

# Dry Deposition Fluxes and Mass Size Distributions of Pb, Cu, and Zn Measured in Southern Lake Michigan during AEOLOS

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As part of the Atmospheric Exchange Over Lakes and Oceans Study (AEOLOS) the dry deposition fluxes and atmospheric size distributions (ASDs) of anthropogenic metals were measured over the southern basin of Lake Michigan. The measurements were made during winter, summer, and fall, concurrently, in Chicago, IL; over Lake Michigan onboard the U.S. EPA RV *Lake Guardian*; and in South Haven, MI. The flux of Pb, Cu, and Zn was substantially higher in Chicago than in either South Haven or over Lake Michigan. The average measured Pb, Cu, and Zn fluxes were 0.07, 0.06, and 0.20 mg m<sup>-2</sup> day<sup>-1</sup> in Chicago; 0.003, 0.01, and 0.01 mg m<sup>-2</sup> day<sup>-1</sup> over Lake Michigan; and 0.004, 0.007, and 0.004 mg m<sup>-2</sup> day<sup>-1</sup> in South Haven. When the wind was from Chicago over the lake, the fluxes and concentrations measured over the lake were higher than when the wind was from other directions. In general, these anthropogenic metals had higher concentrations in the fine particle mode than in the coarse particle mode. Modeled and measured fluxes were in reasonable agreement. Coarse particles were found to be responsible for the majority of the flux at all locations.

## Introduction

The research described in this paper was part of the Atmospheric Exchange Over Lakes and Oceans Study (AEOLOS), whose broad goal was to assess the effect that emissions of hazardous air pollutants into the coastal urban atmosphere have on atmospheric deposition to adjacent Great Lakes. In this portion of the study, temporal and spatial variations in the dry depositional flux of metals of primarily anthropogenic origin (Pb, Cu, Zn) were measured directly with a surrogate surface (dry deposition plate) in Chicago, IL (urban site), over Lake Michigan offshore of Chicago onboard the U.S. EPA RV *Lake Guardian*, and in South Haven, MI (nonurban site).

Typically the dry deposition flux has been calculated indirectly from measured aerosol concentrations and an

assumed or modeled overall dry deposition velocity (1). A hybrid-receptor modeling approach has also been used to assess the flux to Lake Michigan (2, 3). Predicted fluxes of trace metals by Pirrone et al. (3) were in very good agreement with the direct measurements of these metals using a surrogate surface (4). This modeling effort indicated that there is increased dry deposition of trace metals to Lake Michigan when the wind is from the Chicago area.

The bulk of the previous research on these metals involved the measurement of airborne concentrations using PM<sub>10</sub> (<10 μm) and PM<sub>2.5</sub> (<2.5 μm) samplers and cascade or Micro-Orifice impactors (5–11), which provide information only on the <10 μm particle size range. Efforts at determining continuous atmospheric size distributions (ASDs) across a broader particle size range (e.g., 0.1–100 μm) have been relatively limited (12, 4). Previous research has shown that measurement of the ASD in the coarse fraction (>2.5 μm) is important since it accounts for the bulk of the dry deposition flux (13).

Atmospheric dry deposition flux has often been estimated by multiplying a measured airborne concentration by an assumed or modeled deposition velocity. Thus, the magnitude of the chosen deposition velocity has a large influence on the calculation of the flux. There is currently considerable debate among researchers about the selection of an appropriate deposition velocity and the validity of the models currently being used to predict this velocity. In this research, the Sehmel–Hodgson dry deposition velocity model (14) was used in conjunction with measured airborne concentrations to compare to the directly measured flux. The results were used to determine which size fraction of the atmospheric aerosol accounts for the bulk of the deposition.

## Experimental Section

**Measurement of Dry Deposition Flux and Ambient Particle Concentrations.** *Dry Deposition Flux.* The dry deposition flux was measured at the three locations using a dry deposition plate with a sharp leading edge mounted on a wind vane (15). The plate used in this study was similar to those used in wind-tunnel studies (16). It was made of poly(vinyl chloride) (PVC) and was 21.5 cm long, 7.6 cm wide, and 0.65 cm thick with a sharp leading edge (<10°) that was pointed into the wind by a wind vane. Each of three plates was covered on top with four Mylar strips (Graphic Art Systems, Cleveland, OH) (7.6 cm × 2.5 cm) coated with Apezion L grease (thickness ~5 μm) to collect impacted particles (123 cm<sup>2</sup> total exposed surface) (17). The strips were weighed before and after exposure to determine the total mass of particles collected. The strips were then extracted and analyzed for Pb, Cu, and Zn. A previous intercomparison of Frisbee-shaped symmetric airfoils and the knife-edge surrogate surfaces used in this study showed that the measured mass deposition was not statistically different (18).

*Ambient Particle Concentrations.* Atmospheric particles were sampled with a Noll Rotary impactor (NRI) (19, 20), and a Andersen I ACFM nonviable ambient particle-sizing sampler with a preseparator (AAPSS) (21). The NRI collects particles in a 6.5–100 μm size range, which is wider than other traditional size segregated samplers. Stages of the NRI and stages of the Andersen impactor were covered with Mylar coated with Apezion L grease to minimize particle bounce. The media were weighed before and after sampling to determine the total mass collected.

**Description of Sampling Locations, Sampling Periods, and Meteorological Conditions.** Samples were taken at three

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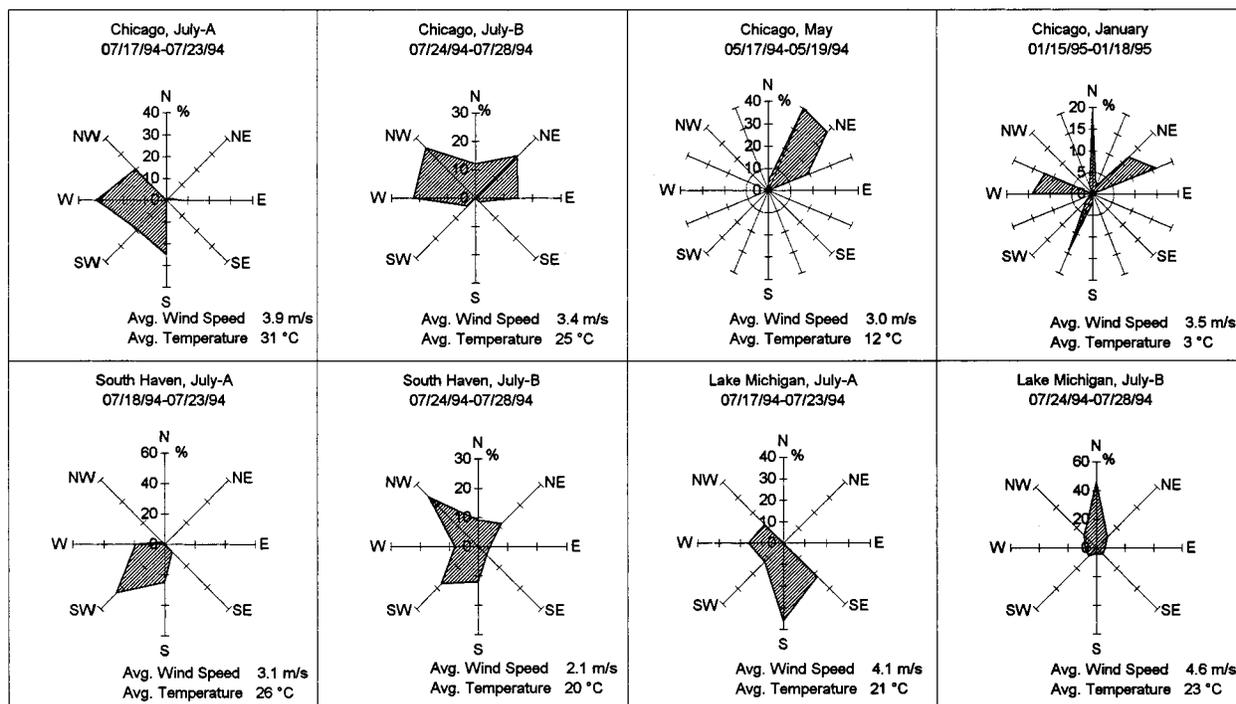


FIGURE 1. Meteorological conditions during sampling (meteorological data are arithmetic average of the dry deposition plate exposure period).

locations (Chicago, Lake Michigan, and South Haven) in May 1994, July 1994, and January 1995. Each sample set was taken over a period of 1 week (day and night) as long as there was no rain or threat of rain and included dry deposition plate, Noll Rotary (NRI), and the cascade impactor samples. Dry deposition fluxes were measured at all sites in the January period, but atmospheric size distributions were only measured in Chicago. NRI samples were obtained approximately every 2 days, each for a total of 3–23 h depending on ambient concentrations. As a result, each instrument may have a different percent exposure time, but all three instruments cover the same sampling period.

**Sampling Periods and Meteorological Conditions.** The sampling periods and the meteorological conditions prevailing during each sampling period are shown in Figure 1. The May samples were collected when the wind was from over the lake, while the July-A samples were collected when the wind was primarily from over the land (toward the lake).

**Chicago.** Samples were taken on a 1.6 m high platform on the roof of Farr Hall, a four-story (12 m height) building located in a mixed institutional, commercial, and residential area on the campus of the Illinois Institute of Technology (IIT), 5.6 km south of Chicago's center and 1.6 km west of Lake Michigan. The IIT campus consists of predominantly low-rise buildings, landscaped areas, and asphalt parking lots. The sampling site is one of five sites in the City of Chicago that are regularly monitored for PM<sub>10</sub> (1 in 6 days) by the Illinois EPA. The annual arithmetic mean PM<sub>10</sub> value for 1993 at this site was 33 μg/m<sup>3</sup>, which is the median value for the five sites (47, 34, 33, 31, and 30 μg/m<sup>3</sup>). It is classified by Illinois EPA as being primarily influenced by area sources in a primarily neighborhood environment (22). The location and the height of the sampling instruments on the platform and meteorological tower on the roof were out of roof wake boundary calculated using techniques in refs 23 and 24.

**Lake Michigan.** Samples were also taken aboard the U.S. EPA RV *Lake Guardian* offshore of Chicago between 6 and 12 km from land. The samplers were placed on the bow of the vessel at a height of 1.5 m off the deck (approximately

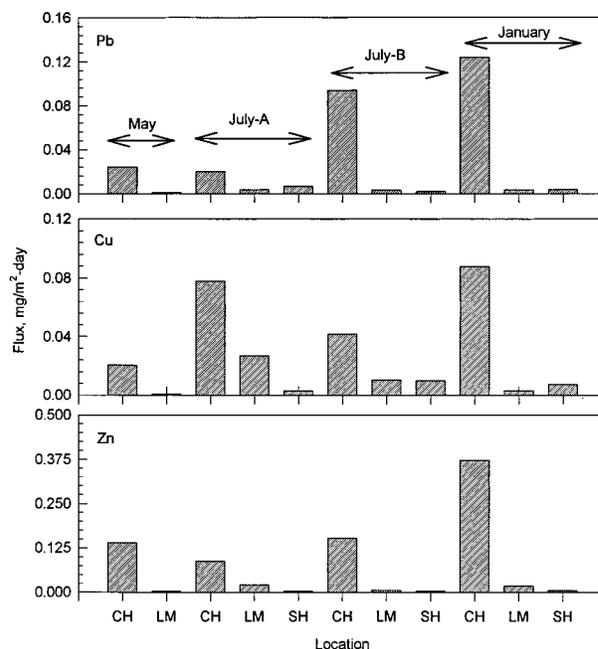


FIGURE 2. Dry deposition flux of copper, lead, and zinc measured with a dry deposition plate in Chicago, IL (CH); in South Haven, MI (SH); and over Lake Michigan (LM). May and July samples were taken in 1994, and the January sample was taken in 1995.

5 m above the water). The ship was anchored while on station and pointed into the prevailing wind at all times.

**South Haven, MI.** The third sampling site, representing nonurban conditions, was on a large platform 1.2 m high built in an open field on the Barden Farm, which is 3.6 km east of Lake Michigan. The platform is surrounded by short grass, and the farm is surrounded by rolling hills and orchards (2–4).

**Extraction and Analytical Protocol for Metals.** Extraction was carried out in the University of Michigan Air Quality

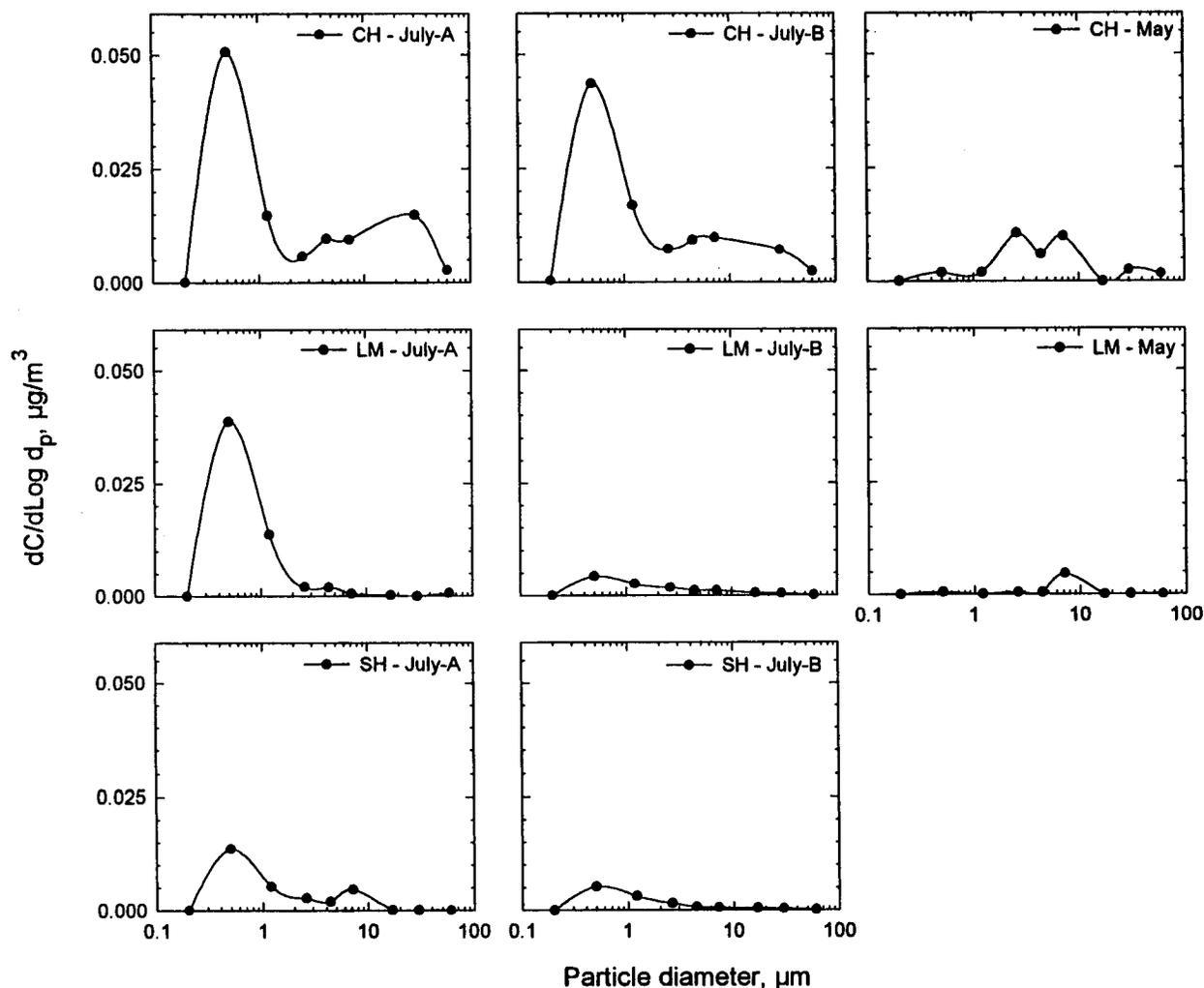


FIGURE 3. Atmospheric size distributions of lead measured with a cascade impactor and Noll Rotary impactor in Chicago, IL (CH), over Lake Michigan (LM), and in South Haven, MI (SH).

Laboratory (UMACL) in a Class 100 clean room. The procedure began with the washing of the greased Mylar strips with 10–20 mL of distilled ultrapure hexane in a Teflon vessel. The hexane was subsequently evaporated by directing a stream of ultrapure nitrogen on its surface. Next, 20 mL of 10% (v/v) ultrapure nitric acid was added; the container was placed in a Teflon bomb and loaded into the microwave oven. Acid digestion was performed for 30 min at 160 °C and approximately 160 psi, following which the Teflon bomb was allowed to cool for a period of 1 h. Samples were subsequently analyzed with a Perkin-Elmer 6000 inductively coupled plasma–mass spectrometer (ICP–MS).

The ICP–MS instrument was calibrated daily. A standard curve was deemed acceptable only if  $r^2$  was greater than 95%. After every 10 samples, a standard was analyzed as a sample. If the variation between this sample and standard concentration was more than 5%, the instrument was recalibrated. Instrument accuracy was checked daily by analyzing a 2% NIST standard to ensure that the percent recovery was between 70 and 120%. Precision was estimated by analyzing split samples (e.g., two separate strips from the same plate) and replicate sample extract analysis (same sample analyzed at different times). The relative standard deviation was less than 15% for both sample types.

Method detection limits (MDLs) for the ICP–MS were calculated by repeated analysis of a sample with concentrations near blank levels. MDL is defined as three times the standard deviation of the concentrations obtained in the

seven runs. MDLs for Pb, Cu, and Zn were 0.02, 0.03, and 0.08 ppb, respectively. Metal concentrations in the process blanks were either below or of the same magnitude as the MDLs.

Blanks were prepared as a sample, weighed, and then taken to the field in a sampling box along with the samples. They were then put on the platform with the lid open while samples were replaced and required preparations were performed for the next samples. The concentration of the metals in field blanks were slightly higher (<0.2 ppb) than the MDLs. To account for trace levels of background contamination, the mass of metal on the field blank was subtracted from the mass collected on the various stages of the cascade impactor, NRI, and flux plate. The average concentration of metals in the samples subjected to ICP–MS analyses was 10–50 times greater than the blanks (2–10 ppb). Stage values less than blanks are assumed to be less than detection limit and not reported.

Extraction efficiencies were calculated by measuring metal concentrations after spiking a 10% nitric acid solution with NIST SRM 1648. Recovery efficiencies for Pb and Cu were 98% and for Zn was 88%. No recovery correction was made.

## Results and Discussion

**Dry Deposition Flux of Metals.** Figure 2 shows the dry deposition flux of Pb, Cu, and Zn measured over the different time periods in Chicago, South Haven, and over Lake Michigan. Fluxes were substantially higher in Chicago than

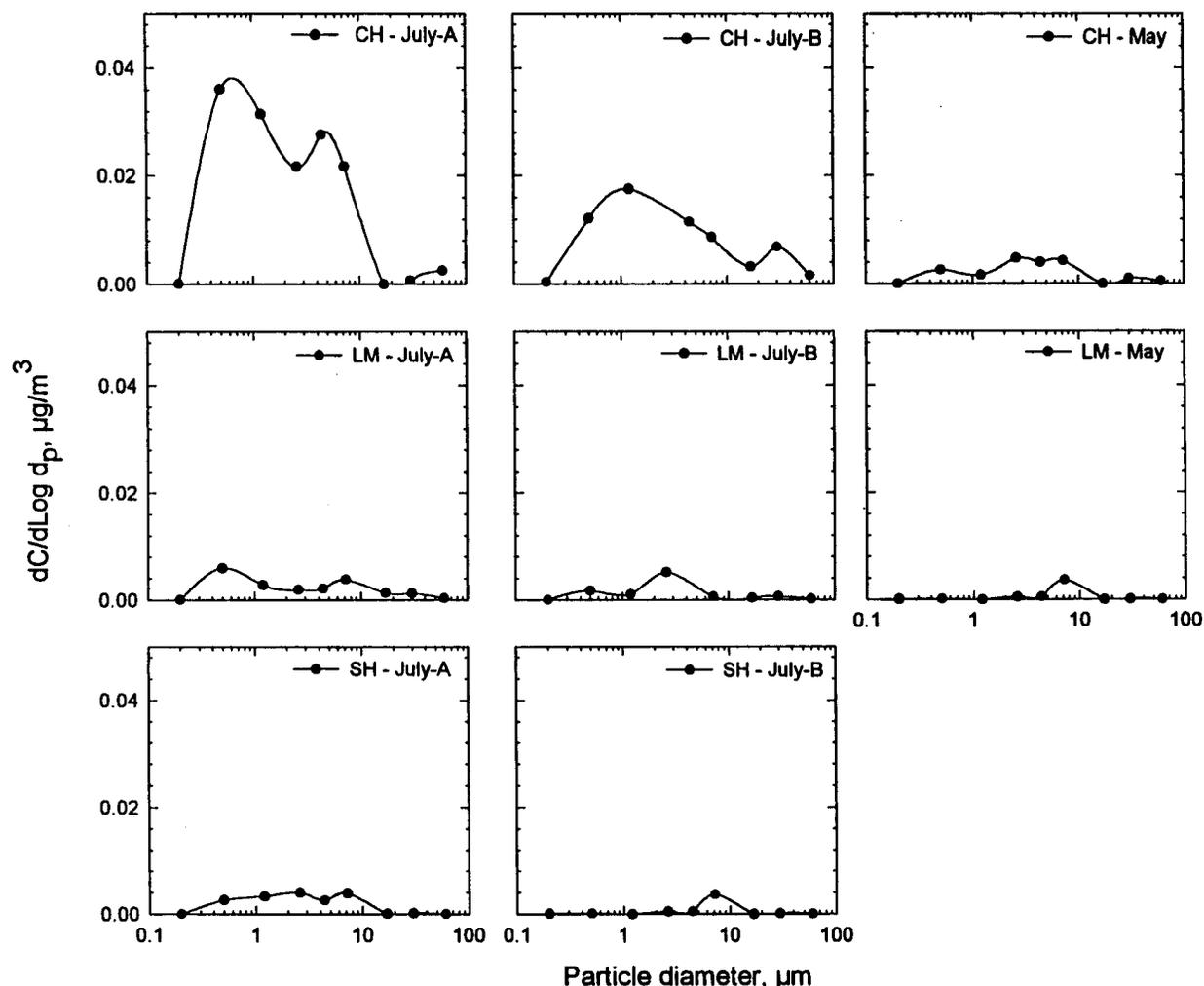


FIGURE 4. Atmospheric size distributions of copper measured with a cascade impactor and Noll Rotary impactor in Chicago, IL (CH), over Lake Michigan (LM), and in South Haven, MI (SH).

either in South Haven or over Lake Michigan. The measured average Pb, Cu, and Zn fluxes in Chicago were 0.07, 0.06, and 0.20  $\text{mg m}^{-2} \text{day}^{-1}$ ; over Lake Michigan they were 0.003, 0.01, and 0.01  $\text{mg m}^{-2} \text{day}^{-1}$ ; and in South Haven they were 0.004, 0.007, and 0.004  $\text{mg m}^{-2} \text{day}^{-1}$ , respectively. The Pb flux measured over the lake and in South Haven were comparable, while the Cu and Zn flux measured over Lake Michigan tended to be higher than in South Haven. The dry flux of Pb, Cu, and Zn over southern Lake Michigan reached levels as high as 20–30% and as low as 3–5% of the flux in Chicago. The average Zn flux was the highest of the three metals possibly because of road/wind-blown dust, the source that accounts for much of the dry deposition flux of this metal (6).

These variations in metal fluxes were strongly influenced by wind direction. In Chicago and in particular over Lake Michigan, lower metal fluxes were measured during May when the wind was primarily from the north over the lake (see Figure 1). The fluxes of Pb, Cu, and Zn over Lake Michigan were higher when the wind had a strong southwesterly and westerly component and consequently were strongly influenced by Chicago area emissions. Even though the July-A wind direction had westerly and southwesterly components (and was therefore effected by urban and industrial areas), higher fluxes were measured in the July-B period for Pb and Zn in Chicago. This difference may be because the weather was fairly stormy during the July-A sampling period and the plates were exposed for only 47% of the time, mostly during the second half of this period. The coarse ( $>2.5 \mu\text{m}$ ) and fine ( $<2.5 \mu\text{m}$ ) particle concentrations

were two times and about six times higher (25), respectively, in the first half of the sampling period (when the plates were not exposed), so it is probable that the flux was substantially higher than measured during this period.

Although Pb and Cu fluxes in Chicago are of the same magnitude, the Cu flux over the lake is higher than Pb, suggesting that the sources and transport of lead and copper are different. (This difference is probably due to their different ASDs as will be discussed below.) Zinc behaved in a manner similar to lead although its fluxes are somewhat higher. Except for the July-A sample, the zinc flux over the lake was less than 5% of the Chicago flux.

The anthropogenic metal fluxes measured here compare well with fluxes obtained in other studies. Noll et al. (26) measured Pb, Cu, and Zn fluxes of 0.12, 0.09, and 0.38  $\text{mg/m}^2\text{-d}$ , respectively, in Chicago. In another study (4), the Chicago fluxes of Pb, Cu, and Zn were 0.12, 0.16, and 0.7  $\text{mg/m}^2\text{-d}$ , respectively. The South Haven fluxes of these metals (same sequence) were 0.02, 0.03, and 0.12, respectively; and the Lake Michigan fluxes were 0.007, 0.01, and 0.025  $\text{mg/m}^2\text{-d}$ , respectively. These results compare well with the flux trends obtained by Pirrone and co-workers (3) using a hybrid-receptor deposition modeling approach. Over Lake Michigan, zinc had the highest flux followed by copper and lead.

The atmospheric deposition into Lake Michigan was estimated by Eisenreich and Strachan (1) in 1992. The estimated lakewide dry deposition flux of Pb is approximately 5 times lower than the average flux measured over the

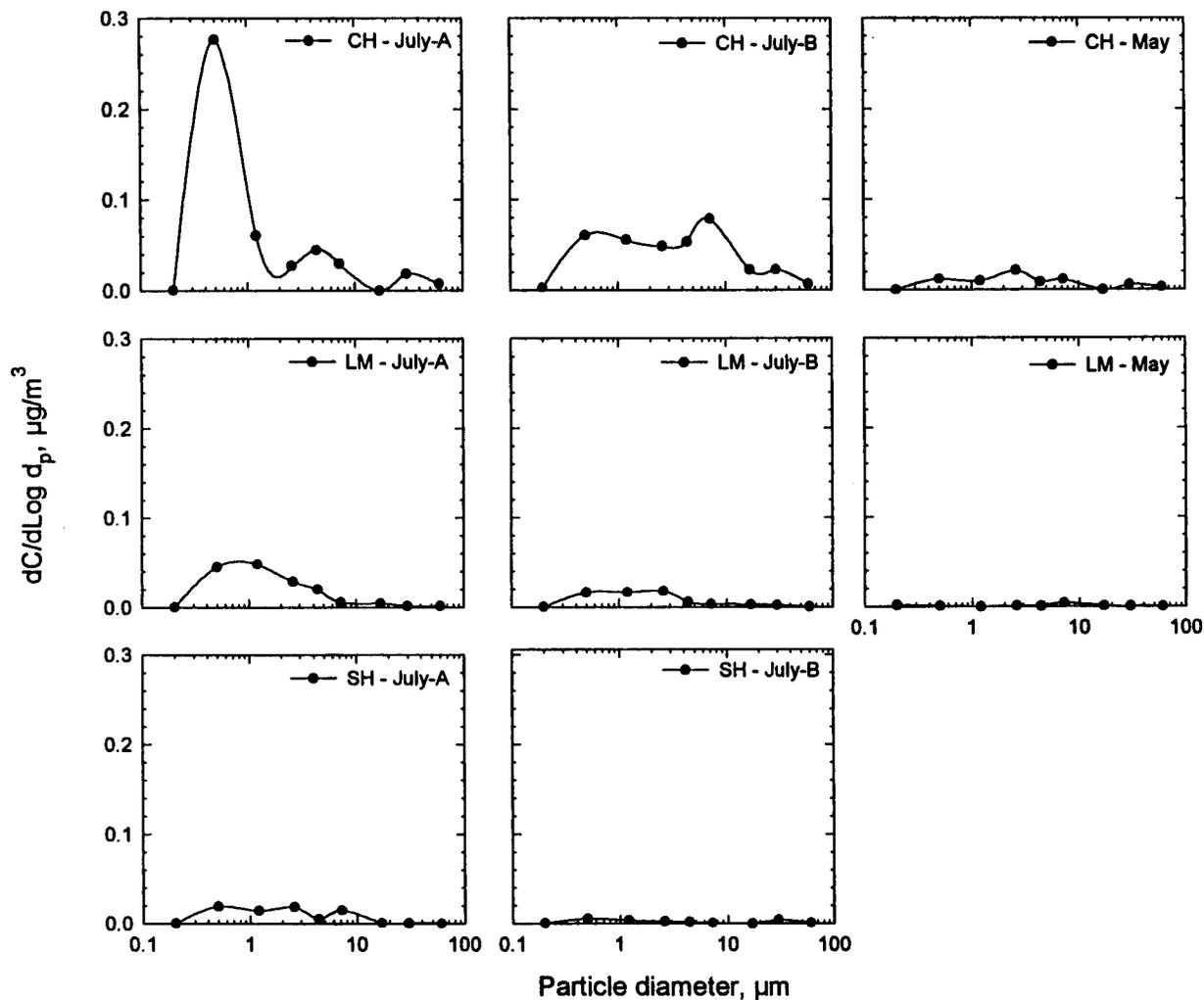


FIGURE 5. Atmospheric size distributions of zinc measured with a cascade impactor and Noll Rotary impactor in Chicago, IL (CH), over Lake Michigan (LM), and in South Haven, MI (SH).

southern portion of Lake Michigan and approximately 100 times lower than the fluxes measured in Chicago. This difference can be attributed, in part, to the large urban signal, which was not taken into account in their calculations. In addition, their calculation technique accounts for only the flux due to fine particles.

**Elemental Size Distributions.** Figures 3–5 present the ASDs (between 0.1 and 100  $\mu\text{m}$ ) of Pb, Cu, and Zn respectively, measured simultaneously in Chicago, over Lake Michigan, and in South Haven during May and July 1994. The figures show ASDs for a singular site at various time periods in horizontal panels and for a specific time period at various sites in vertical panels.

Variations in the July-A ASDs, measured concurrently when the wind was primarily from over the land and toward the lake, show decreasing particle concentrations from Chicago to the RV *Lake Guardian* and South Haven. In particular, the coarse fraction concentration decreased, probably because the majority of biggest coarse particles deposited to the lake. Part of the difference may also be due to exposure of Lake Michigan samples to southerly and southeasterly winds along with westerly components (Figure 1). The fine fraction concentrations measured over the lake were also somewhat lower than those in Chicago.

A significant feature of the spatial ASD profile is the presence of substantial amounts of anthropogenic metals in the coarse fraction ( $>2.5 \mu\text{m}$ ) in Chicago and to a lesser extent over Lake Michigan and in South Haven. Receptor modeling

studies (8) have suggested that the presence of anthropogenic metals in the coarse fraction is primarily from dust emissions from paved and unpaved roads and industries.

These ASDs help explain why the Pb flux over Lake Michigan was much lower than the copper flux even though the Chicago fluxes of these metals were similar. There is relatively higher concentrations of copper in the coarse fraction than there is of lead in the ASDs measured over the lake during July-A and -B. The elemental ASD measured in Chicago and over the lake during the May period, when the wind was from the north, had a relatively flat profile, particularly in Chicago. However, the presence of a significant coarse fraction accounts for the elemental flux in May being similar to that in July, particularly over the lake.

Previous studies have found a bimodal distribution (two peaks) for all anthropogenic metals, with a primary peak in the 0–1  $\mu\text{m}$  range and a secondary peak in the 8–16  $\mu\text{m}$  (26) or the 3–5  $\mu\text{m}$  (11) particle size range. Lundgren and Paulus (27) have also stated that the ASDs are bimodal. Most of the distributions measured in this study have either one or two peaks. However in some cases three peaks were measured; the first in the 0–1  $\mu\text{m}$  range, the second in the 1–10  $\mu\text{m}$  range, and the third in the 10–50  $\mu\text{m}$  region. Size distributions of the metals had one peak (Pb, Lake Michigan, July-A), two peaks (Cu, Chicago, July-B), or three peaks (Zn, Chicago, July-A; Cu, Lake Michigan, July-B).

This research also showed that the highest concentration of anthropogenic metals is not always in the fine particle

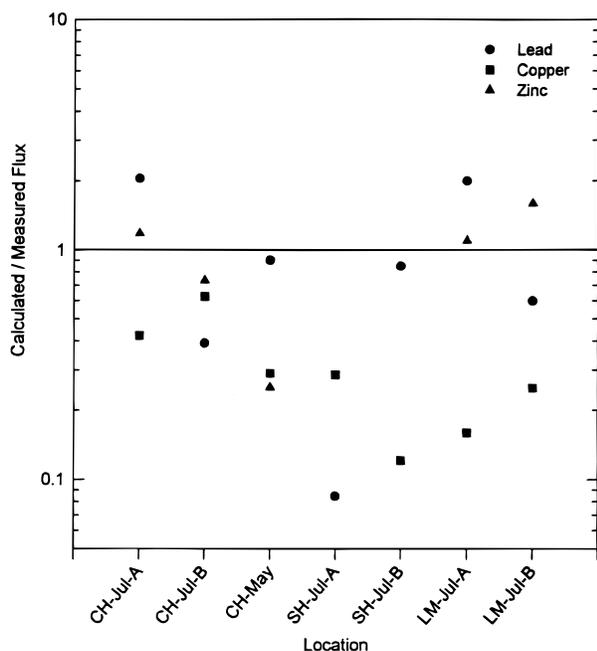


FIGURE 6. Comparison between the flux measured with a dry deposition plate and the flux calculated by the multistep method for lead, copper, and zinc at Chicago (CH), South Haven (SH), and Lake Michigan (LM).

fraction. There were instances when the highest concentration was in the coarse fraction (e.g., Chicago May samples), making the distribution appear to be similar to that of a crustal element.

**Dry Deposition Model.** One of the objectives of this research was to assess the performance of a multistep dry deposition model for anthropogenic metals. This model uses deposition velocities for various particle sizes in conjunction with measured atmospheric concentrations and particle ASDs to predict dry deposition fluxes. Multistep dry deposition models (13) divide the ASD into a number of intervals, assign a deposition velocity to each interval, and sum the calculated flux for each interval to obtain the modeled flux. The flux is given by

$$F = \sum_{i=1}^i C_i V(D_i)$$

where  $F$  is the total flux,  $C_i$  is the concentration for the  $i$ th size interval, and  $V(D_i)$  is the dry deposition velocity for the  $i$ th size interval.

Sehmel and Hodgson's (14) dry deposition velocity model was used to calculate  $V(D_i)$ . This model combines the effects of eddy diffusion and particle inertia on particle motion by an "effective" eddy diffusion coefficient directed toward the surface. The model combined this term with Brownian diffusion and the terminal settling velocity to predict particle deposition velocities. In this study, a plate roughness of 0.001 cm is used. A particle density of 1 g/cm<sup>3</sup> was used because the sampling equipment was calibrated with unit density

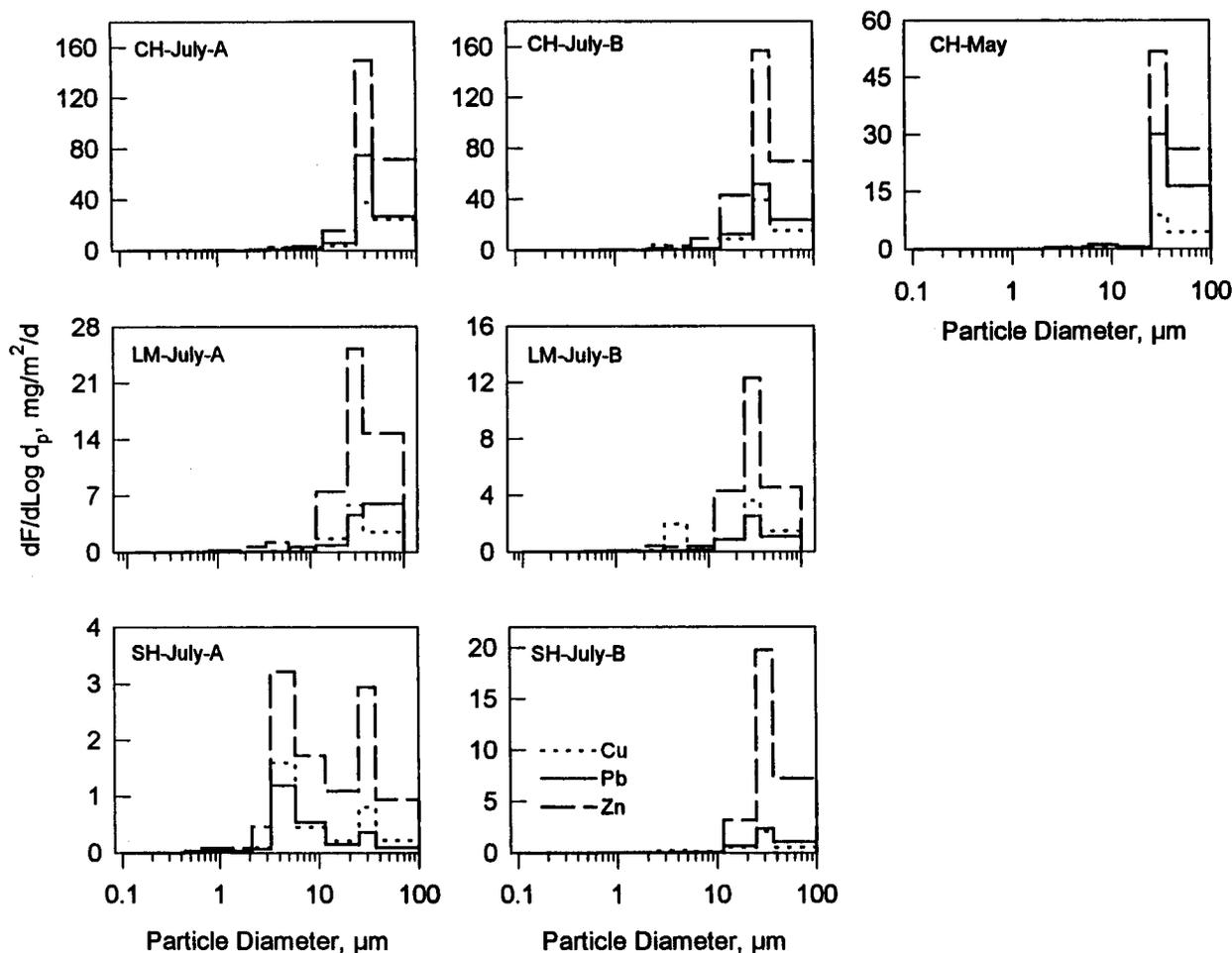


FIGURE 7. Flux size distributions of Pb, Cu, and Zn calculated from the multistep model for Chicago (CH), Lake Michigan (LM), and South Haven (SH).

spherical particles so that all particles are sized aerodynamically equivalent to the reference particles. The model also requires wind speed and ambient temperature as inputs.

Figure 6 compares the measured and modeled flux for all the metals. (Since meteorological conditions were not available for the Lake Michigan sampling location for May 1995, a comparison of the modeled and measured flux could not be made for these samples.) The results suggest that performance of this model for predicting the dry deposition of anthropogenic metals is variable, but in general the results agreed within a factor of 5. For Pb, the ratio of the calculated and measured flux (perfect agreement = 1) varied from 0.1 to 2, while for Cu the ratio varied from 0.1 to 0.8. Zn varied from 0.1 to 1.1. The models generally underpredicted the flux of metals in Chicago, which agrees with the findings of Holsen and co-workers (4). A cause for the underestimation may be that the dry deposition velocity for particles in the 5–80  $\mu\text{m}$  size range tends to be underestimated by this model (28).

Figure 7 presents the relationship between flux and particle diameter for the three metals. The cumulative flux was obtained from the multistep model. As expected and also reported by others (4, 28), in all cases the bulk of the flux (>90%) was due to particles > 2.5  $\mu\text{m}$ , although in most cases anthropogenic metals were primarily in the fine aerosol (<2.5  $\mu\text{m}$ ) fraction.

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