Supporting Information

Dry deposition fluxes and deposition velocities of seven trace metal species at five sites in central Taiwan - A summary of surrogate surface measurements and a comparison with model estimation

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1. Measurement instruments

Polyurethane Foam Sampler

The PS-1 (GPS1 Polyurethane Foam (PUF) Sampler, General Metal Work) collects total suspended particulate mater (TSP). The PS-1 sampler is a complete air sampling system designed to simultaneously collect suspended airborne particles. The maximum collection particle size is appropriately 100 μm (Graseby-Andersen, GMW High Volume Air Sampler, Ohio, USA). The flow rate was adjusted to 200 L/min in this study. A quartz filter (diameter 102 mm) with pore size of 3 μm was used to filter the suspended particles in the study. The filters were first conditioned for 24 hours under an electric chamber at humidity of $35 \pm 5\%$ and temperature of $25 \pm 5 \circ C$, prior to both on and off weighing. Filters were placed in a sealed CD box during transport and storage process. The sampling device and procedures are similar to those described in a previous study by these authors (Fang et al., 2010).

Dry deposition plate

A dry deposition plate (DDP) comprising of a smooth, horizontal, surrogate surface is used in this study. The DDP is expected to provide a lower bound estimate of the dry deposition flux. The DDP consists of a smooth surface plate made of polyvinyl chloride (PVC) that measured 21.5 cm long, 8.0 cm wide, and 0.8 cm thick. The DDP also contains a sharp, leading edge that is pointed into the prevailing wind. All filters were maintained in a condition of $35 \pm 5\%$ relative humidity and a temperature of $25 \pm 5 \circ C$ for more than 24 hours. Prior to sampling processing, all filters were weighed to 0.0001-g significant digits (Fang et al., 2007a, b).

2. Chemical analysis

The samples were placed in an oven one night before being weighed. A quarter of the filter was cut and selected before the digestion process. The filters were cut into pieces thin added into the Teflon cup. 3 ml of hydrochloric acid (HCl) and 9 ml of nitrate (HNO_3) were mixed and then added to this cup. After that the samples were heated at 50^0C on the hot plate for

two hours. Samples after digestion on the hot plate were then filtered. After filtration, the sample solution was then added 0.2 % of HNO_3 and added up to 100 ml solution. Before ICP-MS (Perkin Elmer Sciex ELAN DRCII) analysis of arsenic species and inductively coupled plasma - atomic emission spectrometer (ICP-AES) using a Perkin Elmer N0800540 Plasma Emission Spectrometer was used to analyze the metallic elements (Mn, Fe, Zn, Cr, Cu and Pb), these samples were kept at 4^0C in the refrigerator.

3. Data quality control and availability

The blank test background contamination was monitored by using operational blanks (unexposed projection film and quartz filter) which were processed simultaneously with field samples. The field blanks were exposed in the field when the field sampling box was opened to remove and replace field samples. Background contamination of arsenic was accounted for by subtracting field blank values from the concentrations. Field blank values were very low, usually below or around the method detection limits. In this study, the background contamination is insignificant and can be ignored. The results of the blank test are 0.18, 0.08, 0.12, 0.10, 0.30, 0.35, 0.32, 0.20, 0.19 and 0.22 μg for As, As(III), As(V), Hg(p), Mn, Fe, Zn, Cr, Cu and Pb.

4. Meteorological data

Daily average meteorological data were obtained from the nearby meteorological observation station. The seasonal and annual average wind speed and temperature are listed in Table S1. Note that the height the wind data were collected are different from site to site (12, 7, 5, 12 and 7 meters for Sites 1 to V, respectively).

5. Dry deposition model

Dry deposition velocity (V_d) was calculated using the size-segregated particle dry deposition model described in Zhang et al. (2001):

$$V_d = V_g + \frac{1}{(R_a + R_s)}$$

where V_g is the gravitational settling velocity, R_a is the acronymic resistance, and R_s is the surface resistance. R_s is parameterized as a function of collection efficiencies from Brownian diffusion, impaction and interception processes.

The original model of Zhang et al. (2001) had only 15 land use categories (LUC) and was later extend to 26 LUC to be consistent with Zhang et al. (2003). Input parameters in Zhang et al. (2001) were given for each LUC and for five seasonal categories. This approach was discarded here; instead, the same approach developed in Zhang et al. (2003) was used. That is, for any input parameter X changing with season, a maximum (X_{max}) and a minimum value (X_{min}) were provided and were then interpolated to any day of the year based on the annual variation of leaf area index (*LAI*):

$$X(t) = X(\min) + \frac{LAI(t) - LAI(\min)}{LAI(\max) - LAI(\min)} [X(\max) - X(\min)]$$

Where t represents any day of the year, and *LAI*(min) and *LAI*(max) represents minimum and maximum LAI values, respectively, during the year. Input parameters for the particle dry deposition model that need interpolation include a parameter for the characteristic radius of collectors, a parameter for calculating collection efficiency by Brownian diffusion, and a parameter for calculating collection efficiency by impaction (Zhang et al., 2001). Roughness for each LUC for the particle dry deposition model is the same as for the gaseous dry deposition model as described in Zhang et al. (2003).

6. Supplement figures

Four figures are provided in this document as supplemental materials. Figure S1 shows the seasonal average and standard deviation of dry deposition fluxes (ng m^{-2} min^{-1}) of As, Mn, Fe, Zn, Cr, Cu and Pb measured at the five sites. Figure S2 shows the seasonal average and

standard deviation of deposition velocities (cm s^{-1}) calculated as the ratio of measured fluxes

and concentrations of As, Mn, Fe, Zn, Cr, Cu and Pb at the five sites. Figure S3 shows the

correlation between measured daily flux and concentration for As, Fe, Zn, Cr, Cu and Pb at

the five sites. Regression equation, square of correlation coefficient (R^2), and the significance

of the correlation (P - the probability that the two group variables are not correlated) are

shown in each subset of the Figure. Any P larger than 0.1 represents insignificant correlation.

Figure S4 shows the lognormal size distributions for three modes: PM$_{2.5}$, PM$_{2.5-10}$ and PM$_{10+}$.

Brief discussions of these figures were presented in the main text.

Table S1. Seasonal and annual average of meteorological conditions.

Spring	Site I	Site II	Site III	Site IV	Site V
Temp(℃)	20.61	20.77	20.75	20.73	21.04
RH(%)	83.85	74.69	77.86	81.03	79.62
WS(m/sec)	1.74	1.61	2.39	3.17	2.51
Summer	Site I	Site II	Site III	Site IV	Site V
Temp(℃)	27.36	27.79	27.45	27.11	27.63
RH(%)	81.25	74.92	78.82	82.72	79.46
WS(m/sec)	2.52	2.41	2.59	2.76	2.44
Fall	Site I	Site II	Site III	Site IV	Site V
Temp(℃)	27.88	28.98	28.76	28.53	27.86
RH(%)	70.69	70.75	72.83	74.90	75.68
WS(m/sec)	1.92	1.71	2.09	2.48	1.99
Winter	Site I	Site II	Site III	Site IV	Site V
Temp(℃)	20.05	21.13	21.17	21.21	21.18
RH(%)	72.37	68.66	71.59	74.53	74.14
WS(m/sec)	1.40	1.30	1.72	2.14	1.76
Annual	Site I	Site II	Site III	Site IV	Site V
Temp(℃)	23.97	24.67	24.53	24.39	24.43
RH(%)	77.04	72.26	75.28	78.30	77.22
WS(m/sec)	1.89	1.76	2.20	2.64	2.18

Figure S1. Seasonal average ($\pm$ standard deviation) dry deposition flux (ng m^{-2} min^{-1}) of As, Mn, Fe, Zn, Cr, Cu and Pb measured at the five sites.
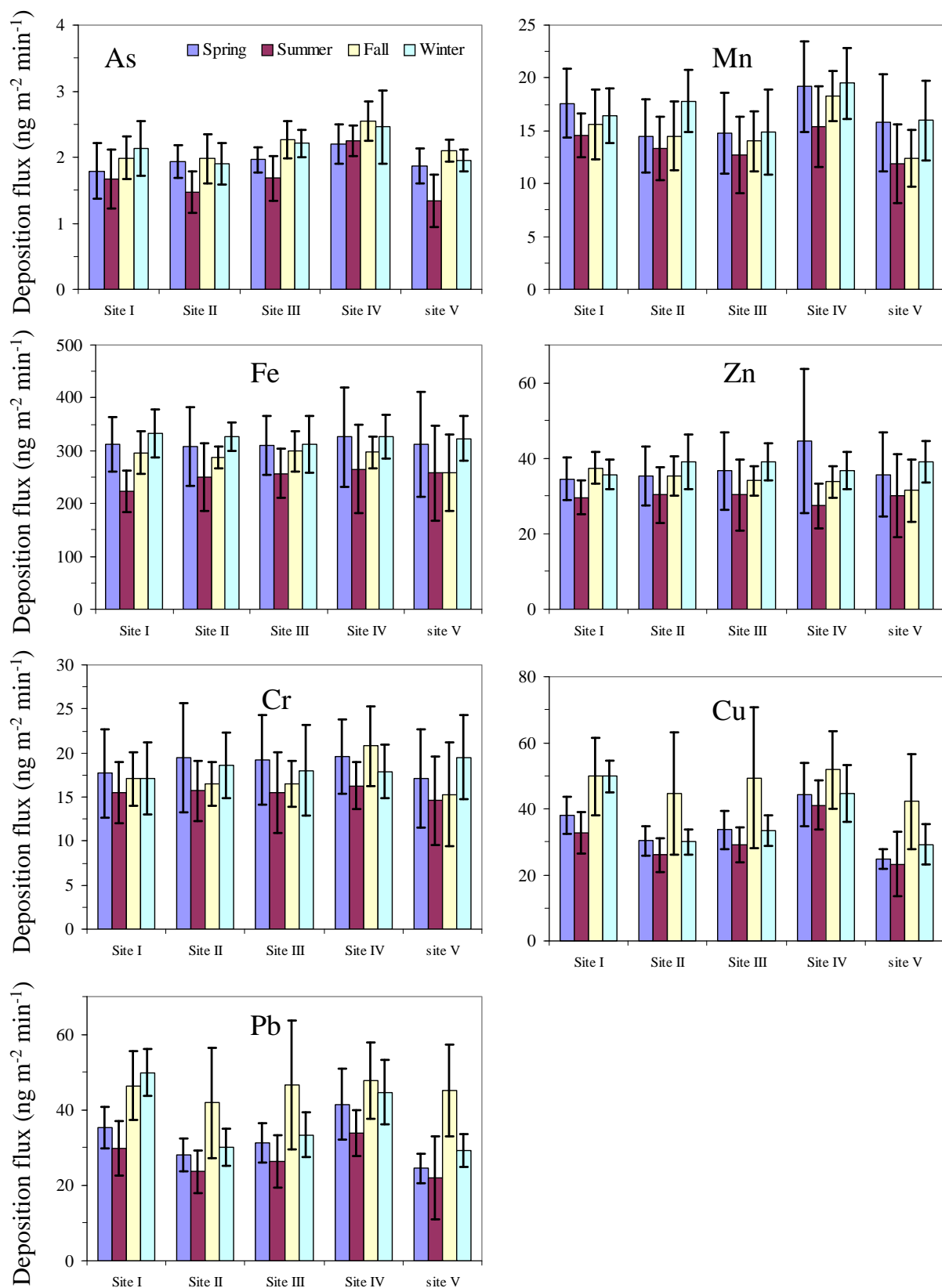
Figure S2. Seasonal average (±standard deviation) deposition velocities (cm s⁻¹) calculated as the ratio of measured fluxes and concentrations of As, Mn, Fe, Zn, Cr, Cu and Pb at the five sites.
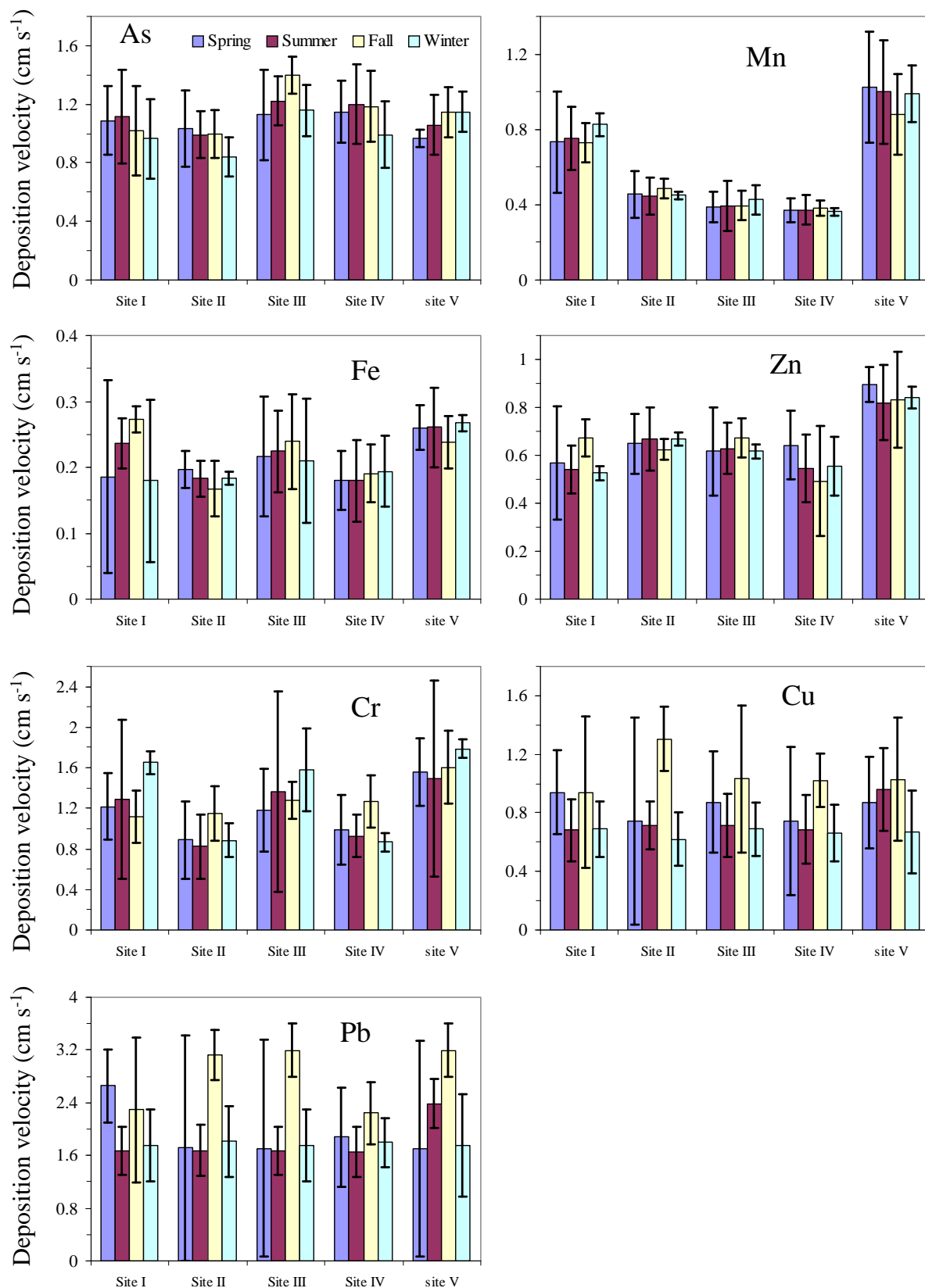
Figure S3. Correlation between measured daily flux (µg m^{-2} day^{-1}) and concentration (ng m^{-3}) for As, Fe, Zn, Cr, Cu and Pb at the five sites
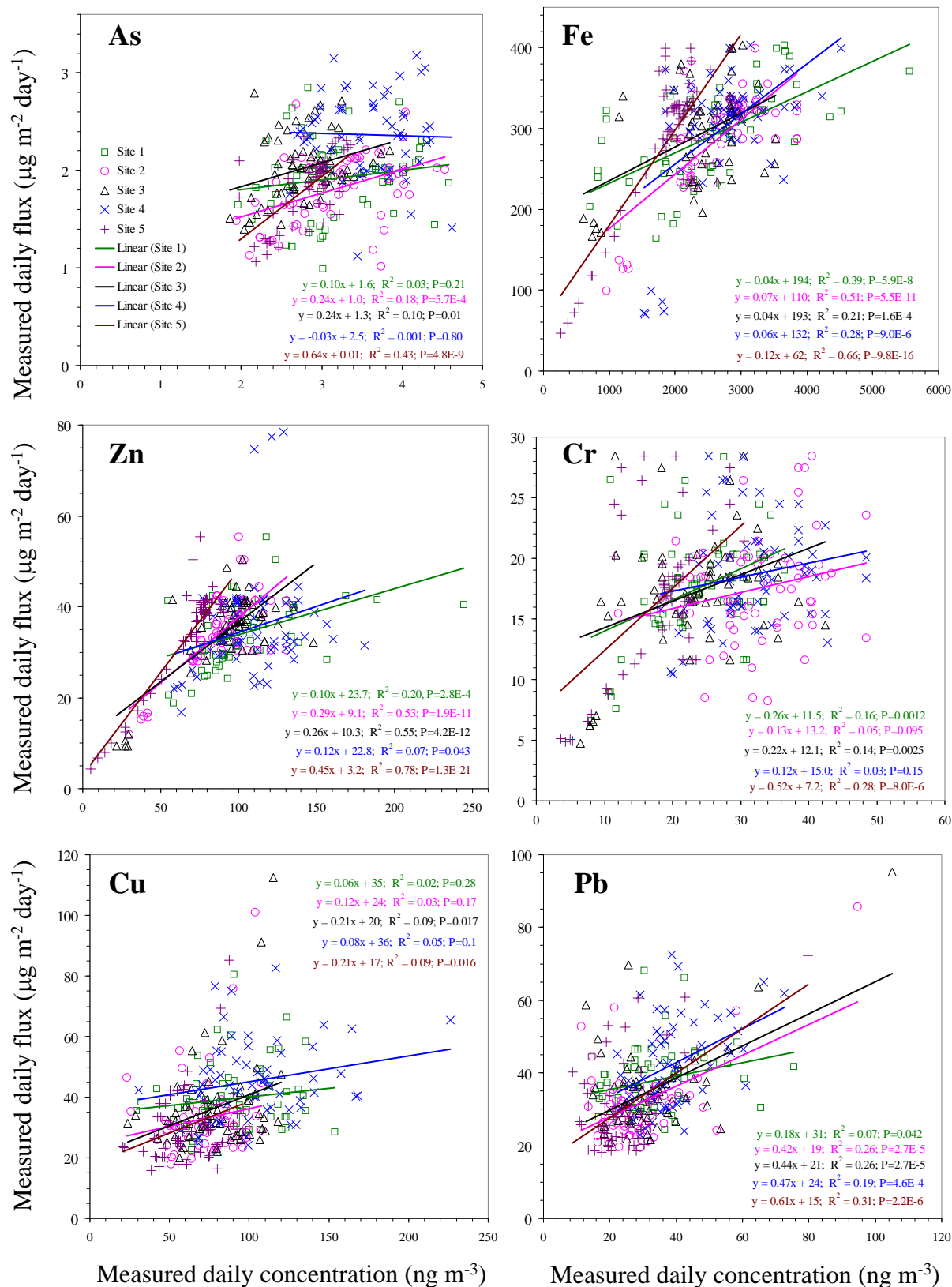
Figure S4. Lognormal size distributions for three modes: $PM_{2.5}$, $PM_{2.5-10}$ and PM_{10+}.