is formed, its coercive field $H_{\rm c}$ is $15\times$ greater than that for slowly cooled magnetite of comparable size and even 50% higher than for crushed Fe₃O₄ (12). Because of the relatively small amount of magnetic material that is generally collected, $H_{\rm c}$ has been measured with a high sensitivity alternating gradient magnetometer, AGM (13). The dependence of $H_{\rm c}$ on particle diameter (Figure 7) is shown for material acquired at a coal fired power station in Eddystone, PA, after separating the particles by size. Since most of the volume in a distribution comes from the largest particles, $H_{\rm c}$ for samples collected near coal fired plants can be estimated from the maximum diameter of the spherical particles, $d_{\rm max}$, as shown by the data from Table 1a that is included in Figure 7

Spherical magnetite also comes from FeS₂ in liquid fossil fuel when it is burned. It is found in abundance adjacent to a petroleum processing plant at Beaumont, TX, and has a maximum size that is smaller than that for coal ash. With $d_{\rm max}=8~\mu{\rm m}$ and a coercive field between 129 and 147 Oe, this material lies at one extreme of the $H_{\rm c}$ vs d curve for

TABLE 3. Coercive Field for Airborne Particles

 $H_{\rm c}$ (Oe)

A. Near Commercial Fossil Fuel Burning				
adjacent to petroleum processing	129-147			
at an oil burning utility	131			
coal burning utilities ^a	113			
B. Downwind and > 10 km but < 100 km from Coal Burning Power Stations				
U.S.A.				
Newtown Square, PA	93			
Boston, MA	90			
U AL Arboretum, Tuscaloosa, AL	80			
Rancocas Valley State Park, NJ	80			
Tullytown, PA	78			
Batsto, NJ	78			
Hartford, NJ	75			
Blue Mt., PA	74			
Moorestown, NJ	72			
Philadelphia, PA	70			
Medford, NJ	69			
U.K. Kew Gardens, London	93			
Embankment Gardens, London	93 90			
St. James Park, London	86			
Buckingham Palace Street, London	79			
Reigate, Surrey	72			
Windsor	69			
C. No Upwind Coal Fired Power Stations within				
Rosthwaite, Cumbria, U.K.	54			
Carkeek Park, N. Seattle, WA	46 43			
Olympic Peninsula, WA Seattle Arboretum, WA	43 42			
Bremerton, WA	31			
,	31			
D. Magnetic Soil Minerals				
volcanic ash (St. Helens)	42			
magnetite mineral crystals	33			
beach sand	25			
E. Road Dust				
Walnut Street Bridge, Philadelphia, PA	45			
bus, London, U.K.	40			
bus, Southern NJ	40			
F. Rail Wear				
British rail trains	47			
London underground	36			
^a Average for seven plants at a 1 km distance.				
Average for seven plants at a 1 km distance.				

separated coal ash particles (Figure 7). At a distance of 16 km from the plant, where the amount of ash is reduced by a factor of 7, H_c remains high, 133 Oe (Table 2).

With increasing distance from a strong source of coal ash, we observe a decrease in H_c [Figure 8a]. For the same set of samples, H_c will also fall off with a smaller $(m/A)_{eq}$ [Figure 8b]. The reason for this smaller coercive field is related to a decrease in the ratio of fly ash to airborne soil particulate. Since the slope of the second quadrant of an M vs H plot is the same for both fly ash ($H_c \sim 135$ Oe) and airborne soil erosion particles ($H_c \sim 33$ Oe), the H_c of a mixture will vary with the fraction of the two materials as the average of the two coercive fields, after weighting each by its respective $(m/A)_{eq}$. While $(m/A)_{eq}$ from the fly ash falls off with distance, $(m/A)_{eq}$ from the background soil erosion remains approximately constant. Based on this model, H_c would be expected to increase linearly from 33 to 135 Oe as the fraction of fly ash goes from 0 to 1. Conversely, H_c should decrease linearly from 135 to 33 Oe with an increasing fraction of particles from the soil. Since this fraction is simply the magnetization from the soil, assumed constant for a given area, divided by the magnetization from both sources, $H_{\rm c}$ should decrease linearly as $(m/A)_{\rm eq}^{-1}$. This behavior is confirmed [Figure 8c] using the data from Figure 8b. In the regions that surround these two sources, $(m/A)_{\rm eq}$ from the background soil erosion is found to be $\sim 3~\mu{\rm emu/cm^2}$, based on the value of $(m/A)_{\rm eq}^{-1}$ extrapolated to $H_{\rm c}=33~{\rm Oe.}$ This background level will vary from one region to another depending on the prevalence of magnetic soil minerals.

For the scanning electron micrographs in Figure 6, H_c decreases progressively from a—e. Most of the magnetic particles in Figure 6a,b are spherical; the larger particles in b have a lower H_c , as indicated in Figure 7. For Figures 6c—e, H_c is reduced as the percentage of spherical ash decreases.

Our results show that the decrease in H_c at greater D, associated with the higher percentage of soil erosion particles, generally outweighs any increase in coercive field that would come from gravitational settling of larger spheres nearer to the source.

As summarized in Table 3, we see that near coal fired stations and petroleum processing plants $H_{\rm c} > 100$ Oe. Farther from a source of fly ash, as measured at a number of locations, the range of coercive fields is between 50 and 100 Oe. Here these intermediate values generally indicate the inclusion of particles from soil erosion minerals (33 Oe), roadways (42 Oe), or rail wear (41 Oe), in addition to fly ash. At four locations in western Washington State and one in the U.K. Lake District, where there are no upwind coal fired power stations, $H_{\rm c}$ remains between 31 and 54 Oe.

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Literature Cited

- Dockery, D. W.; Pope III, A.; Xu, X.; Spengler, J. D.; Ware, J. H.; Fay, M. E.; Ferris, B. G., Jr.; Speizer, F. E. N. Engl. J. Med. 1993, 329, 1753.
- Raask, E. Mineral Impurities in Coal Combustion; Hemisphere: Washington, DC, 1988; Chapter 18.
- (3) Raask, E. Mineral Impurities in Coal Combustion; Hemisphere: Washington, DC, 1988; Chapter 15.
- (4) Norton, G. A.; Markuszewski, R.; Shanks, H. R. Environ. Sci. Technol. 1986, 20, 409.
- (5) Wibberley, L. J.; Wall, T. F. Combust. Sci. Technol. 1986, 48, 177.
- (6) Flanders, P. J. J. Appl. Phys. **1994**, 75, 5931.
- (7) Jacobson, A. R.; Morris, S. C. Air Pollution, 3rd ed.; Stern, A. C., Ed.; Academic: New York, 1976; Vol. I, p 184.
- (8) Bureau of Census, U.S. Statistical Abstract of the United States: 1977, 117th ed.; Washington, DC, 1997; Table 944.
- (9) National Air Pollution Emission Trends, 1900–1995, EPA-454/ R-96-007, October 1996; p A21, Table A-5.
- (10) Hoefnagle, G. F. Power Eng. 1997, 101, 44.
- (11) Hunt, A. Phys. Earth Planet. Inter. 1986, 42, 10.
- (12) Heider, F.; Dunlop, D.; Sugiura, N. Science 1987, 236, 1287.
- (13) MicroMag 2900 AGM; Princeton Measurements Corp.: 31 Air Park Road, Princeton, NJ 08540.

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