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Dry Deposition Fluxes and Atmospheric Size Distributions of Mass, Al, and Mg Measured in Southern Lake Michigan during AEOLOS

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ABSTRACT. In this study, which was a part of the Atmospheric Exchange Over Lakes and Oceans Study (AEOLOS) investigation, the dry deposition fluxes and atmospheric size distributions (ASDs) of mass and crustal metals (aluminum and magnesium) were measured over the southern basin of Lake Michigan (in Chicago, over Lake Michigan, and in South Haven, Michigan). Airborne crustal metals arise primarily from fugitive dust emissions and are associated with the coarse fraction of atmospheric aerosol. Consequently, they can serve as fingerprints for the atmospheric behavior of fugitive dust. The flux of these metals were substantially higher in Chicago than in either South Haven or over Lake Michigan. The measured average mass, aluminum, and magnesium fluxes were 138, 2.23, and 5.32 mg/m^2-day in Chicago, 47.8, 0.24, and 0.28 mg/m^2-day over Lake Michigan, and 37.4, 0.17, and 0.12 mg/m^2-day in South Haven, respectively. The ASDs of crustal metals measured in Chicago had higher concentrations of coarse particles than ASDs measured over Lake Michigan and in South Haven. The calculated flux of metals using a multistep model and dry deposition velocities obtained from the Sehmel-Hodgson model were in general agreement with measured fluxes of crustal metals. Particles >10 μm were found to be responsible for the majority of the flux. AEROSOL SCIENCE AND TECHNOLOGY 29:281–293 (1998) **© 1998 American Association for Aerosol Research**

INTRODUCTION

This research is a part of the AEOLOS whose broad goal is to assess the effect that emissions of air pollutants into the coastal urban atmosphere have on the atmospheric depositional fluxes to adjacent Great Waters (the Great Lakes, the Chesapeake Bay, Lake Champlain, and other coastal waters). Urban and industrial activities in areas near the Great Waters result in elevated concentrations of particles and chemicals that can be transported over the adjacent Great Waters and removed by dry and wet deposition

One of the important emission sources in urban areas is fugitive dust, which can arise from a wide variety of sources including paved and unpaved roads, industrial areas, and construction and agricultural activities. Fugitive dust is usually associated with the coarse fraction (>2.5 μm) of atmospheric aerosols. Since crustal metals (e.g., Si, Al, Ca, Fe, Mg) in the atmosphere are derived

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mainly from fugitive dust emissions, these metals can serve as surrogates or fingerprints for this source. Thus, an understanding of the behavior and properties of suspended/wind blown dust can be obtained by monitoring crustal metals. For example, a high dry deposition flux of aluminum or silica at a particular site would signal a high contribution from fugitive dust emissions. Receptor modeling studies (Chow et al., 1992) have shown that PM_{10} source contribution from road dust can be as high as 31% and that the coarse fraction of many anthropogenic metals is associated with road dust (Chow, 1995; Sweet and Vermette, 1993).

This paper presents the results of the investigation of the dry deposition and atmospheric size distribution (ASD) of mass and crustal metals (aluminum and magnesium) in the southern basin of Lake Michigan. An objective of this work was to compare the dry depositional flux and ASD of these crustal metals in Chicago (urban site), over Lake Michigan onboard the R/V Lake Guardian in the vicinity of Chicago, and in South Haven, Michigan (nonurban site).

This study includes the measurement of ASDs across a wide particle size range (0.1–100 μm) using the Noll Rotary and Cascade Impactors, during various seasons at the three locations listed above. Since the majority of the airborne mass and crustal metals are derived from dust emissions, an assessment of the change in their concentration during transport over Lake Michigan would indicate the amount that windblown dust is contributing to dry deposition to the lake. This research also included an investigation of the relationship between flux and particle diameter to determine which particle size range dominates the dry deposition flux of crustal metals.

EXPERIMENTAL SECTION

Description of Sampling Locations, Sampling Periods, and Meteorological Conditions

Samples were taken at three locations (Chicago, Lake Michigan, and South Haven) in May 1994, July 1994, and January 1995 (Fig. 1). Each sample set was taken over a period of one week and included dry deposition plate, Noll Rotary, and the Cascade Impactor samples. Noll Rotary Impactor (NRI) samples were obtained approximately every two days for between 3 and 23 hours to avoid overloading. As a result, each instrument may have a different percent exposure time, but all three instruments cover the same sampling period.

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, 5.6 km south of Chicago's center and 1.6 km west of Lake Michigan. The institute's 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_{10} (one in six days) by the Illinois EPA. The annual arithmetic mean PM_{10} value for 1993 at this site was 33 $\mu g/m^3$, which is the median value of the five sites (47, 34, 33, 31, and 30 $\mu g/m^3$). It is classified by Illinois EPA as being primarily influenced by area sources in a primarily neighborhood environment (Franek, 1996). The location and the height of the sampling instruments on the platform and meteorological tower on the roof were out of the roof wake boundary calculated using techniques in Wilson (1979) and Hanna et al. (1982).

Lake Michigan. Samples were taken aboard the U.S. EPA R/V 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 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 sur-

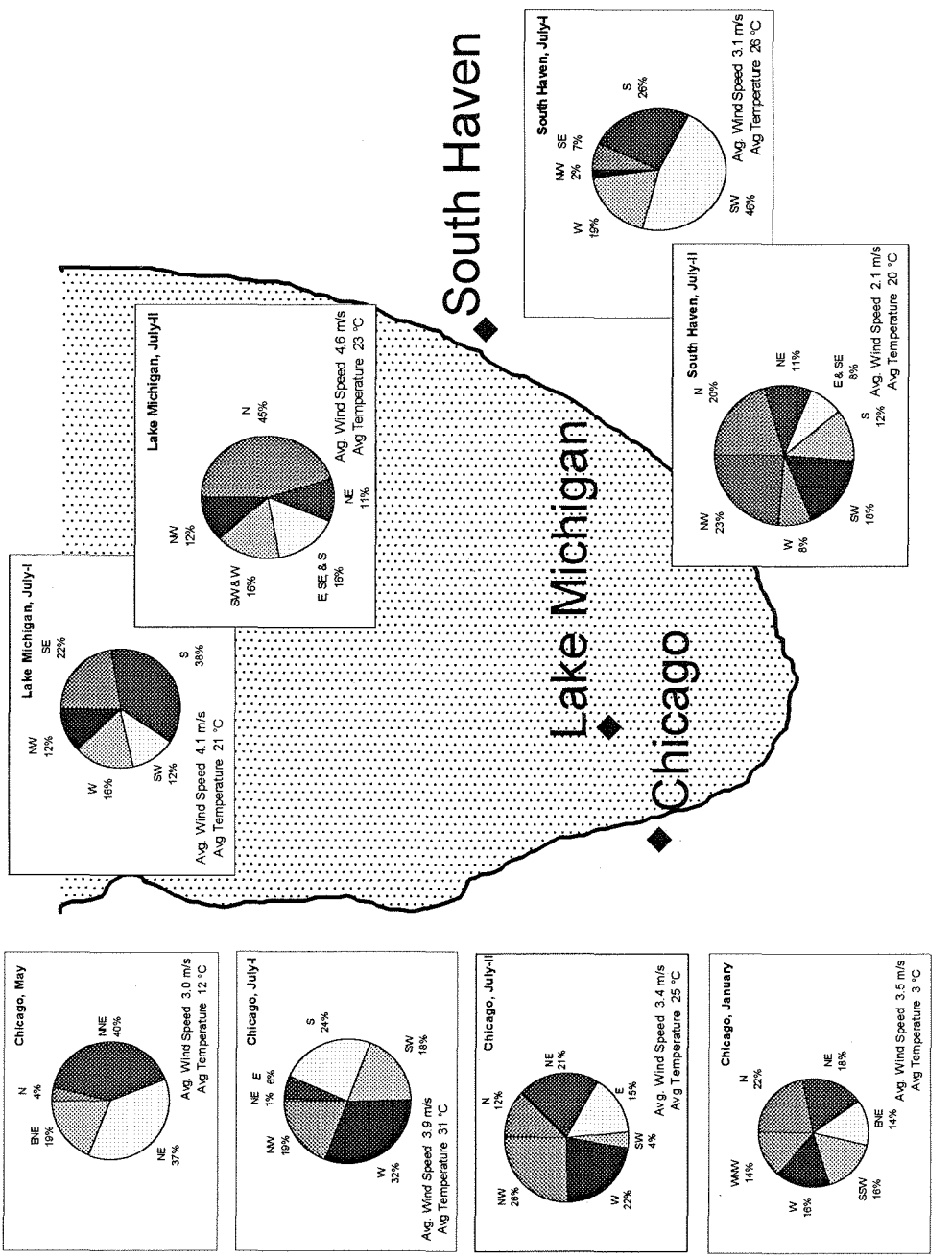

FIGURE 1. Map of Lake Michigan showing the three sampling sites and meteorological conditions during sampling.

rounded by short grass and the farm is surrounded by rolling hills and orchards.

Sampling Periods and Meteorological Conditions. Detailed information on the sampling periods and the meteorological conditions prevailing during the sampling periods are found elsewhere (Sofuoglu, 1995) and summarized in Fig. 1. Wind direction data shows that the May sample was collected when the wind was from over the lake, while the July-I sample was collected when the wind was from primarily from over the land (toward the lake).

Measurement of Dry Deposition Flux and Ambient Particle Concentrations

Dry Deposition Flux. The dry deposition flux was measured at the three locations using a smooth surrogate surface with a sharp leading edge (dry deposition plate), mounted on a wind vane (Noll et al., 1988). The plate used in this study was similar to those used in wind tunnel studies (McCready, 1986). It was made of polyvinyl chloride and was 21.5 cm long , 7.6 cm wide, and 0.65 cm thick with a sharp leading edge ($<10^{\circ}$) 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 x 2.5 cm) coated with Apezion L grease (thickness $\sim$ 5 μm) to collect impacted particles (123 cm^2 total exposed surface)(Cahill, 1979). The strips were weighed before and after exposure to determine the total mass of particles collected. The strips were then extracted and analyzed for Al and Mg using the protocol described below.

Ambient Particle Concentrations. Atmospheric particles were sampled with a Noll Rotary Impactor (Noll et al., 1985, Noll and Fang, 1986), and an Andersen I ACFM nonviable ambient particle-sizing sampler with a preseparator (Graseby Andersen Instruments Inc., 1985). The NRI is designed to collect the particles conventional samplers exclude. It is a multistage rotary inertial impactor that collects coarse particles by simultaneously rotating four rectangular collectors (stages) of different dimensions through the air. The stages are covered with Mylar strips coated with Apezion L grease to minimize particle bounce. Total collection areas were 1.2 cm^2, 3.1 cm^2, 10.3 cm^2, and 10.3 cm^2 for stages A–D, respectively. The strips were weighed before and after sampling to determine the total mass collected.

Extraction and Analytical Protocol for Metals

Extraction was carried out in the University of Michigan Air Quality Laboratory 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 evaporated subsequently by directing a stream of ultrapure nitrogen on its surface. Next, 20 mL of 10% (v/v) ultrapure nitric acid was added, and the container was placed in a Teflon bomb and loaded into the computer-controlled microwave oven. Acid digestion was performed for 30 minutes at 160°C and approximately 160 psi, following which the Teflon bomb was allowed to cool for a period of one hour. Samples were analyzed subsequently 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 the r^2 (coefficient of determination) 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 difference between certified and measured concentrations was between 70% to 120%. Precision was calculated 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 split and replicate samples.

Method detection limits (MDLs) for the ICP-MS were calculated by repeated analy-

sis of a low concentration sample. MDL is defined as three times the standard deviation of the concentrations obtained in the seven runs. MDLs for Al and Mg were 1.3 and 1.1 ppb, respectively. Metal concentrations in the process blanks were either below or of the same magnitude as the MDLs. To account for trace levels of background contamination, the mass of metal in the field blank was subtracted from the mass collected on the various stages of the cascade impactor and NRI, and the dry deposition plate.

Extraction efficiencies were calculated by measuring metal concentrations after spiking a 10% nitric acid solution with urban particulate matter (NIST SRM 1648). Recovery efficiencies for aluminum and magnesium were 60% and 72%, respectively.

RESULTS AND DISCUSSION

Dry Deposition Flux

Figure 2 shows the dry deposition flux of total mass, aluminum, and magnesium measured at different time periods in Chicago, South Haven, and over Lake Michigan. As reported earlier (Holsen et al., 1993; Noll et al., 1990), there is a significant spatial variation in the fluxes of mass and crustal metals. The fluxes were substantially higher in Chicago than either in South Haven or over Lake Michigan. The July-I flux was uncharacteristically low possibly because the weather was fairly stormy during the first half of the July-I sampling period, and the deposition plates were exposed for only a short period of time. The particle concentrations during this period were approximately two times higher for the coarse fraction and about six times higher for the fine fraction than during the second half of the sampling period in which the plates were mostly exposed.

The measured average mass, aluminum, and magnesium fluxes in Chicago were 138, 2.23, and 5.32 mg/m^2-day, over Lake Michigan were 47.8, 0.24, and 0.28 mg/m^2-day; and in South Haven were 37.4, 0.17, and 0.12 mg/m^2-day, respectively. These fluxes compare well with results of previous studies. Holsen et al. (1993) reported aluminum fluxes of 2, 0.8, and 0.5 mg/m^2-day and magnesium fluxes of 3, 1.5, and 0.5 mg/m^2-day in Chicago, over Lake Michigan, and in South Haven, respectively. Dulac et al. (1989) reported aluminum flux of 0.87 mg/m^2-day over the western Mediterranean sea. Although spatial flux trends for anthropogenic metals were similar, the magnitude of flux was an order of magnitude lower than for these crustal metals (Paode et al., 1998).

The crustal elements' flux over Lake Michigan is due primarily to fugitive emissions from the Chicago area and other areas around southern Lake Michigan. In this study the ratio of the Chicago to Lake Michigan flux was almost four times for mass and 11 (Al) to 26 (Mg) times for crustal metals. This finding implies that fugitive dust emissions from the Chicago region are partly removed before they reach the sampling station on the lake. The ratio of the Chicago to Lake Michigan flux for crustal metals was comparable to the same ratios for anthropogenic metals. While Pb and Zn had values similar to magnesium (23 and 25 respectively) the ratio for Cu (15) was similar to aluminum (Paode et al., 1998).

Contrary to expectations, fluxes for crustal metals were not lower in January, probably because the temperature was well above freezing (avg. 3°C) and the ground conditions were relatively dry. Noll et al. (1985) have reported 30% lower fluxes when the ground was wet and 90% lower fluxes when the ground was frozen.

The depositional profile of metals was influenced by wind direction. In Chicago and over Lake Michigan, lower fluxes were measured during May when the wind was primarily from the lake (Fig. 1). As a consequence the dry deposition in May was impacted to a lesser degree by the wind blown dust emissions from the Chicago area. Similar trends were seen in our research on anthropogenic metals (Paode et al., 1998).

Atmospheric Size Distributions

Figures 3, 4, and 5 present the atmospheric size distributions (0.1 to 100 μm) of mass,

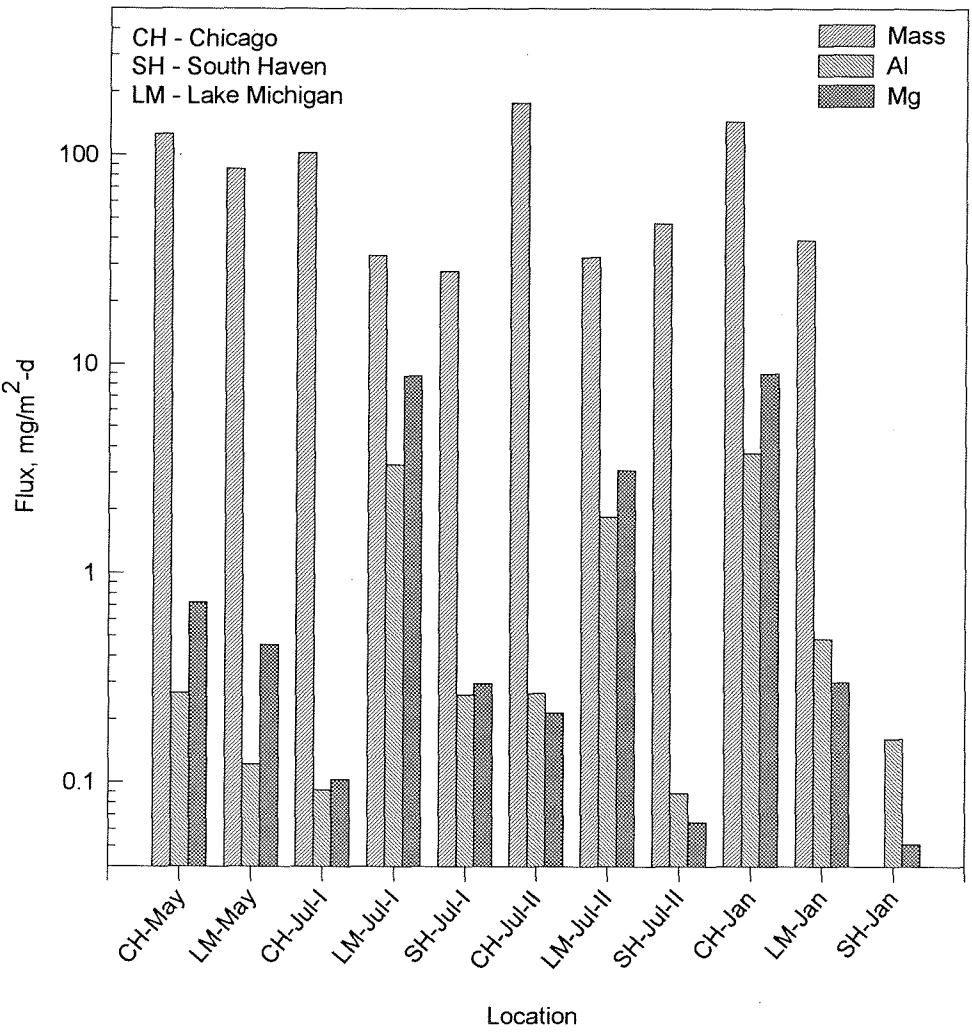

FIGURE 2. Dry deposition flux of mass, aluminum, and magnesium measured with the dry deposition plate in Chicago, IL, in South Haven, MI, and over Lake Michigan. May and July samples were taken in 1994, and January sample was taken in 1995.

aluminum, and magnesium, respectively, during various sampling periods at the three locations. The figures were constructed to permit the comparison of ASDs for a single site at various time periods (horizontal panels), and for a specific time period at various sites (vertical panels).

Figures 3, 4 and 5 show the atmospheric size distributions measured simultaneously in Chicago, on the Lake Guardian, and in South Haven during May and July of 1994. The ASDs measured over the Lake and South Haven have far lower concentrations in the coarse fraction than the Chicago samples. In particular, the Lake Michigan and South Haven ASDs reflect a virtual absence of particles in the >20 μm size fraction. These particles have large deposition velocities and are probably deposited to the lake before they reached the ship. In the study of

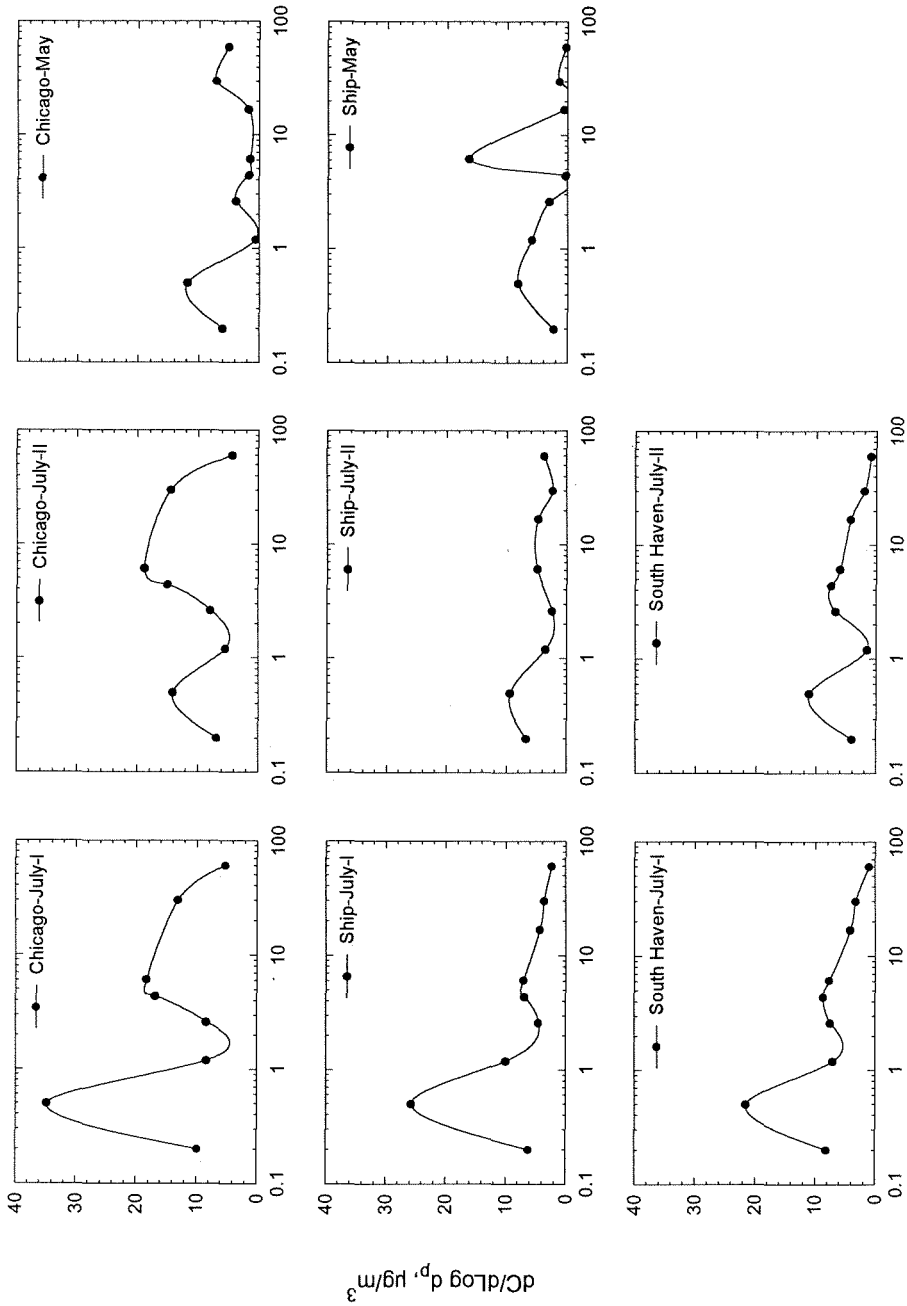

FIGURE 3. Atmospheric size distributions of mass measured with a Cascade Impactor and Noll Rotary Impactor in Chicago, IL, in South Haven, MI, and over Lake Michigan.

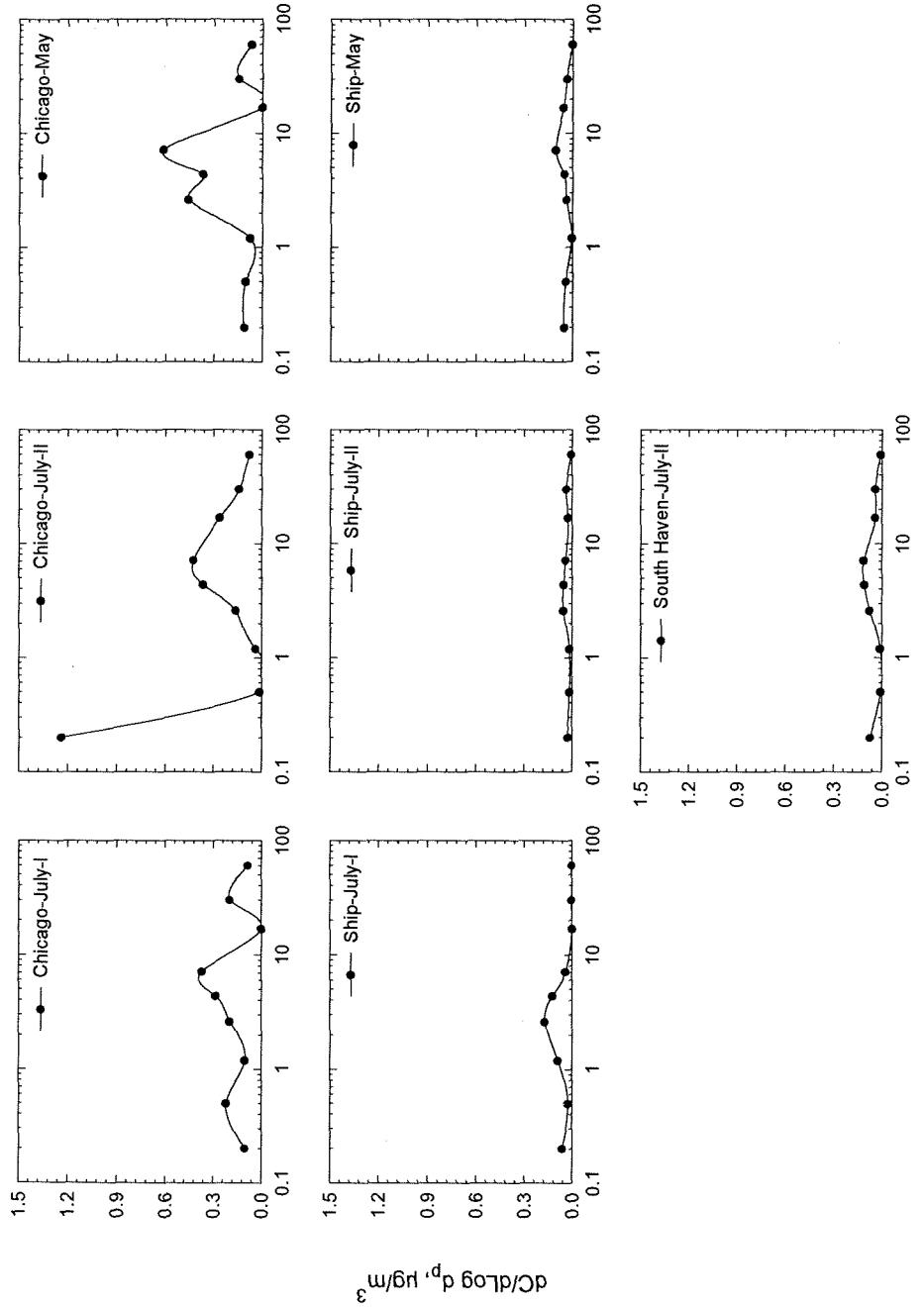

FIGURE 4. Atmospheric size distributions of aluminum measured with a Cascade Impactor and Noll Rotary Impactor in Chicago, IL, in South Haven, MI, and over Lake Michigan.

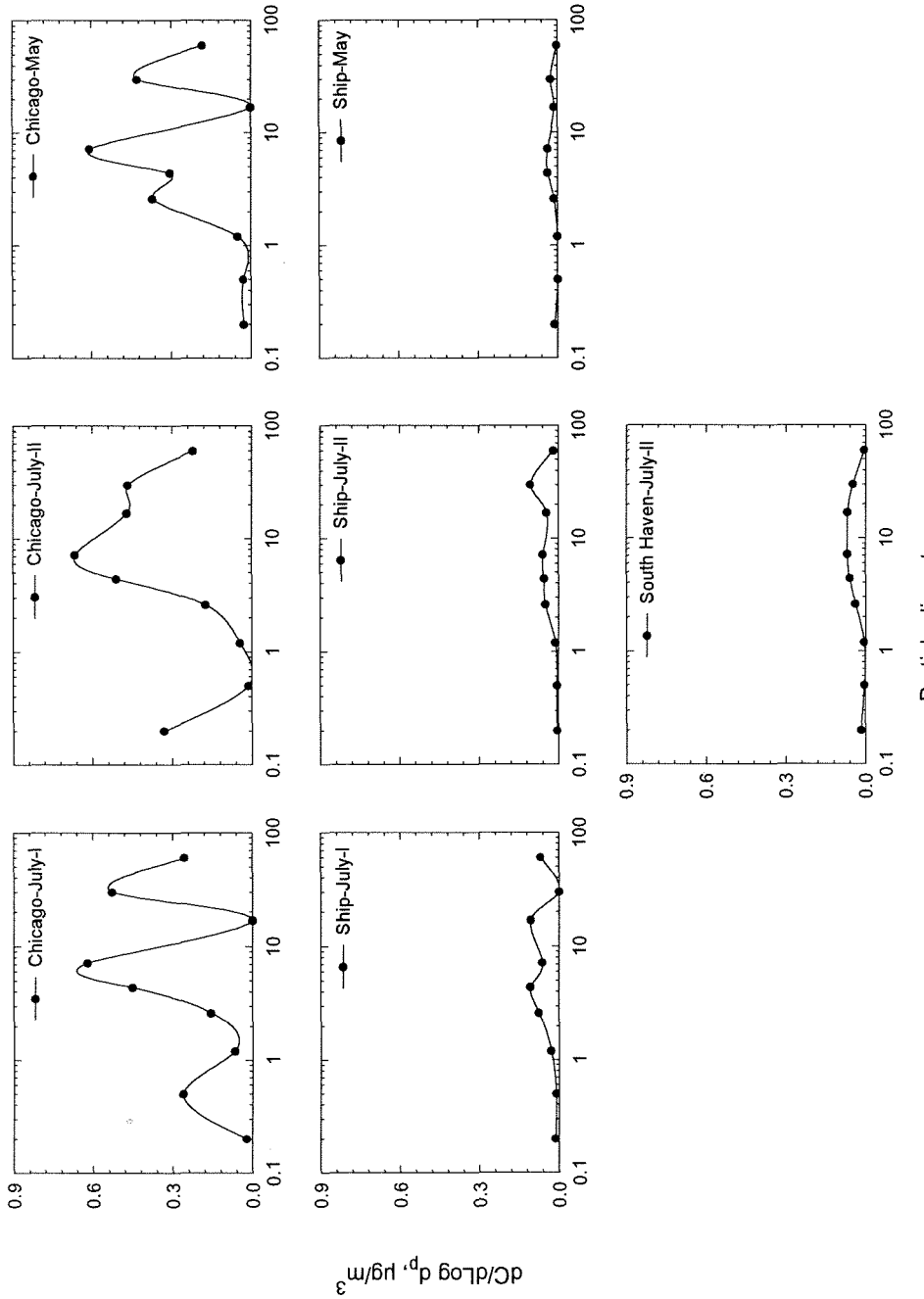

FIGURE 5. Atmospheric size distributions of magnesium measured with a Cascade Impactor and Noll Rotary Impactor in Chicago, IL, in South Haven, MI, and over Lake Michigan.

anthropogenic metals (Paode et al., 1998) Cu, Pb, and Zn also had decreasing concentrations from Chicago to South Haven, especially in the coarse fraction.

Previous studies have suggested a unimodal distribution (one peak) for all crustal metals in the coarse fraction, with a peak occurring in the 8–16 μm (Noll et al., 1990), or the 5–10 μm (Milford and Davidson, 1985) particle size range. In this study, there were examples of the existence of two possible peaks; the first in the 7–10 μm range, the second in the 20–50 μm region. Size distributions of crustal metals had one peak (magnesium, South Haven, July-II), or two peaks (aluminum, Chicago, July-I) in the coarse fraction. There were also instances where a peak was observed in the fine region (aluminum and magnesium, Chicago, July-I), suggesting there are anthropogenic sources of these metals in Chicago area.

Dry Deposition Model

The performance of a multistep dry deposition model in combination with the Sehmel and Hodgson (1978) dry deposition velocity model was evaluated for mass and the crustal metals. The multistep model uses deposition velocities for various particle sizes in conjunction with measured atmospheric concentrations and particle ASDs to predict the dry deposition fluxes. The multistep dry deposition model (Holsen and Noll, 1992) divides the ASD into a number of intervals, assigns a deposition velocity to each interval, and sums 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 ith size interval, and $V(D_i)$ is the dry deposition velocity for the ith size interval. Sehmel and Hodgson's (1978) 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^3 was used because sampling equipment used in study was calibrated with unit density spherical particles so that all particles are collected and 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 fluxes for mass and the two metals. Since meteorological conditions were not available for the Lake Michigan sampling location for May, a comparison could not be made for these samples. The results suggest that performance of this model for predicting the dry deposition of crustal metals is variable but the results agreed within a factor of four. For aluminum and magnesium the ratio of estimated and measured flux varied from 0.3 to 4. The largest discrepancies between measured and modeled fluxes had ratios of 4.8 (Chicago-July-I) for Mg, and 4.4 (Chicago-July-I) for Al in July-I sample. During the first week of sampling in the July-I period there was consistent rain and exposure times of the dry deposition samples were short. In general, measured mass flux was in good agreement with the modeled flux. The model generally underpredicted the flux in Chicago, which agrees with the findings of Holsen et al. (1993) and Paode et al. (1998). A cause for the underestimation may be that the dry deposition velocity for particles in the 5–80 μm size range tends to be underestimated by this model (Lin et al., 1994).

Relationship Flux and Particle Diameter

Figure 7 presents the relationship between flux and particle diameter for the metals. The cumulative flux was obtained from the multistep model. As previously reported (Holsen et al., 1993; Lin et al., 1994; Holsen and Noll, 1992), the coarse fraction accounts for a bulk of the flux (>90). Taking 90% of the flux as a reference point, the particle size

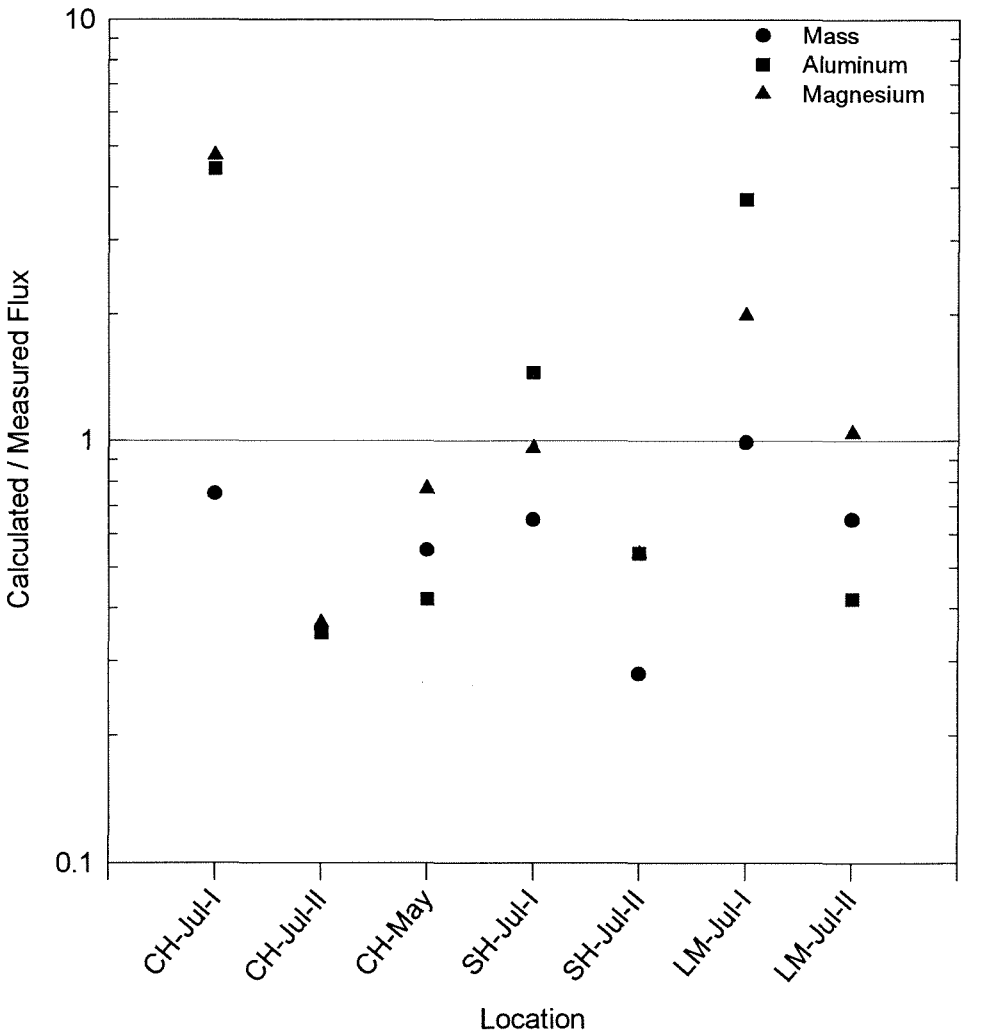

FIGURE 6. A comparison between the flux measured with the dry deposition plate and the flux calculated by the multistep method for mass, aluminum, and magnesium in Chicago (CH), S. Haven (SH), and over Lake Michigan (LM).

fraction accounting for this proportion of the flux ranged from >3 to >30 μm. For anthropogenic metals this range was >2 to >20 μm, although in most cases anthropogenic metals were primarily in the fine (< 2.5 μm) fraction (Paode et al., 1998).

CONCLUSIONS

There was substantial spatial variation in the dry deposition fluxes of crustal metals. Fluxes measured in Chicago were substantially higher than the fluxes measured over Lake Michigan or in South Haven. In this study the atmospheric size distributions of the coarse fraction (>2.5 μm) of crustal metals had one or two peaks. The calculated fluxes using the multistep model and dry deposition velocities obtained from the Sehmel-Hodgson model tended to underpredict the dry deposition flux. The flux and ASD profiles indicate that when the wind is

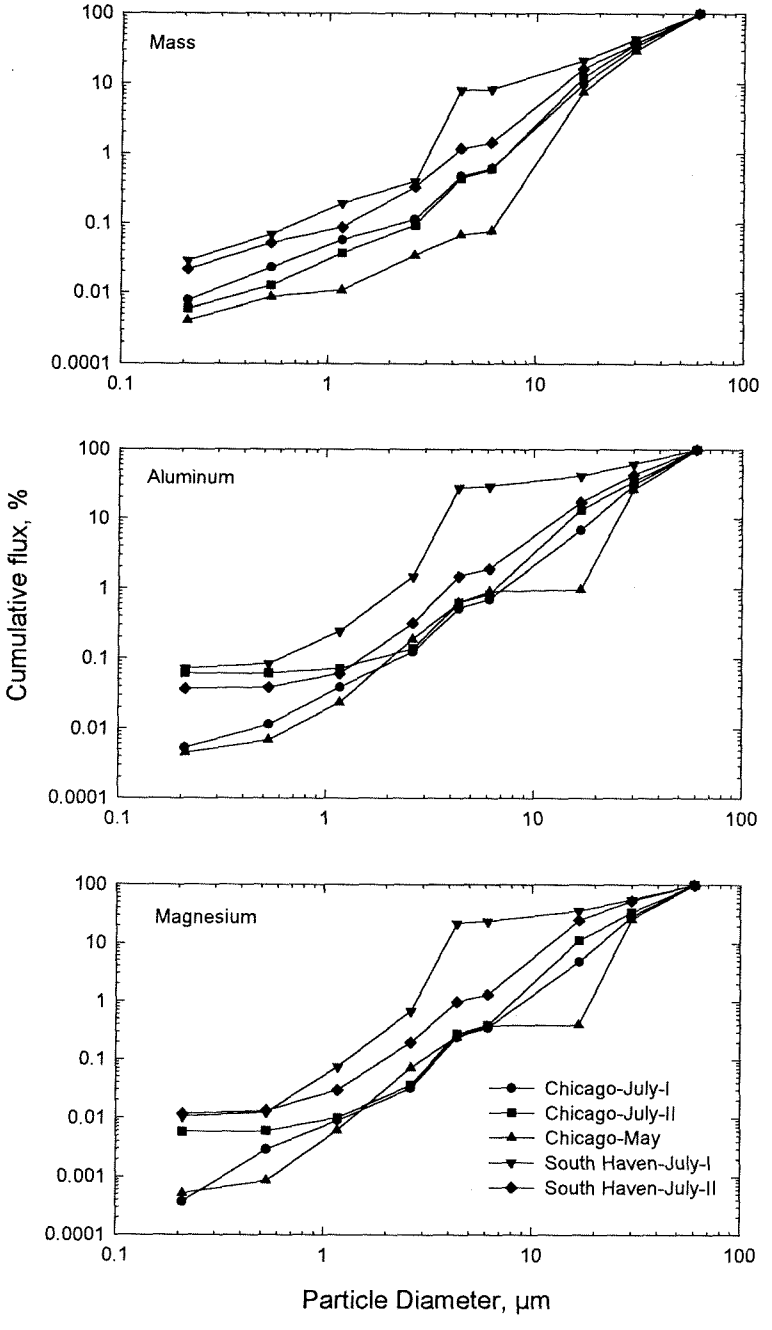

FIGURE 7. Cumulative flux data calculated from the multistep model for mass, aluminum, and magnesium at various locations.

from the land over the lake the fugitive dust emissions are partially removed before they reach the sampling site over the lake.

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