Atmospheric deposition by wet and dry processes is known to be an important source of several anthropogenic, particulate-bound metals in critically important waters such as the north Atlantic Ocean, the coastal mid-Atlantic waters, and the Great Lakes. Lake Michigan is especially subject to deposition of anthropogenic air pollutants as it lies in close proximity to the heavily polluted urban and industrial areas stretching from Chicago to Gary, Indiana, i.e., an area containing 20% of the US steel production.

The University of Maryland (at College Park, UMCP) aerosol chemistry group has used the NBSR reactor for instrumental neutron activation analysis to characterize atmospheric aerosol particles and gases for more than 20 years. Detailed and accurate multielement analyses are routinely achieved, non-destructively, for up to 40 elements in samples collected for periods of several hours to a few days on various types of filters and in cascade impactors which size fractionate the aerosol into as many as 10 size domains. As many as 30 elements can be determined in as little as 100 mg of a size-segregated particulate fraction, allowing the application of receptor modeling techniques to determine the sources of size segregated aerosol particles. This is extremely important in assessing dry deposition of aerosol particles and their constituents because deposition velocity is highly sensitive to particle diameter (see Fig. 1). Some of the elements measured, e.g., As, Cd, and Hg, are highly toxic and are, therefore, of epidemiological interest, especially in the Chesapeake Bay, Lake Michigan, and Coastal Marine environments. Equally important is that information on elemental constituents remains a powerful, fundamental tool with which atmospheric sources, transport, and processes may be elucidated.

As a part of EPA’s Great Water’s project, Atmospheric Exchange Over Lakes and Oceans (AEOLOS), size segregated aerosol particulate samples were collected, simultaneously, with 10-stage cascade impactors sited in south Chicago, aboard ship at sites 20 km east of Chicago, and on the Eastern shore, about 90 km down wind (Fig. 2). Sampling campaigns were conducted in spring, summer, and winter periods to observe seasonal differences in the concentrations and size distributions of inorganic elemental constituents. Several hundred size-segregated fractions were collected and analyzed by X-ray fluorescence (XRF) and Instrumental Neutron Activation Analysis (INAA).

A great deal of useful information about the sources of aerosol particles can be obtained from size spectra of constituent “marker” elements. In Fig. 3, vanadium V, a marker of particles emitted from fuel oil combustion, is bimodal. Vanadium on large (coarse) particles (i.e., those >1 μm) is associated with urban and

![FIGURE 1. Deposition velocity vs. aerodynamic particle diameter](image1)

![FIGURE 2. Location of sampling sites](image2)
rural dust particles. Vanadium on sub-micrometer (i.e., “fine”) particles was emitted from oil-fired power plants. Likewise, selenium is a marker of coal combustion particles; Zn, incineration; and As, steel mill emissions. Iron- and Mn-containing particles were also emitted from steel mills, but have substantial large particle components from this source. Manganese clearly has both a fine and coarse particle component.

Often, the size distributions of particles emitted from different sources overlap, and must be resolved by, for example, a chemical mass balance (CMB) method. The CMB involves solving a system of equations, constructed to explain the concentration of an elemental aerosol constituent at a sampling (receptor) site as a linear sum of the concentrations of elements from N number of sources for which the composition of particulate emissions is well characterized. Results for As and Zn are show in Fig. 4. This is the first successful resolution of the contributions of toxic constituents of aerosol particles by size and source. These data, and much more like it, have been used to provide the policy makers with far more accurate estimates of the deposition fluxes of toxic substances to Lake Michigan. Similar analyses are being performed by our group for the Chesapeake Bay.

REFERENCES


FIGURE 3. Concentration vs aerodynamic particle size distributions for several important “marker” elements determined from samples collected on Lake Michigan.

FIGURE 4. Distributions of As (upper) and Zn (lower) in particles sampled over Lake Michigan, by size and source.