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Project Summary

Local and Regional Contributions to Urban Particulate Matter

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This report summarizes the data analysis of two extensive field studies on urban particulate matter: the 1974–77 St. Louis (RAPS) and the July/August 1982 Philadelphia (PAFS) studies. Special emphasis is placed on the "dichot" measurements of particles that segregated the fine fraction (<2.5 μm) and the coarse fraction (2.5 – 15 μm in St. Louis and 2.5 – 10μm in Philadelphia).

The major conclusion of this study is that in both cities the majority (more than 50%) of the total mass collected by the dichots is of regional, not local, origin. The regional share is about equally large for long-term (yearly, seasonal, monthly) and short-term (24-hours) averaging of concentrations. In the fine fraction, the regional component is even larger, 60% in St. Louis and 83% in Philadelphia. This conclusion is supported primarily by the observation that with the exception of a single site in both St. Louis and Philadelphia, all other sites within the extensive monitoring network, including remote rural sites, show a very low gradient of concentrations. This observation shows that the majority of collected aerosols do not originate from local sources, but must come from distant, regional sources that impact all monitors equally.

The annual average composition of PM-15 in St. Louis is 21% sulfate as SO_4, 39% crustal matter (SiO_2, Al_2O_3, Fe_2O_3, CaO, MgO), 35% "unknown" (i.e., not determined by the routine analytical method, X-ray fluorescence), and 5% others (mainly trace metals). The summer monthly average composition of PM-10 in Philadelphia is 34% SO_4, 14% crustal, 51% unknown, and 1% others. The unknown contains carbonaceous matter (elemental carbon and condensed organic matter, ammonium, nitrate, and water). Peak 24-hour compositions are not greatly different from the above.

Given the large proportion of regional contribution to and the chemical makeup of PM-10(15), neither dispersion nor receptor modeling based on local emission inventories and elemental composition is likely to accurately predict or interpret particle levels in urban airsheds.

This Project Summary was developed by EPA's Atmospheric Sciences Research Laboratory, Research Triangle Park, NC, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

Introduction

This report summarizes the findings of a two-year research effort on inhalable particle characteristics in urban airsheds. The research is based on data of two extensive urban field programs conducted by EPA in St. Louis (1974-1977) and in Philadelphia (1982). The St. Louis Regional Air Pollution Study (RAPS) was probably the largest monitoring effort ever undertaken to characterize the temporal and spatial characteristics of air quality in an urban environment, with special emphasis on inhalable particles. Among other monitoring devices, the RAPS campaign op-

erated 10 dichotomous inhalable particle samplers continuously for over three years, covering an area of 30 × 90 km. In this study, one year of data (1976) was analyzed. The Philadelphia Aerosol Field Study (PAFS) was of shorter duration, four weeks in July/August 1982, and of smaller spatial extent—six dichotomous samplers covering an area of 20 × 30 km.

This report provides an in-depth discussion and analysis of the two data bases, including determinations of particle composition, temporal and spatial characteristics, and meteorological influences. The data are then analyzed to separate the components of particulate pollution concentrations that are attributable to sources within or outside the local urban area. These are referred to as local and regional components, and the effort helps define the amount of air quality improvement that can be obtained from local emission controls. Next, dispersion and receptor (statistical source apportionment) models are applied to the data sets to evaluate model performance individually and in comparison. Finally, the report discusses the concept of combining dispersion and receptor modeling approaches in a mixed model that could improve predictive and analytic capabilities geared toward recommendation of control strategies.

Ambient Particle Data

High PM-10 and PM-15 levels generally result from large contributions of both fine and coarse size fractions. While fine and coarse fractions are about equal in St. Louis, in Philadelphia the fine fraction is dominant at most sites. Based on the different compositions and low correlations, the two size fractions appear to have largely separate origins.

About one-half of the fine fraction particles and one-third of PM-10 consist of SO_4. SO_4 levels are highest in the summer, probably due to the faster oxidation of SO_2 to SO_4 associated with increased photochemical activity, high temperatures and high humidities. At most urban and rural sites, concentrations of fine fraction mass and SO_4 are similar and their pollutant roses are nearly identical; both imply the importance of regional sources. At high concentrations, the fraction of trace metals, indicative of local sources, increases in St. Louis but decreases somewhat in Philadelphia, possibly due to the dominance of regional SO_4 in the latter city.

Coarse fraction particles contain large amounts of Al, Si, Ca, Fe and Mn and appear to be of crustal origin. This crustal material composes a relatively constant fraction of the aerosol. On average, coarse fraction levels in Philadelphia are much lower than in St. Louis (summer averages of 9 vs. 25 $\mu g/m^3$), probably due to the change in sampler design, which excludes larger particles. Source differences between the two cities may also be important. The highest coarse fraction concentrations occur during dry and dusty periods; concentrations tend to decrease during prolonged or heavy precipitation.

Local and Regional Components of Particles Concentrations

Because particles are formed and transported over long distances, both local and regional or distant emission sources contribute to ambient particle concentrations. Thus, particle concentrations may be viewed as the sum of contributions from local and regional emission sources. The regional component of the total particle mass collected at a receptor is termed "background." Hereafter, "regional component" and "background" are used interchangeably. Local sources, situated within the airshed, produce concentration levels which generally increase toward the sources. This is the "urban increment." The regional or background component arises from the long-range transport of pollutants into the airshed and attains about equal levels at all locations within the airshed.

It is assumed that the upwind or regional monitoring site receives the lowest concentration in the monitoring network. In both rural and urban areas, the local increment or contribution is the difference between the highest and lowest concentrations.

The annual average concentrations at the St. Louis sites of the PM-15, fine and coarse fractions, were calculated and the background share averages 57.5% of the PM-15 at the 10 sites. The fine fraction background is 60%; the coarse fraction background is 55%. The ratio of average concentrations at the central industrial site to the most outlying site is only about 2 for PM-15, 1.75 for fine fraction, and 2.3 for coarse. There are two significant conclusions to be drawn. Concentrations at rural sites are due almost entirely to background particles and therefore receive little contribution from the metropolitan St. Louis sources. Equally important, the background concentration is a significant fraction of the total concentration at each site, even for the central city sites. The interesting conclusion is that on low pollution days local sources may contribute more to the total concentrations. On high pollution days (when standards are likely to be exceeded), local impacts become less pronounced and most particles seem to come from outside the network.

The monthly average concentrations were considered at the six Philadelphia sites. Site averaged concentration are computed for five sites only, excluding one with peculiar results. The background share of the average PM-10 concentrations is 77%. The fine fraction background is 83% and the coarse 64%. The ratio of concentrations at the center city site to that at the most rural site is less than 1.2 for total mass and fine fraction, and 1.6 for the coarse fraction. Thus, in Philadelphia the background share is even larger than in St. Louis, and the relative difference between city center and outskirts is smaller. Looking at daily average concentrations at sites in Philadelphia, the ratio to background is usually less than two. As in St. Louis, the range is greater at lower overall concentrations. This again indicates that the proportionate contribution of local sources is less at high concentrations when exceedances might occur.

In summary, the regional component composed well over half of the average fine fraction levels at most monitors in both cities and about half of the coarse fraction concentrations. The background fraction is relatively constant; thus, local and regional levels appear to change together.

Dispersion Modeling

A long-term version of the Particle Episodic Model (PEM) is applied to apportion and predict particle concentrations in St. Louis and Philadelphia. In the evaluation, only the local contributions are modeled. The observed local component is obtained by subtracting the regional component as described in the preceding section.

Estimated Philadelphia fine fraction emissions are about twice the coarse fraction, whereas in St. Louis the coarse fraction emissions are estimated to be higher. These estimates may in part explain the observed differences of ambient particle levels in the two cities. Area sources are the primary contributors of

particles. Host cells, the area source cell in which the receptor is located, account for about half of the total particle prediction. Point sources produce roughly half of the SO_2 and SO_4. Generally, a few sources provide most of the contributions at the receptors.

In St. Louis, however, particle levels are overpredicted, especially in the coarse fraction, and predictions do not have the correct spatial distribution. Most likely, the St. Louis emission inventory does not accurately reflect PM-15 emissions. In St. Louis, fine and coarse particle emissions were derived from TSP data, which in many cases may not be related to PM-15 emissions but rather to fugitive dust. At short averaging times, model predictions have low correlation (0.2-0.3) with observations. In Philadelphia, long-term predictions are in reasonable agreement with measurements. The mean levels and the spatial variation at most sites are quite well modeled.

Statistical Models for Source Apportionment

Receptor methods are useful only for the apportionment of those sources that have distinct composition and are predominantly of local origin, e.g., vehicles, incinerators, metal processing industries, and some oil-related sources. Receptor models do not separate local from regional sources that have similar elemental composition.

According to the multiple linear regression (MLR), 56% of the fine fraction and 34% of the total PM-15 levels in St. Louis are because of SO_4. Previous studies using summer data only, attributed a greater percentage to SO_4 (59 to 84% of the fine fraction). This could be due partially to higher transformation rates in the summer. Apportionments of other sources are similar to the previous studies. Crustal sources account for 85% of the coarse fraction and 16% of PM-15. Other sources tentatively identified include road salting, industrial emissions and incineration. The MLR model explains 50-90% of the variance of particle concentrations and indicates that S, Ca and Cu are stable tracers.

In Philadelphia, the MLR approach indicates that SO_4 accounted for 52-65% of fine concentrations and 39-52% of PM-10 concentrations. Crustal components account for 30-50% of the coarse fraction; oil and/or refinery sources (V) account for 2-11% of PM-10 concentrations; vehicular sources may contribute 8-15% of PM-10. The identification of other factors is more speculative. Incineration (Cu) may account for 5-8% of fine fraction concentrations; and crustal or fertilizer sources (P, K) may compose 6-25% of particle concentrations.

MLR results indicate that the mass loading factor of elemental S is 5.1 in St. Louis and 4.1 in Philadelphia. The stoichiometric factor for $(NH_4)_2SO_4$ is 4.1; thus, in St. Louis, the sulfur components may have carried additional matter, perhaps water.

Mixed Models

With a few exceptions, statistical (receptor) and physical simulation (dispersion) models remain separate approaches in air quality modeling. To provide more accurate results, a "mixed model" was developed that incorporates features of both dispersion and receptor modeling.

In some respects, the mixed model described here resembles the "inverse" dispersion model and state-space approaches. In the inverse dispersion model problem, source emissions (rather than ambient concentrations) are predicted using ambient observations. The mixed model differs in that multiple pollutants, source, compositions and prior information are considered.

The model consists of several components. First, a dispersion model is used to calculate transfer coefficients. These coefficients indicate the contribution of local emission sources to receptors for the meteorological conditions during the sampling period. The Particle Episodic Model (PEM) is used with optimized dispersion parameters. Second, predictions of mass and elemental concentrations at receptor sites are made using the transfer coefficients and prior information. The latter includes the elemental composition and the particle emission rate of sources. Spatial and temporal aggregation is used to reduce the number of unknowns and simplify the estimation problem. Third, the prior information—emission rates and source compositions—is revised so that predicted mass and elemental concentrations best correspond to ambient measurements. Measurements collected at all receptor sites are used. Linear Bayesian estimates are used to correct for missing data. As the distributions of the parameters are unknown, a parametric approach is used in which ranges of uncertainties and covariances are selected. The primary outputs of the model are posterior estimates of emission rates and elemental compositions. Apportionments are derived as the product of the estimated emission rates and the transfer coefficients.

Mixed models may be used in many applications. First these models can reconcile different apportionments generated by simulation and receptor models. Second, the results provide a check on the accuracy of the source inventory. Third, the appropriateness of source compositions may be assessed. Fourth, it may be possible to identify unknown sources or detect locations of accidental releases. Fifth, the approach forces the explicit quantification of uncertainty.

Conclusions

While the two field studies were different in scope and extent, and even used different instrumentation and schedules, the main conclusion is the same: the majority (more than 50% of the inhalable particle mass, whether averaged over a day, month, or year is not attributable to sources that lie within the metropolitan city limits, but is probably because of regional sources.

Other conclusions regarding inhalable particles in St. Louis and Philadelphia follow:

- In St. Louis, the fine fraction (less than 2.5 μm diameter) and coarse fraction (2.5-15 μm) masses are about equal; in Philadelphia the fine fraction (<2.5 μm) and coarse fraction (2.5-10 μm) masses are about 3:1.
- In St. Louis, about 40% of the fine fraction consists of sulfate (SO_4); this percentage is even larger if it is assumed that most of the sulfate consists of hydrated ammonium sulfate. In Philadelphia, 43% of the fine fraction is SO_4. The coarse fractions are dominated by crustal components.
- In St. Louis, 24-hourly PM-15 concentrations exceeded 150 $\mu g/m^3$ several times, which is the proposed lower range of the 24-hour PM-10 standard for particles. In Philadelphia, during the four weeks of monitoring, this range was approached only once at one site.
- The ratio of annual network averages of inhalable particle mass to total suspended particle mass is about 0.5; however, the ratios of 24-hour averages may have a wider range, from 0.25 to 0.75.

- Dispersion modeling, at best, can account only for the local increment of inhalable particles; not the background that, as indicated above, appears to be of regional origin. Dispersion modeling requires an accurate emission inventory of inhalable particles (and their gaseous precursors), which cannot be obtained simply by assuming that there is a constant ratio of emissions of IP/TSP.
- Receptor modeling is only useful for the apportionment of those sources that have distinct compositions and are of local origin, e.g., vehicles, incinerators, metal processing industries, and oil combustion-refining. Many sources have similar profiles and cannot be separated by receptor methods.
- The mass loading factor of elemental sulfur (sulfur-related mass divided by sulfur mass) is about five in St. Louis and about four in Philadelphia. This factor is larger than the stoichiometric factor of three for SO_4, indicating that elemental sulfur carries along some other species, probably ammonium and water.
- A "mixed" dispersion-receptor model was developed in which multiple pollutants, source compositions, and dispersion model-derived transfer coefficients were used. The mixed model is computationally very intensive but yielded some useful information regarding emissions.

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The complete report, entitled "Local and Regional Contributions to Urban Particulate Matter," (Order No. PB 86-236 965/AS; Cost: $11.95, subject to change) will be available only from:
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