SOURCE APPORTIONMENT AND THE UNITED STATES CLEAN AIR ACT

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The United States Clean Air Act of 1977 requires that the Environmental Protection Agency review the National Ambient Air Quality Standards (NAAQS) at five year intervals. As part of a review of the standards themselves, methods for measuring compliance with the standards have to be published. During the current review, there will be a major alteration in the NAAQS for particulate matter; the total suspended particulate (TSP) standard, currently based on collection devices with 50% collection efficiency at 15μm particle diameter, will be replaced with the PM10 standard, based on 50% collection efficiency at 10μm.

Questions on the comparability of TSP and PM_{10} measurements have attracted much research effort (1), for two important reasons:

1) The historical database of air quality data obtained with high volume samplers with 15 μm inlets represents a substantial investment which cannot be abandoned. Longer term trends in pollution levels form an integral part of control strategies in achieving attainment in terms of State Implementation Plans (SIPs).

2) Investment in existing 15 μm samplers is such that transition to new μm inlets would have to be phased in over a period of time. The conversion factor from TSP to PM_{10} measurements is found to be approximately 60%, averaged over large numbers of samples.

One important consequence of choosing a smaller diameter upper cutpoint is that the fine aerosol component, i.e. the conversion aerosol less than 3 μm aerodynamic diameter, forms a correspondingly larger fraction of the PM_{10} measurements. For a region struggling to meet the particulate standards, this causes additional problems, as up to 50% of urban particlate mass can consist of fine aerosol transported into that region from distant sources! A disproportionate, perhaps even impossible, task would then be placed on local regulators if they tried to attain the standard by ever more stringent control of local industrial, vehicle and domestic sources.

For this reason, the EPA requires as part of the State Implementation Plans for each region, that the sources contributing to ambient pollution should be quantitatively identified. The methods for achieving this, broadly referred to as SOURCE APPORTIONMENT techniques, should be quantitative, reproduceable and verifiable. The verification is achieved by obtaining similar results from two or more independent techniques.

The longest established family of techniques for source apportionment are dispersion models. These require as input detailed source inventories of the specified region, including source location, emission levels and variations, and local meteorology. With this information, mathematical models are formulated to predict levels of pollution at a given location, and the relative amount contributed by each source. Predictions provided by current models are generally adequate for longer term averages, i.e. for seasonal or annual averages, but are less good for 24-hour predictions. Disperson models are particularly useful for planning purposes, since the impact of additional sources can easily be included in an existing model. Shortcomings of disperson models are that: model results give no indication of missing sources (i.e. not included in the inventory); fugitive sources are difficult to model; only a few models make allowance for chemical transformation or settling of particles during transport; and models do not cope well with complex, mountainous terrain.

A second group of source apportionment techniques fall into the class of RECEPTOR MODELS. In these, the chemical and physical characteristics of samples collected at an ambient site (the receptor site) are examined. These properties are then matched to particle properties of various sources, and apportionment of the relative contribution is made by mathematical or statistical means. Numerous analytical techniques are available for use in receptor modeling; Light Microscopy, Automated Scanning Electron Microscopy (SEM), Ion Chromatography (IC), Instrumental Neutron Activation Analysis (INAA), Xray Diffraction (XRD), Xray Fluorescence (both conventional – XRF – and particle induced – PIXE).

Light Microscopy is one of the oldest and perhaps the easiest to grasp conceptually. Receptor modelling proceeds by optical identification and counting of particles in various categories, say soil, carbonaceous, fly-ash, pollen and auto-exhaust particles. The relative numbers in each class are assumed to be in proportion to the contribution from each of the corresponding source types. Optical counting is, however, a highly skilled and labour intensive operation and even experienced microscopists can reach different results from the same sample. The advent of automated scanning electron microscopy, which can characterize individual particles on the basis of their chemical composition has removed much of the tedium and arbitrariness from individual particle analysis.

If bulk chemical analysis of filter samples is performed, various mathematical techniques may be applied. Regression, principal components and factor analyses are frequently used where multiple samples (preferably forty or more) are available. These statistical techniques are useful in situations where detailed knowledge of local sources is not available. If, however, local sources can be sampled, then Chemical Mass Balance (CMB) modelling may be applied. In these CMB models, observed chemical concentrations of a receptor sample are matched to a set of

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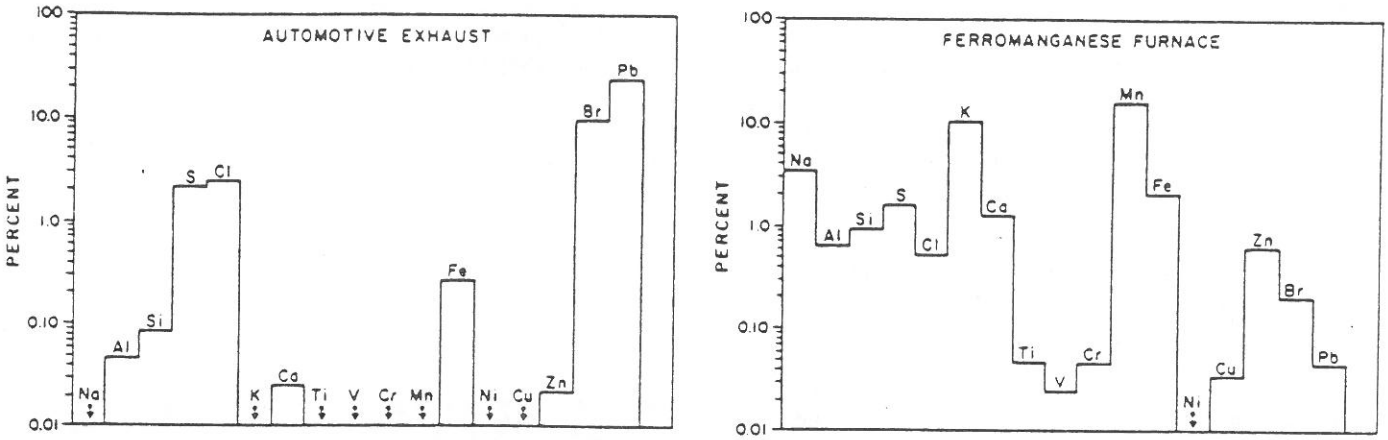

Figure 1. Examples of elemental source fingerprints used in Chemical Mass Balance receptor modeling. (From Portland Air Characterization Study, Watson and Cooper)

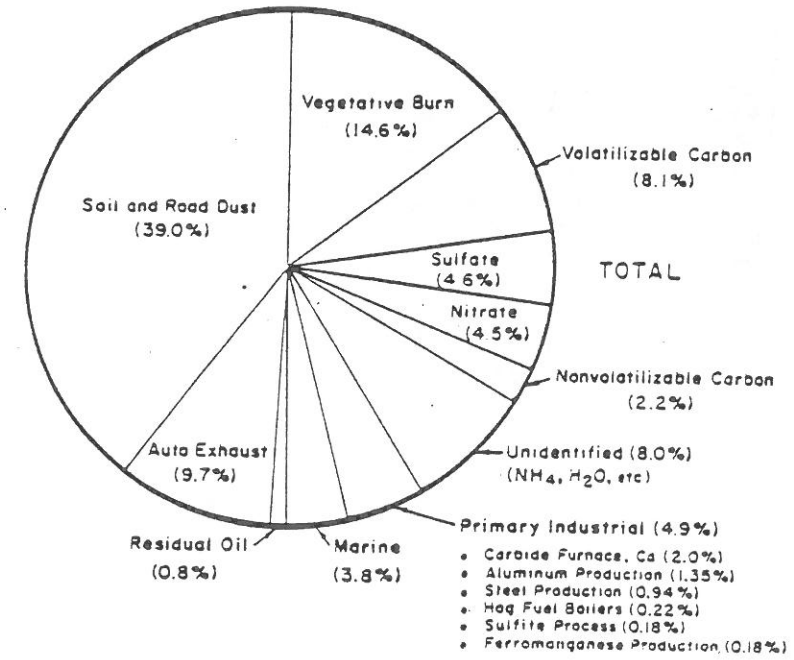

Figure 2. Source contributions to air particulates in downtown Portland, Oregon. Annual Stratified Arithmetic Average. (From Portland Air Characterization Study, Watson and Cooper)

'fingerprints' or 'profiles' of the chemical composition of the sources, as shown in figure 1.

Typical results for a CMB model are shown in figure 2, taken from the Portland Air Characterization Study. An unexpected outcome of that study was that industrial sources, originally thought to be major contributors to particulate pollution, were apportioned less than 5%of the total. Domestic wood-burning stoves (vegetative burn) were found to be larger contributors, up to 20%, to ambient pollution.

CMB modelling has the great advantage of being applicable for the analysis of single samples or events (as opposed to forty or more samples required for the multi-variate statistical techniques). It is thus suitable for short term (24-hour) and worst episode analyses. CMB, while more precise than the multivariate statistical techniques, does require that all contributing sources be identified, and that major sources be 'fingerprinted'. (CMB does have limited ability to identify the characteristics of sources missing from the inventory). CMB has the ability to identify source categories; if sources have similar fingerprints, e.g. agricultural soil, road dust and coal fly ash, then CMB would have difficulty in distinguishing between them.

Receptor models, including CMB, do not have any predictive power and are therefore not suitable planning tools. However, CMB has been used to advantage in calibrating the results of dispersion models, and in this sense provides an important tool for independent validation of dispersion models in particular areas.

As part of the developing methodology of CMB modelling, a great deal of emphasis has been placed on the planning stages. Since the cost of source sampling and sample analyses can be considerable, it is imperative that sampling campaigns be designed to achieve the required degree of precision within budgetary limits. The old 'shot-gun' approach to sampling, in which large numbers of samples were collected and analysed, and only then was thought given on how to interpret them, is now fortunately being replaced with coherent and objective methods for designing sampling strategies.

A readable introduction to CMB modelling is given by Cooper and Watson (2). For the more technically minded, a comprehensive overview of CMB receptor modelling appeared in a series of six articles in Atmospheric Environment. (3) Public domain software for running CMB models is available from the U.S. EPA; copies have been acquired by the Nuclear Environmental Group at the Schonland Research Centre at Wits. Through various research programmes and workshops with interested parties, it is hoped to evaluate and develop CMB methods for application in the South African context.

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