

## A DETERMINATION OF SOURCES OF AEROSOL IN CHICAGO BY CHEMICAL ELEMENT MASS BALANCE

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### Synopsis

*A chemical element mass balance (CEB) receptor model was used to determine the contributions from coal combustion, cement and limestone, mobile sources, refuse incineration, soil, and steel industries to the ambient particulate matter in Chicago.*

*Twenty-four hour average measurements were made of inhalable particulate (IP) matter consisting of fine particles (FP) diameter  $< 2.5 \mu\text{m}$  and coarse particles (CP) diameter  $> 2.5 \mu\text{m}$  but  $< 15 \mu\text{m}$ ; sulfur ( $\text{SO}_4^{--}$ ), and the concentration of Al, Br, Ca, Co, Cr, Cu, Fe, K, Mg, Mn, Na, Ni, Pb, Se, Si, Ti, V and Zn for both size fractions; and  $\text{SO}_2$ , temperature, wind speed and wind direction on 17 days during a 4 month period from May to August 1984. The CEB was applied as an independent analysis to each sampling day.*

*The average FP, CP and IP concentrations were  $21.0 \mu\text{g}/\text{m}^3$ ,  $28.4 \mu\text{g}/\text{m}^3$  and  $49.4 \mu\text{g}/\text{m}^3$  respectively. Based on study period average, coal, limestone, mobile, refuse, soil and steel contributes  $0.40 \mu\text{g}/\text{m}^3$ ,  $7.7 \mu\text{g}/\text{m}^3$ ,  $5.1 \mu\text{g}/\text{m}^3$ ,  $7.8 \mu\text{g}/\text{m}^3$ ,  $5.5 \mu\text{g}/\text{m}^3$  and  $1.2 \mu\text{g}/\text{m}^3$  of the IP respectively. The contribution from the unexplained particulate matter were  $16.3 \mu\text{g}/\text{m}^3$ ,  $5.9 \mu\text{g}/\text{m}^3$  and  $21.6 \mu\text{g}/\text{m}^3$  for the FP, CP and IP respectively. This suggests that the aerosol is concentrated in the small size fractions contributed by the secondary pollutants.*

*On average, the model was able to explain 22.2%, 79.3% and 56.3% of the FP, CP and IP respectively for the Chicago site.*

### Introduction

To design optimal control strategies for atmospheric particles, air pollution authorities need models that can relate observed concentration of the total suspended particulate (TSP) matter or of certain species on particles to the particles sources. The understanding of the chemical and physical characteristics of particulate air pollution sources and the determination of their emission strengths contribute to the development of a rational control strategy. However, the relationship between multiple source emission and air quality is often non-linear and imperfectly understood. Sophisticated dispersion models have been developed that relate air pollution emission to ambient concentration by considering factors such as macro- and micrometeorology, spatial distribution of sources and receptors, emission inventory, topography, chemical reaction and pollutant deposition. But no single model has been proved to be the best for all environment situations.

In contrast to model based on source dispersion, receptor models are derived solely from monitored air pollution and sources chemical characteristic. No a priori assumption about source location and strength, meteorology or topography are required. If one knew the compositions of particles from each type of source, one could resolve the concentration pattern of ambient particles into a set of components contributed by the various types of sources. Such a resolution would indicate the concentration of each source to the concentration of each element as well as to the TSP.

A number of studies have performed quantitative source resolution by applying the chemical element balance (CEB) receptor model developed by Miller et. al.<sup>1</sup> The study was to resolve the chemical composition of airborne particles into components from several types of emission sources. Data on concentrations of several elements were used to determine the abundance of sea salt, soil, automobile emissions and fuel-oil fly ash in total suspended particulate matter at Pasadena, California. Later, Friedlander<sup>2</sup> applied the method of Pasadena, including additional components for cement and tyre dust. Winchester and Nifong<sup>3</sup>, Gatz<sup>4</sup>, and Scheff<sup>5</sup> used element balance methods to estimate source components for the Chicago area. Kowalczyk et. al.<sup>6</sup> resolved aerosol in the Washington D.C. area into six components; soil, marine, coal, oil, refuse incineration, and motor vehicle emissions. By adding sulfate and nitrate to their predicted six components, about 80% of the TSP mass was accounted.

The CEB model is based on the assumption that the concentration of element  $i$  at the receptor location equals to sum of the contributions of  $i$  from  $j$  sources. The model further assumes that mass is conserved, that is the fraction of element  $i$  in the emission of source  $j$  is unchanged from the time it is released to the environment to the time it is collected at the receptor. With all elemental concentrations expressed as fractions of the total mass (same units for sources and receptor), the CEB model can be written as:

$$C_i = \sum_{j=1}^p M_j X_{ij} \quad \dots (1)$$

Where  $C_i$  is the concentration of the chemical component  $i$  measured at the receptor and  $X_{ij}$  is the pollution source elemental concentration or source profile matrix with units of micrograms of  $i$  per gram of total primary particulate matter emitted from source  $j$ . The source contribution  $m_j$  is the ratio of the mass contributed from source  $j$  to the total mass collected at the receptor site. It is this fraction of particulate pollution measured at a receptor which is primary interest in CEB calculation.

If the  $C_i$  and  $X_{ij}$  at the receptor for all  $p$  of the source types suspected of affecting the receptor are known, and  $p < n$  ( $n$  = number of chemical species), a set of  $n$  simultaneous equations exist from which the source type contribution  $m_j$  may be calculated by weighted least square (WLS) methods.

Similarly, the purpose of this study is to illustrate the application of the CEB model in predicting the source of particulate matter affecting the Illinois Institute of Technology in Chicago. The elemental composition of particulate matter collected in two size ranges were analyzed using x-ray fluorescence spectrometer. Six source categories were studied including soil-derived aerosol, mobile sources, refuse incineration, cement and limestone, coal combustion, and iron and steel production.

### Methodology

Ambient 24-h average particulate samples were collected every sixth day from May — August 1984. Air sampling was carried out on the roof top of a building (13 m height) nearby the students dormitories at Illinois Institute of Technology located 2 miles south of downtown Chicago. The fine particulate, FP (0 — 2.5 microns) and coarse particulate, CP (2.5 — 10 microns) fractions of the inhalable particulate matter (IP = FP + CP) were collected using a Sierra Andersen dichotomous sampler (Model 244) operating at  $1 \text{ m}^3 \text{ hr}^{-1}$ . The samples were collected on 37 mm diameter polyvinylchloride filters with 0.8 micron pore size. Each IP sample was analyzed for Al, Br, Ca, Co, Cr, Cu, Fe, K, Mg, Mn, Ni, Pb, S, Se, Si, Ti, V and Zn by x-ray fluorescence spectrometer (Rigaku 3064). Details of the sampling analysis and instrumental calibration procedures have been presented.<sup>7</sup>

The meteorological data was obtained from the National Weather Bureau for all the seventeen sampling days. The gaseous sulfur ( $\text{SO}_2$ ) concentrations were taken from three  $\text{SO}_2$  monitoring stations (Cook County of Environmental Control) located around the sampling site. The final concentration of sulfur was found by adding up the concentration of sulfur in gaseous and particulate from ( $\text{SO}_4^{2-}$ ) as measured on the filter before the CEB analysis was carried out. Note that conservation of mass is a basic requirement of the CEB method.

### Source Characterization

The elemental composition of the modeled sources (source profile) used in equation (1) were developed from published data and is listed in Table 1. All data selected are based on the elemental composition of samples collected downstream of pollution control devices. In this way, the profile reflects actual source emission more accurately. The coal profile is an average of data from nine reports on the elemental composition of coal fly ash<sup>3, 6, 8-14</sup>. Limestone is a composite of the composition of Portland cement<sup>2, 3</sup>, cement plant emission<sup>15, 16</sup> and crustal limestone<sup>17</sup>. The mobile source profile is based on tunnel data<sup>18</sup> and an absolute concentration of lead of 10% by weight was assumed<sup>16</sup>. While the sulfur content (in the mobile source) was based on road test data<sup>19</sup>. Mobile profile from other CEB studies are also included<sup>20, 21</sup>. The refuse profile is based on stack data from three incinerators<sup>22, 23</sup>. The soil data are derived from geological survey data of the composition of surficial materials in Chicago area<sup>24</sup> and are supplemented by worldwide estimates of soil and crustal composition<sup>17</sup>. The steel profile is a composite of four published studies of particles emitted from iron and steel industries<sup>3, 14-16</sup>. Sulfur data for steel, refuse, limestone, and coal profiles were supplemented by an EPA emission inventory of  $\text{SO}_2$  emission from point sources in Chicago area<sup>25</sup>.

Table 1 Source profile matrix

| Element* | Coal      | Limestones | Mobile Source | Refuse    | Soil    | Steel   |
|----------|-----------|------------|---------------|-----------|---------|---------|
| Al       | 120,000   | 40,700     | 3,840         | 14,000    | 30,000  | 20,000  |
| Br       | 180       | 70         | 38,300        | 1,500     | 2.5     | 80      |
| Ca       | 43,000    | 221,000    | 8,850         | 17,000    | 8,000   | 30,000  |
| Co       | 55        | 7          | 11            | 6.6       | 20      | 6       |
| Cr       | 350       | 166        | 75            | 490       | 50      | 10,000  |
| Cu       | 390       | 235        | 750           | 1,700     | 30      | 7,400   |
| Fe       | 85,000    | 25,445     | 3,870         | 6,500     | 25,000  | 300,000 |
| K        | 10,400    | 18,488     | 7.8           | 0         | 20,000  | 35,000  |
| Mg       | 8,200     | 16,743     | 7,420         | 13,000    | 7,000   | 12,000  |
| Mn       | 410       | 943        | 310           | 730       | 300     | 44,000  |
| Na       | 9,500     | 8,700      | 390           | 82,000    | 6,300   | 10,000  |
| Ni       | 215       | 44         | 41            | 150       | 30      | 3,000   |
| Pb       | 380       | 27.5       | 100,000       | 81,000    | 30      | 7,000   |
| S**      | 3,882,000 | 248,390    | 7,590         | 1,960,000 | 260     | 240,000 |
| Se       | 290       | 40         | 8.1           | 37        | 0.1     | 45      |
| Si       | 233,000   | 163,200    | 16,950        | 0         | 270,000 | 6,600   |
| Ti       | 10,000    | 4,720      | 0             | 2,400     | 3,000   | 1,100   |
| V        | 520       | 104        | 10.6          | 31        | 100     | 670     |
| Zn       | 530       | 190*       | 1,960         | 120,000   | 50      | 9,000   |

\* — concentration in ug element i/gram emission

\*\* — total sulfur (gas + particle)

Equation (1) was solved for the source coefficients m for the FP, CP and IP fractions of the ambient aerosol on each of the 17 particle sampling days for the source profile. Occasionally, the WLS procedure estimated a negative source coefficient, which is physically meaningless. It is therefore necessary to assume that all negative coefficients are zero and to reestimate the other nonzero coefficients with the zero source removed from the analysis.

### Results and Discussion

Table 2 presents the averages and standard deviations of the FP, CP and IP source coefficients for the study period days. The unexplained particulate matter for the six sources in each size fraction is also included. The coefficients are calculated by the weighted least square procedure using all the six sources and nineteen elements in the source profile.

Table 2 Average fine, coarse and inhalable particulate matter source coefficients

| Particulate Fraction | Mean Source Coefficient, ug/m <sup>3</sup> (Standard Deviation) |                 |                |                |                |                | Unexplained* Particulate |
|----------------------|---|-----------------|----------------|----------------|----------------|----------------|--------------------------|
|                      | Coal  | Limestones      | Mobile Source  | Refuse         | Soil           | Steel          |                          |
| FP                   | 0.01<br>(0.05)  | 0.04<br>(0.09)  | 0.43<br>(0.57) | 3.14<br>(2.04) | 0.83<br>(0.89) | 0.23<br>(.26)  | 16.33<br>(14.91)         |
| CP                   | 0.60<br>(0.97)  | 9.76<br>(14.53) | 5.03<br>(7.33) | 2.22<br>(1.33) | 4.02<br>(8.30) | 0.92<br>(0.84) | 5.89<br>(11.80)          |
| IP                   | 0.40<br>(0.98)  | 7.75<br>(8.54)  | 5.14<br>(5.19) | 7.81<br>(2.47) | 5.47<br>(9.40) | 1.24<br>(1.07) | 21.64<br>(16.46)         |

\* Unexplained Particulate = Average Particulate Concentration — sum of six sources above

Table 3 further summarizes the averages, standard deviations and percentage source contribution of the FP, CP and IP source coefficients. The IP source contribution is considered as the studies best estimate.

This is because due to the fact that the source profile was never developed on FP or CP size fractions but rather it was based on particulate emissions (not specific to size) at the source.

**Table 3 Summary of fine, coarse and inhalable particulate matter source coefficients**

| Particulate fraction source | Source Coefficients ( $\mu\text{g}/\text{m}^3$ ) |                |                         |
|-----------------------------|--|----------------|-------------------------|
|                             | Mean   | Std. Deviation | Source Contribution (%) |
| <b>FP</b>                   |  |                |                         |
| Coal                        | 0.011  | 0.046          | 0.1                     |
| Limestone                   | 0.044  | 0.093          | 0.2                     |
| Mobile                      | 0.427  | 0.575          | 2.0                     |
| Refuse                      | 3.144  | 2.042          | 15.0                    |
| Soil                        | 0.835  | 0.889          | 4.0                     |
| Steel                       | 0.210  | 0.264          | 1.0                     |
| Unexplained                 | 16.335   | 14.908         | 77.8                    |
| <b>CP</b>                   |  |                |                         |
| Coal                        | 0.602  | 0.970          | 2.1                     |
| Limestone                   | 9.764  | 14.534         | 34.3                    |
| Mobile                      | 5.028  | 7.335          | 17.7                    |
| Refuse                      | 2.225  | 1.338          | 7.8                     |
| Soil                        | 4.020  | 8.301          | 14.1                    |
| Steel                       | 0.920  | 0.838          | 3.2                     |
| Unexplained                 | 5.894  | 11.797         | 20.7                    |
| <b>IP</b>                   |  |                |                         |
| Coal                        | 0.403  | 0.980          | 0.8                     |
| Limestone                   | 7.755  | 8.537          | 15.7                    |
| Mobile                      | 5.139  | 5.192          | 10.4                    |
| Refuse                      | 7.812  | 2.470          | 15.8                    |
| Soil                        | 5.466  | 9.403          | 11.1                    |
| Steel                       | 1.239  | 1.069          | 2.5                     |
| Unexplained                 | 21.636   | 16.465         | 43.8                    |

As shown in Table 3, refuse combustion make up the highest fraction of the FP and IP with 15% and 15.8% respectively. Based on the period average FP of  $21.0 \mu\text{g}/\text{m}^3$  and IP  $49.4 \mu\text{g}/\text{m}^3$ , refuse represents  $3.1 \mu\text{g}/\text{m}^3$  and  $7.8 \mu\text{g}/\text{m}^3$  respectively. This value is higher than previous studies.<sup>5</sup> Analytical error in measuring major elements in refuse is suspected. However, as expected refuse source is found to be predominant in the FP size fraction.

Limestone derived aerosols make up the highest fraction of CP (34.3%) and in IP (15.7%) next to refuse. Only small percentage of limestone source is found in the FP size range. Similarly, soil derived material contribute more in the CP (14.1%) than FP (4.0%). Most mechanical derived aerosols like soil and limestone are expected to be in the CP rather than FP size fraction. This finding is consistent with other studies<sup>26</sup>. Steel was found to contribute quite consistently in all size fractions. While coal combustion is higher in the CP than FP. Mobile contributes 2.0%, 17.7% and 10.4% in the FP, CP and IP respectively.

It is interesting point to note that while all the three size fractions were analyzed separately, the contributions from the unmodeled particulate matter fractions were  $16.3 \mu\text{g}/\text{m}^3$  and  $5.9 \mu\text{g}/\text{m}^3$  in the FP and CP respectively giving a total of  $22.9 \mu\text{g}/\text{m}^3$ . This is close to the unexplained particulate mass found in the IP ( $21.6 \mu\text{g}/\text{m}^3$ ). The unexplained particulate is the largest component in the FP fraction and it is believed that this particulate is predominantly consists of the secondary pollutant.

On average, the model was able to explain 22.2%, 79.3% and 56.3% of the FP, CP and IP respectively for the site.

#### *Point Source Validation*

A wind direction approach to model validation for point sources (coal, limestone, refuse and steel) was

considered. The wind directions were divided into four sectors at the receptor site; NW, NE, SW and SE. The daily IP source coefficients were grouped accordingly with respect to their vector average wind direction for the day. The highest average source coefficient in a given category will indicate the direction of the highest point source emissions upwind of the receptor and this is summarized in Table 4.

Table 4 Average point source contribution, inhalable particulate and unexplained concentration by wind direction

| Source                          | Wind Direction and Concentration in $\mu\text{g}/\text{m}^3$ |               |                |               |
|---------------------------------|--|---------------|----------------|---------------|
|                                 | NW [6]   | NE [4]        | SW [5]         | SE [2]        |
| Coal                            | 0.518<br>(41)  | 0<br>(0)      | 0.749<br>(59)  | 0<br>(0)      |
| Limestone                       | 7.008<br>(24)  | 2.446<br>(8)  | 13.657<br>(47) | 5.859<br>(20) |
| Refuse                          | 7.035<br>(23)  | 7.528<br>(24) | 9.350<br>(30)  | 6.843<br>(22) |
| Steel                           | 0.982<br>(16)  | 0.382<br>(6)  | 1.466<br>(24)  | 3.159<br>(53) |
| Ave. Inhalable<br>Particulate   | 47.565   | 19.295        | 76.591         | 47.642        |
| Ave. Unexplained<br>Particulate | 20.587   | 4.699         | 34.656         | 25.864        |
| Ave. Temperature*               | 66.5   | 64.0          | 74.0           | 70.5          |
| Ave. Wind Seed**                | 8.83   | 8.00          | 10.4           | 7.00          |

- [ ] — Number of samples  
 ( ) — Percent contribution  
 \* — Temperature of °F  
 \*\* — Wind speed in mph

Coal combustion was found to originate from NW and SW wind directions. The largest number of coal fired power plants are located in these directions<sup>25</sup>. Contribution from other directions were all zero. To a lesser extent, the coal impact would also be expected from the SE wind direction.

Refuse was found to contribute evenly from all four directions. Although, high refuse emission would be expected from the S wind directions. This finding further supports the conclusion that there may be an analytical error in the measurement of the major elemental components for refuse source.

In contrast to coal and refuse, limestone and steel were found to agree with known point source location relative to the receptor. Major steel industries are located in SE and SW of the receptor. For limestone, the SW, NW and SE wind directions were responsible for 47%, 24% and 20% of the limestone emission impact on the receptor respectively. Similarly, these findings confirm that most cement and limestone related industries one located in the SW and SE wind directions.

#### *Effect of Temperature on Unexplained Particulate Matter*

The unexplained particulate predicted by the CEB model were also considered for analysis. Assuming that a fraction of this unexplained mass if not all of it, consists of photochemical produced components, then, more of the unexplained would be expected to be more in hot-sunny days than cool days. Average daily IP unexplained source contributions below 70 °F and above or equal 70 °F and above or equal 70 °F were compared and is presented in Table 5. More unexplained was found in the higher temperature range. Thus, this concurrently increase the amount of IP in the environment. This confirms that higher temperature increases the reactivity of gaseous constituents in the atmospheric air resulting in greater particle production.

#### **Conclusion**

The application of CEB model as an apportionment technique in solving six sources in Chicago has been

Table 5 Effect of temperature on inhalable and unexplained particulate concentration\*\*

| Temperature, (°F)     | < 70 | > 70 |
|-----------------------|------|------|
| Number of Samples     | 8    | 9    |
| Unexplained Mass*     | 17.3 | 25.3 |
| Inhalable Particulate | 42.8 | 55.4 |

\* Inhalable particle size range

\*\* Concentration in  $\mu\text{g}/\text{m}^3$

presented. The solution of the CEB by weighted least square procedure is sensitive to daily changes in particulate source emissions and can be applied to estimate day to day variations in source contributions. Results indicate that a source profile matrix for FP and CP need to be developed in the future. But this requires intensive sampling programs and researches. The study has also demonstrated the utility of using the combination of the dichotomous sampler, XRF and CEB techniques for particulate monitoring and evaluation.

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