



Designing ambient particulate matter monitoring program for source apportionment study by receptor modeling

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ABSTRACT

Particulate matter (PM) receptor modeling requires specific intensive input data that is always a challenge to produce cost effectively. A well-designed monitoring program is important to collect such PM ambient data in urban areas with diverse and densely distributed sources. This paper presents a general framework for designing such a monitoring program while emphasizing appropriate quality assurance and quality control elements that are particularly applicable where limited resources are available. Topics for discussion include selection of monitoring sites, sampling and analytical techniques, and the uncertainty estimation for ambient concentration input data. The design framework is illustrated by a case study of a monitoring program for PM source apportionment in the Bangkok Metropolitan Region in which 24-h fine and coarse PM samples were collected using two collocated dichotomous samplers. Comparison between black carbon measurements by Smoke Stain reflectometry and Thermal Optical Transmittance method is highlighted.

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1. Introduction

Available monitoring data show high particulate matter (PM) levels in the world megacities (Molina and Molina, 2004), especially in the developing countries (Kim Oanh et al., 2006; HEI, 2004; Gupta et al., 2006; Kim et al., 2006; Chan et al., 2001; Tsai and Chen, 2006; Hopke et al., 2008). Fine PM, especially those with an aerodynamic diameter $\leq 2.5 \mu\text{m}$ (PM_{2.5}), in particular, are believed to be one of the most harmful pollutants to human health (HEI, 2002; Pope et al., 2009). PM can also introduce adverse effects on the atmosphere and climate (Bond et al., 2004; Ramanathan and Carmichael, 2008). Understanding the nature (e.g. source, size, chemical composition, etc.) of PM pollution in an airshed is the first step towards managing particulate air quality to minimize its associated effects.

Particles present in the ambient air are of different origins. Some particles are directly emitted from the sources (primary particles) while others (secondary) are formed in the atmosphere, for example, through the chemical transformation from gaseous precursors. A quantitative assessment of the source contribution to the ambient PM is required to prioritize the sources for the abatement purpose. The “bottom-up” approach based on emission inventories is often limited by the lack of accurate emission factors,

especially in developing countries. In addition, the emission inventory cannot directly account for secondary particles that are formed in the atmosphere. Dispersion modeling, in principle, can estimate the impact of source emissions to ambient air pollution levels and advanced chemical transport models have the ability to predict secondary aerosol formation in the atmosphere (Mysliwiec and Kleeman, 2002; Sun and Wexler, 1998). However, advanced 3D dispersion models dealing with PM formation require intensive input data, e.g. detailed gridded emission inventory, and other resources that may not be readily available everywhere. Receptor modeling, known as a “top-down” approach, on the other hand, does not require the emission inventory in detail, but it does need intensive ambient monitoring data at a receptor site to permit accurate source apportionment. Receptor modeling can be used as an independent check for the accuracy of emission inventories, and in many cases, it reveals important sources that may have been overlooked by the emission inventories. In the absence of a reliable emission inventory in a study area, source apportionment analysis alone may offer useful information on the major contributing sources to plan abatement strategies. The bottom-up and top-down approaches, however, complement each other to provide a better understanding of source–receptor relationships that is required for the development of effective air quality management strategies.

As a result of complex processes of emission, dispersion and transformation, PM in the atmosphere is a complex mixture of various chemical species that may carry signatures of its origins.

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Principally, receptor modeling hypothesizes that PM emitted from a certain source type is characterized by its distinct chemical and/or physical properties that subsequently appear in the ambient PM at a receptor site and, therefore, can be used to identify and quantify the contributing sources. Receptor models can be divided into two groups: i) the microscopic models that use physical features such as color, shapes, or configuration of particles to qualitatively identify their types and the sources (Cooper and Watson, 1980; Kim et al., 2004; Zhou et al., 2005a), and ii) the chemical-based models that require the chemical composition of PM at ambient receptor sites and at emission sources. The chemical-based models include those that produce qualitative source information (enrichment factor or time/spatial series) and others that produce quantitative source apportionment (chemical mass balance and advanced multivariate analysis). The source fingerprints or marker species are used to characterize respective source emissions, either by their abundance or their relative proportions (profile). The source profiles normally represent a category of sources in urban areas, such as diesel exhaust, rather than individual emitter such as a single diesel vehicle. Depending on the types, receptor models require the source profiles either explicitly (e.g. Chemical Mass Balance or CMB) or implicitly (multivariate models).

Receptor modeling results contain many uncertainties associated with the accuracy of source information, the quality of the ambient PM composition data, and the assumptions in the formulation of a particular model. The evaluation of receptor modeling results is not as straightforward using a direct comparison with the monitoring data as dispersion models. Normally, the receptor models have a set of criteria for model performance that are mainly statistical estimates such as χ^2 , R^2 , TSTAT for CMB (Watson et al., 1997) or Q value and scaled residual distributions for the Positive Matrix Factorization (Paatero and Tapper, 1994). Recently, the advanced tools such as the nonparametric regression (NPR) and the conditional probability function (CPF) that analyze the local source contributions in relation to wind have been applied to find the source directions (Kim et al., 2003a). Likewise, the potential source contribution function (PSCF) has been applied to analyze regional source contributions based on the air mass trajectories (Kim et al., 2004; Prapat and Kim Oanh, 2007). These tools can help to qualitatively verify the receptor modeling results. There are also other trajectory ensemble methods such as Residence Time Analysis (Poirot et al., 2001), Residence Time Weighted Concentrations (RTWC) (Stohl, 1996) and Simplified Quantitative Transport Bias Analysis (SQTBA) (Zhou et al., 2004) that can be used to evaluate receptor model results. These tools can help to increase the degree of confidence in the receptor model results but cannot increase the accuracy of the results. The accuracy of the model output mainly depends on the quality of the ambient input data, the availability of required information on the source profiles, and the proper application of the models.

Thus, reliable and representative PM ambient monitoring data at receptor sites are the first requirement for the accurate source apportionment by receptor modeling. PM data with established quality assurance and quality control (QA/QC) protocols such as those available from the Interagency Monitoring of Protected Visual Environments Network (IMPROVE) (<http://vista.cira.colostate.edu/improve/>), State and Local Air Monitoring Stations (SLAMS) (<http://www.epa.gov/ttn/amtic/pmqaifn.html>) and Speciation Trends Network (STN) (<http://www.epa.gov/ttn/amtic/speciepg.html>) in the US do not exist in many other places, especially in developing countries. Therefore, the first task in a receptor modeling study is to collect required PM ambient data. This is a resource consuming activity that needs appropriate monitoring design with an established QA/QC protocol to produce reliable data in a cost-effective way.

This paper presents the design of a monitoring program for fine ($PM_{2.5}$) and coarse ($PM_{10-2.5}$) PM for receptor modeling purpose on an urban scale. The program was implemented to collect data in the Bangkok Metropolitan Region (BMR) of Thailand. The research is a part of the activities of AIRPET (Asian Regional Air Pollution Research Network; <http://www.serd.ait.ac.th/airpet>) during phase 1 (2002–2004). The focus of this paper is on the QA/QC related to sampling and analysis of PM, namely collocated sampling, calibration of a reflectometer for black carbon (BC) measurement, and estimation of the uncertainty of input data for receptor models. Details of PM compositions, the receptor modeling results, and the source profiles developed for Asian cities are presented in separate papers.

2. Monitoring design

2.1. Sampling

2.1.1. Siting

Siting is always a challenging task in crowded and congested urban areas in developing countries, where mixed land use patterns are present, especially when limited resources prohibit having numerous sites. Sampling sites with known strong impacts from certain sources such as roadside or industrial estates are expected to have higher contributions from the nearby sources and may also provide relevant source signatures. They, however, would not represent the overall picture of the PM pollution in an urban airshed. Instead, for source apportionment, a few but representative mixed sites are generally desirable. Because of the complex urban activities forming different air bubbles (sub-airshed affected by common sources) in a large urban area a few sites are required for PM source apportionment. For example, Gordon (1988) mentioned that at least 6 sites are required in most of the receptor modeling studies and more would be required if there are unique sources present. These sites would capture common urban air bubbles such as upwind, downwind, city center, commercial, residential, industrial, etc. Thus, each site should represent an air bubble and should reflect the representative mixture of PM from major contributing sources to that air bubble. Careful consideration of the emission source distribution, meteorology (windrose) and typical dispersion patterns (upwind or downwind sites of major sources) in a study city during a year should be taken when sites are selected. The sites are expected to receive emissions from major sources in the air bubble under the prevalent wind directions. Ideally, they should be located in such a way that major sources can be discriminated from each other by wind trajectories. Additional information on major sources such as stack height (affecting dispersion) or emission temporal variations is also important. Sites located upwind and downwind of a city help in understanding the air pollution transport across the study area. If remote sites are added, then regional scale phenomena such as long range transport can also be reflected. Auxiliary information such as meteorology and gaseous pollutants at selected monitoring sites can greatly aid to the evaluation of source apportionment results. Some advanced receptor models, such as those based on the Multilinear Engine (ME) algorithm (Kim et al., 2003b; Zhou et al., 2005b), can directly input this information into analysis; hence availability of this information should also be considered in the site selection.

For illustration, a monitoring for PM source apportionment study for BMR, which has an area of over 7700 km² with around 10 million people and mixed human activities, is presented. BMR climate has two distinct seasons: dry season (mid October to mid May) under the influence of NE monsoon with a total rainfall of 550 mm, and the wet season with SW monsoon and 1150 mm total rainfall. All the year around wind is normally low, 1–3 m s⁻¹, and the average temperature

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