Exposure to Airborne Particles and Gases and Apportionment to Major Sources in the Detroit Area (The Detroit Exposure Aerosol Research Study)

Revised Study Plan after Peer Review
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1.0 INTRODUCTION

1.1 Background

The goal of EPA's air program is to reduce emissions, which in turn, will improve air quality, reduce exposure to pollutants that cause health impacts, and improve health (US EPA OIG Report, 2002). To achieve this goal, the Clean Air Act includes provisions that address criteria pollutants, such as particulate matter (PM), and hazardous air pollutants or air toxics. For criteria pollutants, the Agency develops and implements air quality standards that are protective of health by establishing ambient concentration levels above which health effects are determined to be unsafe. For air toxics, the Agency is directed to develop standards that address significant sources of pollutants like major industrial sources and mobile sources and to address urban air toxics by developing standards for the smaller area sources. Many of these standards will require a risk assessment, which involves an understanding of how human exposures are affected by specific sources of air toxics. Setting a standard that is defensible and protective of human health requires a comprehensive scientific understanding of exposure to ambient pollutants and the exposure-response relationship.

Particulate Matter
In July 1997 based on an extensive review of the literature, the EPA Administrator issued new National Ambient Air Quality Standards (NAAQS) for particulate matter (PM) that revised the standard for PM_{10} and added a standard for $PM_{2.5}$. The new standards were developed largely on the basis of the epidemiological studies that found consistent associations between PM concentrations measured at central site monitors and various adverse health effects. However, individuals develop an adverse health response to PM in the air that they breathe, not the air at a central site monitor. It seemed almost counter-intuitive that monitors representing the widely distributed PM mass within an air shed could serve as a surrogate for individual human exposures, given the diversity of life-styles and activities. Thus, understanding personal exposures to ambient PM provides a critical link between regulatory monitoring and health outcomes. Specifically, understanding the relationship between PM measured at central site monitors and residential outdoor, indoor, and personal exposure concentrations to ambient PM is essential to understanding risks.

In 1997, Congress requested that the National Research Council (NRC) review EPA's Office of Research and Development's (ORD's) PM research program with the objective of clearly defining research which would reduce "uncertainties in the scientific evidence used to guide regulation of airborne particulate matter in the United States." Ten priority research areas were identified in the first NRC report. Two of the ten highest priority research activities were directed towards understanding PM exposures. **Research Topic 1** focused on understanding how susceptible sub-populations were exposed to ambient PM mass and how these exposures related to concentrations at the central monitor. **Research Topic 2** extended the research to address potentially toxic components of PM and the general population as well as susceptible sub-populations.

The need to link PM measured at central site monitors to personal exposure was recognized early on by ORD. Understanding the source-to-personal exposure component of the risk paradigm became a primary focus of the PM Research Program.

The goal of ORD's exposure program is to develop data and models that characterize and predict human exposure to PM of ambient origin relative to that measured at central sites. Research was initiated in 1997 with a focus on susceptible sub-populations and PM mass to address NRC Research Topic 1. Longitudinal PM exposure studies were conducted to characterize inter-personal and intra-personal variability in exposure to PM mass, and to describe the relationship between personal exposures and ambient exposure estimates based on central site monitoring. Results from these studies have verified that for fine PM mass and sulfate, the central monitoring site should serve as an adequate surrogate for exposure in community-based epidemiological studies. Differences between ambient levels and estimates of personal exposure should not change the conclusions regarding epidemiology-based health outcomes, although the strength of the association has been shown to vary by location, housing characteristics, and season. Since individuals are typically exposed to lower levels of ambient PM than would be predicted by central site monitors, the strength of the impact may be underestimated. In addition, since the ratio of personal exposure concentrations to central site concentrations (i.e., the attenuation factor) can change by city and season, a single nationwide PM standard may provide a different degree of protection for different populations. Recent studies have not shown significant differences in personal exposure to ambient PM as a result of disease-state.

As a result of our work in NRC Research Topic 1, databases have been developed to evaluate exposure relationships for PM mass. EPA's exposure program is now focusing on understanding exposures and exposure relationships for PM components as specified under NRC Research Topic 2. In the next ten years, it is anticipated that EPA regulations will dramatically reduce ambient air concentrations of fine particulate mass and sulfates. As demonstrated in recently completed exposure studies, these two species are well behaved, and exposure and health effects can be reasonably predicted from central site monitoring data. To ensure that our standards protect human health in the future, however, we must be able to evaluate exposure and health effects for those PM species that will remain after particulate sulfates are removed. Preliminary data show that outdoor to indoor correlations are poor for several species including ultrafines, acid aerosols, and nitrates. Other studies have shown that outdoor concentrations of elemental carbon and several organic species are not homogenous across air sheds and are influenced by both mobile and stationary sources. For those species that show only weak associations between central site concentrations and exposures or are not well distributed across air sheds, it is unlikely that community-based epidemiological studies can be used to adequately evaluate health impacts. For these species, data and models will be needed to develop better exposure surrogates for epidemiological studies and to conduct high quality exposure and risk assessments.

Understanding the association between central site monitoring and ambient exposure (i.e. the ambient component of total personal exposure), understanding the non-ambient

exposure component, and knowing the correlation between ambient and non-ambient exposure for PM mass, PM species and PM sources will be important in the process of determining whether to revise existing standards. Currently, a national monitoring network for PM (NCore) is being developed which includes both integrated and real-time measurements of PM species. It has been proposed that these data, along with data from the Speciation Trends Network (STN), can provide a foundation for future epidemiological studies. In addition, the Office of Management and Budget (OMB) is suggesting that epidemiological studies should be conducted in 50 cities across the U.S. using the Aerosol Research and Inhalation Epidemiology Study (ARIES) as a model where health outcomes for PM species were evaluated using a single community monitor. Before these data can be used in epidemiological studies, it must be shown that the central site monitor adequately represents exposure for PM species. In addition, models and information on exposure factors need to be developed to predict exposure based on measurements at a central site monitor.

As originally stated, NRC Research Topic 2 was to evaluate exposure relationships for the causal agents of PM toxicity. Although substantial research has been conducted to understand the mechanisms of PM toxicity and to identify causal agents, specific toxic agents have not yet been identified, rather there is evidence that supports health effects associated with most of the originally hypothesized toxic agents. Concurrently, several epidemiological studies have shown health effects associated with PM from specific sources rather than focusing on individual components. In light of these findings, emphasis is now being placed on understanding exposure and health effects of PM from specific sources. Source apportionment techniques are being incorporated into exposure research in order to evaluate the ambient-personal exposure relationship for PM from various sources as well as for individual PM species. Linking specific sources through central site concentrations and human exposures to health effects is likely to provide data that can be applied to regulatory policy more quickly, and can help support identification of biologically important characteristics and constituents as well.

Air Toxics

With the July 19, 1999 Federal Register publication of the *National Air Toxics Program: Integrated Urban Strategy,* the EPA provided a road map for its air toxics program. This program is designed to characterize, prioritize, and equitably address exposures to air toxics and their serious impact on the public health and the environment through a strategic combination of regulatory approaches, voluntary partnerships, ongoing research and assessments, and education and outreach. The program addresses air toxic emissions from large and small stationary sources, mobile sources, and indoor air sources as part of its strategy for reducing risks from exposure to air toxics. In addition, the program prioritizes its actions and measures progress through the use of assessments conducted at multiple scales (e.g., national, regional, local).

EPA is currently working to characterize the extent of the air toxics problem. National Air Toxics Assessments (NATA) are one of the four components in EPA's risk-based National Air Toxics Program and include all of the exposure and risk assessment activities. NATA is intended to provide EPA and others with improved characterization

of air toxics exposures and risks based on emissions data for both stationary and mobile sources, as well as relative risks from indoor air exposures. The NATA are estimated using the hazardous air pollutant exposure model (HAPEM) to calculate the expected inhalation exposures of air toxics for population groups using census data, human activity patterns, ambient air quality levels, climate data and indoor/outdoor concentration relationships. Ambient levels of air toxics are predicted in the NATA using the assessment system for populaiton exposure nationwide model (ASPEN), an air dispersion model based on the industrial source complex long term model (ISCLT). The ASPEN model uses emissions data from both point and mobile sources along with meteorological data to estimate dispersion and atmospheric processes that affect the pollutants (deposition, reactions, secondary formation, etc.). NATA activities also integrate various Office of Air Research (OAR) office-specific components such as:

- Residual risk assessments for regulated stationary sources;
- Listing and de-listing of hazardous air pollutants;
- Research in mobile source air toxics assessments
- Research to support the development of an Indoor Air Strategy as well as the conduct of indoor air assessments; and,
- Community-based risk assessments and risk reduction projects.

Unfortunately there are limited monitoring data for assessing the extent of the air toxics problems. EPA is establishing an ambient air monitoring network. Starting in 2003, thirteen National Air Toxics Trends Sites (NATTS) will be established, with nine more being added in 2004. These trends sites will measure several key air toxics pollutants that have been determined to drive risk in assessments conducted to date. In addition to the NATTS, the ambient air toxics monitoring program will also include community-based air toxics monitoring projects designed to characterize air toxic concentrations across a community and to identify sources of potential concern.

There have been some notable air toxics human exposure studies conducted in the past. These include the TEAM studies (Wallace et al., 1985) and the RIOPA studies (Naumova et al., 2003). The efforts above were limited in scope in that they were not linked to the development of human exposure models. Likewise, they did not attempt to estimate the contribution of major line, point, and regional pollutant sources upon personal and residential settings.

The Detroit Exposure and Aerosol Research Study (DEARS) is intended to provide additional data that can be used to evaluate the extent of the problem. Detroit was selected based on the presence of major industrial and mobile sources. Homes within the study will be selected to evaluate the impact of these sources on exposures and to determine high-end exposure. These data will be used to further evaluate and refine human exposure models that characterize the magnitude of exposure along its uncertainty and variability. In addition, the methods developed and applied in this study can be used as a prototype for other community based air toxic programs.

1.2 Recent Literature

1.2.1 Studies Linking Exposure to Sources

Recent studies have made progress in relating air pollution to sources and the role individual sources have had upon estimating potential human exposures. Some of these studies have gone beyond central site-only measurements to include residential and/ or personal exposures. A number of the most recent studies have been summarized in detail in Appendix A and will not be presented here. However, a summary of some of the most important findings is appropriate and will be presented to provide insight as to the current state-of-the-science in this area and how the proposed study would relate to past research.

In brief, many of these studies have attempted to understand the role that mobile source emissions from local automotive traffic have had upon personal and residential concentrations (Van Vliet et al., 1997, Roorda-Knape et al., 1998, Fischer et al., 2000, Kingham et al., 2000, and Janssen et al., 2001). PM mass concentrations in addition to mobile source-related VOCs and gases were typically studied. In a number of instances, these studies revealed that distance from the roadway itself (proximity) was shown to be a major contributing factor in establishing a source association. One study indicated that the source influence on air concentrations decreased rapidly when the residential setting was more than 50 meters from the roadway. The selection of pollutants to be measured was also observed to be very important. The strength of the central site-residential, central site-personal correlations associated with mobile source pollutants was determined to be highly variable among the metrics in many instances. These latter two points indicate that even where specific source-derived ambient pollutants exist, their spatial variability across a metropolitan area might be sufficiently high as to result in a very high level of uncertainty associated with estimating their concentrations based upon a single measurement location. This might be especially important if a single central site monitor was used as a surrogate for true personal exposures for all individuals living in a large metropolitan area where traffic patterns vary.

In addition to the exposure research described above, a number of studies (Van Vliet et al., 1997, Laden et al., 2000, and Hoek et al., 2002,) have attempted to determine the role of sources, including that related to mobile sources, upon epidemiological findings. For example, Laden et al., (2000) were able to use a specialized factor analysis to resolve the major components of ambient particulate matter and the impact these sources had upon mortality in six U.S. cities. They discovered that elements associated with resuspended soil, motor vehicle emissions, and coal combustion has the strongest assocations with the observed health effect. Both Van Vliet et al. (1997) and Hoek et al. (2002) found that select exposure measures related to mobile sources (e.g., black smoke) as well as cohort proximity to roadways were important variables in associating the incidence of asthma among the local population.

A recently completed investigation of the PM and air toxics associated with vehicle interiors, roadways and central sites observed a wide range of variability between pollutant concentrations (Riediker et al., in press). In-vehicle concentrations were usually

greater than roadside and central site concentrations for certain mobile source-related air pollutants, such as VOCs, carbonyls, metals, and carbon monoxide. This variability may be an important factor for individuals spending extensive periods of time involved in vehicle travel and any resulting health effects.

While numerous studies have attempted to investigate the role of location between personal, residential and central site PM measurements, few studies have attempted to relate non-occupational personal exposures directly to ambient sources across a wide metropolitan area. Studies attempting the simpler case include the longitudinal human exposure panel studies recently performed by the U.S. EPA and collaborating university groups (U.S. EPA, 2002; Liu et al., in press). These studies have shown that the contribution of ambient-derived $PM_{2.5}$ mass to one's total personal exposure falls into a range of ~45-60%. While defining the role, magnitude and variability of individual ambient sources upon total personal exposure was not a primary goal of the cited studies, additional source apportionment work such as that reported by Hopke et al., (in press) is being performed where possible. Data from these secondary efforts might be used as an aid in the final design of this proposed study. In addition to the above cited U.S. studies, the EXPOLIS study performed throughout Europe (Edwards et al., 2001, Kousa et al., 2002) also focused primarily upon the basic PM mass concentrations relationships. Additional European studies having a similar study design have been reported by Van der Zee et al. (1998), Houthuijs et al. (2001), Ruuskanen et al. (2001), and Hoek et al. (2002).

The literature indicated that no substantial or definitive studies have been performed that have attempted to determine the role of numerous ambient sources upon total personal exposure or residential settings. This has undoubtedly been due to the lack of appropriate source apportionment tools, the means to adequately measure source markers at the central site, residential and potentially the personal setting, and the need to integrate such an effort into a multi-component human exposure study.

1.2.2 Source Apportionment Studies

Recent source apportionment studies for PM_{10} and $PM_{2.5}$ suggest that both mobile and stationary sources are responsible for substantial contributions to the ambient PM measured in urban and rural locations of the U.S. (Table 1-1). Many previous studies have focused on elemental analysis. These results were often readily available from inexpensive XRF analysis of filter samples. Combined with information gathered from direct source signatures (such as that obtained from stack monitoring), and/or central site monitoring, source apportionment using receptor modeling was performed. Particulate matter resulting from the combustion of leaded gasoline often yielded a unique means of apportioning this source. Unfortunately, a specific marker for automotive emissions currently does not exist. It is now recognized that the inclusion of organic components into the analysis is needed to adequately define the source (Schauer et al., 1996; Schauer and Cass, 2000). These efforts have typically focused upon the use of particle-phase organics such as straight-chained hydrocarbons, polynuclear aromatic hydrocarbons, carboxylic acids, hopanes, and sugars as source markers. In some instances, discrete

markers such as levoglucosan have been used to uniquely identify the contribution of sources such as woodsmoke.

More recently, the Multilinear Engine (ME) and Positive Matrix Factorization (PMF) source apportionment tools have been used (Paatero et al., 1999, Hopke et al., in press). These approaches use a wide number of exposure variables in assigning source contributions and can easily incorporate data on elemental, organic concentrations, and a large number of other exposure variable data into their models. Review of concentrations in the literature cited in Table 1-1 indicate that while a large number of ambient source apportionment studies have been performed, few data have been reported involving residential and especially personal-based measures. Özkaynak et al. (1996) was the first to attempt such an apportionment using mostly elemental mass concentrations from residential and ambient-based measurements collected in the PTEAM study. Even so, a large concentration of the total PM mass could not be accounted for in the apportionment. Work proposed in the DEARS would represent a major contribution to the science in that data from a human exposure population would be integrated with extensive residential (indoor, outdoor) and central site data. Inputs into the source apportionment modeling would include central site, residential and potentially even personal concentrations of PM mass, VOCs, SVOCs, carbonyls, sulfates, elements, carbon and criteria gases. State-of-the-science source apportionment tools, extensively advanced since the PTEAM study, should enable a large part of the mass to be attributed to specific sources.

Table 1-1 Source Apportionment Study Results

Sampling Location	Time Period	Percent Contribution								$PM_{2.5}$ Conc
		Diesel	Gasoline	Total Mobile Sources	Road Dust/Soil	Biomass Burning	Secondary Sulfate	Secondary Nitrate	Misc. Sources	
Pasadena, CA (Schauer, 1996)	1982	18.8	5.7	24.5	12.4	9.6	20.9	7.4	24.1	28.2
Los Angeles, CA (Schauer, 1996)	1982	35.7	6.5	42.2	11.1	5.8	20.3	9.2	18.9	32.5
West Los Angeles, CA (Schauer, 1996)	1982	18.0	5.7	23.7	12.2	11.0	24.1	7.8	23.3	24.5
Rubidooux, CA (Schauer, 1996)	1982	12.8	0.7	13.5	13.1	1.2	13.8	24.7	21.6	42.1
Bakersfield, CA (Schauer, 2000)	1995	9.5	3.5	13.0	1.5	16.9	5.0	29.2	25.8	53.8
Fresno, CA (Schauer, 2000)	1995	9.7	2.5	12.2	1.8	49.5	3.5	25.7	19.3	65.9
Sacramento, CA (Motallebi, 1999)	Winter, 1991-96	---	---	24.5	1.2	18.1	4.5	36.6	---	39.5
Bakersfield, CA (Magliano, 1998)	Winter, 1996	---	---	16	<3	20	7	34	---	52
Fresno, CA (Magliano, 1998)	Winter, 1996	---	---	13	<3	19	5	32	---	63
Philadelphia, PA (Dzubay, 1988)	Summer, 1982	---	---	8.5	4.4	---	81.9	---	4.5	27.0
Camden, NJ (Dzubay, 1988)	Summer, 1982	---	---	9.2	3.2	---	81.3	0.4	5.7	28.3
Clarksboro, NJ (Dzubay, 1988)	Summer, 1982	---	---	5.8	2.7	---	84.6	---	2.7	26.0
Welby, CO (Lawson, 1998)	Winter, 1997	10	28	38	16	5	10	25	6	no data
Brighton, CO (Lawson, 1998)	Winter, 1997	10	26	36	11	2	15	32	4	no data
Reno, NV (Gillies, 2000)	Summer, 1998	---	---	68	14.5	4	11	2	0.6	7.8
Phoenix, AZ (Ramadan, 2000)	Summer, 1995-98	10.9	36.2	47.1	1.8	15.0	---	---	36.0	8.3
Phoenix, AZ (Ramadan, 2000)	Winter, 1995-98	14.5	38.9	53.4	1.1	8.9	---	---	36.8	13.8
Baltimore, MD (Hopke, 2003)	Summer, 1998	--	--	16.8	--	--	53.7	<1%	--	22.0
Steubenville, OH (Laden et al., 2000)	1979-1988	--	5.0	--	14.0	--	57.3	--	23.7	30.5

2.0 STUDY OBJECTIVES AND OVERVIEW

2.1 Objectives

The overall goal of NERL's Exposure Research Program is to develop the data and models that characterize and predict human exposure. The Detroit Exposure Aerosol Research Study (DEARS) is an important step in this program. The study builds upon the results from previous longitudinal panel studies with several very important differences. The DEARS will examine the spatial variability of $PM_{2.5}$ and its components to determine the suitability of conducting health outcome studies using a central site monitor in an urban area like Detroit where there are many point sources. Source apportionment techniques will be used to evaluate the relationship for PM and air toxics from specific sources. Finally, the study is designed to look at and quantify the impact of local ambient sources on the relationship between central site monitors and exposures. Results from this study will be critical in providing exposure data for developing future standards.

Six objectives have been defined for this study.

(1) To determine the associations between concentrations measured at central site monitors and outdoor residential, indoor residential and personal exposures for selected air toxics, PM constituents, and PM from specific sources.
(2) To describe the physical and chemical factors that affect the relationship between central site monitors and outdoor residential and indoor residential concentrations, including those that affect ambient source impacts.
(3) To identify the human activity factors that influence personal exposures to selected PM constituents and air toxics.
(4) To improve and evaluate models used to characterize and estimate residential concentrations of and human exposures to selected air toxics, PM constituents, and PM from specific sources.
(5) To investigate and apply source apportionment models to evaluate the relationships for PM from specific sources and to determine the contribution of specific ambient sources to residential concentrations and personal exposures to PM constituents and air toxics.
(6) To determine the associations between ambient concentrations of criteria gases (O_3, NO_2, and SO_2) and personal exposures for these gases as well as personal exposures to air toxics, PM constituents, and PM from specific sources.

Achieving the first objective will establish the longitudinal correlations between measurements performed at an central site monitor and residential outdoor/indoor/personal exposure concentrations. Results will be used to determine if central site measurements for air toxics and PM species can be used as exposure surrogates in community-based epidemiological studies. As part of this objective, we will determine if proximity to mobile or stationary sources has an impact on these relationships. Several design elements are required to meet this objective. Concurrent monitoring for all targeted pollutants must be conducted at the central monitoring site, outdoors and indoors at the residence, and on the person. Samples must be collected over multiple days in order to evaluate longitudinal correlations. The central monitoring site must have similar requirements for placement as monitors that would be used for epidemiological

research (i.e., as the speciation sites in NCore or at Supersites). Finally, residences need to be selected based on proximity to sources.

Under the second objective, we will evaluate and describe the physical and chemical factors that determine the impact of various ambient sources on outdoor and indoor residential concentrations and the spatial variability of these concentrations in relation to the source locations. Results will be used to improve the algorithms in exposure models and to develop input data for these models. The study design must be directed toward collecting data at central site, outdoor residential and indoor locations that will describe the most important factors. For outdoor residential concentrations, this will include location of the residence in relation to the central site monitor, location of the residence in relation to mobile or stationary sources, composition and strength of source emissions, and meteorology. For indoor residential concentrations this includes collecting data on indoor and outdoor concentrations, air exchange rates, housing characteristics, HVAC operations, and indoor source use. These data when collected over multiple days will be used to calculate infiltration factors, penetration rates and removal rates for different chemicals and different housing conditions. Monitoring will be conducted during both a winter and a summer season for each participant to evaluate the impact of climate on various factors.

Under the third objective, we will identify factors that influence personal exposures to selected PM constituents and air toxics using the personal monitoring data and detailed time-activity information. The impact of personal activities and the time spent in various locations (residential, non-residential, in vehicles) on personal exposures will be evaluated. Real time measurements for $PM_{2.5}$ will be made using personal nephelometers. These data will be used to determine the impact of spending time in nonresidential locations and personal activities on exposure. This will be critical in understanding the importance of commuting and work place activities on exposures.

For the fourth objective, the measurement data and results of the data analysis will be used to improve the inputs and algorithms used in exposure models. The residential indoor and outdoor measurement data collected during the study, and the important factors identified by the data analysis conducted for the second objective, will provide critical information for improving the inputs and algorithms used in population exposure models for PM constituents and air toxics. A variety of physically- based mechanistic and stochastic modeling tools will be applied in order to quantify predicted impacts of major pollutant source categories on outdoor and indoor concentrations, and personal exposures to $PM_{2.5}$, PM constituents (e.g., SVOCs, EC/OC) and air toxics (e.g., VOCs) in the Detroit area. The principal modeling tools that will be chosen for this specific application, are: the MENTOR (Modeling Environment for Total Risk Studies) system, and the SHEDS (Stochastic Human Exposure and Dose Simulation) model. The MENTOR/SHEDS modeling system will be modified and applied to the Detroit study area. The primary objective of the MENTOR/SHEDS applications will be to test the modeling tools developed using the DEARS monitoring data in order to evaluate the performance of the model and perform appropriate model refinements. A secondary objective of these model applications will be to estimate source-specific contributions of PM and air toxics emissions to outdoor and indoor concentrations and personal exposures in the Detroit area.

Under the fifth objective, we will refine and apply source apportionment techniques to understand exposure relationships for PM and air toxics from specific ambient sources. This has been included as a separate objective since it requires state-of-the-art monitoring for marker compounds. In addition, the extensive use of source apportionment techniques in personal exposure studies is a new and challenging application. To meet this objective, source apportionment models will be used to estimate PM concentrations from specific sources in central site, residential outdoor and residential indoor samples. These results will then be used to evaluate and quantify the relationship between central site monitors and residential indoor and outdoor concentrations for PM from specific sources. Monitoring requirements to meet this objective are large. Source apportionment models require concentration data on a large number of marker compounds including elements, elemental carbon, criteria gases, carbonyls, and semivolatile organic compounds (SVOCs). Concentrations of marker species will be measured in central site and residential samples. The feasibility of collecting these data in personal exposure samples and thus extending source apportion methods to personal exposure will also be evaluated. Source profiles that accurately represent the sources in the study area would greatly enhance the models. Although not in the current design, opportunities to generate source data are being investigated.

The sixth objective is intended to determine if central site concentrations of gaseous co-pollutants (O_3, NO_2 and SO_2) are surrogates or confounders of exposures to air toxics, PM constituents and PM from specific sources in epidemiological studies. Previous studies showed that the gaseous co-pollutants acted as surrogates of personal exposures to $PM_{2.5}$ and sulfate (Sarnat et al. 2001). Statistical analyses similar to that used by Sarnat et al. will be used to determine if similar relationships exist between exposures to the gaseous co-pollutants and air toxics, PM constituents and PM from specific sources.

2.2 Study Overview

The proposed study is a three-year field monitoring study that will be conducted in Detroit, Michigan and is designed to measure exposure and describe exposure relationships for air toxics, PM components, PM from specific sources, and criteria pollutants. Monitoring will be conducted at 120 residences over a three-year period. Measurements of air toxics, PM, PM constituents, and criteria gases will be collected in each home and from one participant in each home for five days during both a winter and summer season for a total of 1200 household-person/days of measurements. A combination of both weekday and weekend sampling will be conducted in order to evaluate expected variations in industrial source emissions, traffic volumes, and personal activities. Monitoring is anticipated to start in the summer of 2004.

The residence will be the primary unit for selection and monitoring. The sampling approach will select households within census tracts that are in close proximity to point and/or mobile sources in addition to those that are further removed from sources. Census tracts will be further subdivided into census blocks to contain approximately 50 households. A single participant in each home will be selected for personal monitoring and to provide time/activity information. The central site monitoring is located at Allen Park and is part of the Speciation Trends Network. To further investigate the distribution of pollutants across the study area, one residential outdoor

location will be selected each week to serves as a secondary community site. To achieve this, a dichotomous sampler will be added to the normal residential outdoor monitoring scheme for a selected home. These data will be used to determine how spatially representative the primary and secondary community measures (Allen Park and the selected residential outdoor sites) are relative to all other outdoor locations.

Measurements will include personal, residential indoor, residential outdoor, and central site monitoring for $PM_{2.5,}$ VOCs and carbonyls. All $PM_{2.5}$ filters will be analyzed for mass, elemental carbon (EC), selected elements, and sulfate as sulfur. Since participants cannot carry a large number of personal monitors, some pollutants will only be measured indoors, outdoors, and at the central monitoring site. These include PM_{coarse}, nitrates, selected SVOCs, and EC/OC as collected on quartz filters. The criteria gases will only be measured in central site and personal exposure samples. Survey information will be collected on household characteristics, HVAC operation, local ambient sources, indoor and personal sources, and time activity patterns.

Data analysis will include statistical analysis to specifically address the study objectives. Human exposure models and source apportionment models will also be applied to the data. These analyses will be used to complete annual performance measures set forth in the PM and air toxics multi year plan.

2.3 Collaborations and Partnerships

The goals in this study are the research needs of the NERL's Human Exposure program. Therefore, the data measurements and resulting modeling efforts target these needs. There are however, opportunities for the measurements performed in this work to support other research areas. The EPA's National Health and Environmental Effects Laboratory (NHEERL) currently has a planned health study involving asthmatic children in the Detroit-Windsor, Canada metropolitan area in 2005-2006. While the proposed NHEERL study (the Detroit Children's Health Study) does not involve any direct human exposure measurements and there is no planned overlap between subjects participating in the two studies, ambient and local PM and air toxics measurements performed in the DEARS would serve as inputs into the resulting epidemiological calculations. This would have potential benefits of reducing the uncertainties in exposure estimates relative to those based on ambient pollutant concentrations.

In addition to the use of the data described above, there is the potential for the NERL to collect criteria gas pollutant and VOC data at representative schools associated with the Detroit Children's Health Study. These samples will be collected in accordance with the study design currently being developed for the Detroit Children's Health Study and the DEARS.

The daily collection of large quantities of size-fractionated PM from the Detroit metropolitan area is also of interest to NHEERL researchers. Collection of milligram quantities of PM mass are needed for toxicological studies. These studies would assess the chemical and toxicological properities of the various PM size fractions as they relate to potential emission sources. There is also a possibility that a study funded by the Electric Power Research Institute (EPRI) that follows the outlines of an earlier EPRI study in Atlanta called ARIES may take place in Detroit. Part of this study involves an exposure study to be carried out by the Harvard School of Public

Health (Petros Koutrakis, PI). This exposure study would be a good complement to our study since it would consist partly of "scripted activities," possibly including such activities as commuting during rush hour. This would provide valuable information on a microenvironment of interest to the Office of Transportation Air Quality (OTAQ) and to exposure modelers. Once again, central site data collected in the DEARS would be shared with these collaborators to meet their research data needs and thereby saving valuable research funding (permitting a larger scripted population size to be incorporated into the EPRI study). The ARIES-type study design has been highly touted by OMB as an important study in the determination of source-based influence upon observed human health effects.

Environment Canada and Health Canada are currently considering research efforts to leverage those proposed in the DEARS. Limited information is available about any potential collaboration but it is one in which the NERL will thoroughly consider following its development. One of preliminary consideration might involve the use of an Environment Canada's mobile laboratory (van) to help determine concentrations of select pollutants in Wayne County neighborhoods being considered for DEARS subject recruitment. Data would help determine the magnitude of pollutant concentrations in neighborhoods being considered and sources potentially influencing these neighborhoods. These vans have the capability of monitoring various PM size fractions, VOC concentrations and other air toxics. The NERL would propose providing technical assistance to any proposed Environment Canada and Health Canada personal and residential field studies conducted across the border in Windsor, Canada. In addition, data from the DEARS and Canadian studies would be integrated as appropriate to provide the basis for an investigation of transborder (U.S.-Canadian) issues.

3.0 SURVEY DESIGN

3.1 Overview

The proposed study is a residential and personal exposure field monitoring study. The primary goal of the study is to evaluate and describe the relationship between air toxics and PM constituents measured at a central site monitor and measurements of residential and personal concentrations. An emphasis is placed on understanding the impact of local sources (point and mobile) on outdoor residential concentrations and the impact of housing type and house operation on indoor concentrations. Personal monitoring will be conducted to determine the impact of time spent in nonresidential locations and personal activities on exposure. Given this emphasis, the residence will be the primary unit for selection and monitoring. A sampling approach will be taken to select households within census tracts, in close proximity to point and/or mobile sources and those that are further removed from sources. Census tracts will be further subdivided into census blocks to contain approximately 50 households. Personal exposure data and time activity information will be collected on one participant in each home. A single central site monitor will be used throughout the study to measure concentrations of PM constituents and air toxics similar to those measured at the residential and personal level. The central monitoring location will be selected to mimic community-based monitors that are used in epidemiological studies. Ideally, the central site monitor should be representative of the airshed.

This section provides general information on the study area. Details are then provided on selection of housing units, individual participants, and the central site monitor. Finally, the approach for recruiting study participants is given.

3.2 Study Area

3.2.1 Detroit Selection

Detroit, Michigan was considered the best candidate for this study because of its current and projected future non-attainment status, the number of point and mobile source influences present, its geographic location, meteorology, ambient monitoring networks, potential state and local collaborations, and community-based partnerships. Most importantly, Detroit is currently in non-attainment status for $PM_{2.5}$, and is also projected to be in non-attainment status after sulfur reductions in 2010 (Clear Skies initiative). There are a large number of industrial point sources of PM and air toxics in the Detroit area, including coke ovens, iron/steel manufacturing, coal-fired power plants, sewage sludge incineration, automotive industry, refineries, and chemical plants. The border crossing between Windsor, Canada and Detroit via the Ambassador Bridge also provides a large diesel and automotive source from idling motor vehicles. There are 4 major interstates and many heavily traveled roadways, which will serve as line sources of vehicle emissions in Detroit and surrounding Wayne County. Historical PM and traffic count data are also available from the area.

The prevailing wind direction (southwest) indicates there should be considerable spatial variation across the Detroit metropolitan area given the industrial sources located along the eastern edge of Wayne County. Spatial variation in PM and air toxics concentrations will aid in source apportionment and modeling. Located in the mid-west, the seasonal fluctuations in temperature and weather patterns make Detroit a better candidate than a southern city with less of a seasonal component. There are a number of central monitoring sites currently running in the Detroit area including a National Air Toxics Trends Site, a Speciation Trends Network site, and a number of monitoring sites operated by the state of Michigan. The data from these sites will be available during the study period and we will be able to run additional monitors at an existing site to better suit our sampling needs.

Recent studies in the area have used a community-based participatory research approach which has involved the local community in studying 300 school aged children with asthma (Community Action Against Asthma, CAAA). Working with community partnerships that are already in place will enhance our ability to recruit and retain participants in a three-year study. The potential for collaboration with the state of Michigan, Lake Michigan Air Directors Consortium (LADCO), and local universities also make Detroit a good study location. A number of other cities were considered for this study, including Houston, TX, Tampa, FL, St. Louis, MO, and Chicago, IL.

3.2.2 General Information and Demographics

Wayne County is located in southeastern Michigan approximately 60 miles north of the Michigan/Ohio border (Figure 3-1). It is located on the western edge of Lake St. Clair and separated from Windsor, Canada by the Detroit River. While the Detroit metropolitan statistical area (MSA) has a population of 4.4 million and includes six counties (Wayne, Oakland, Macomb, Monroe, Lapeer, and St. Clair), nearly half (2.1 million) reside in Wayne county alone.

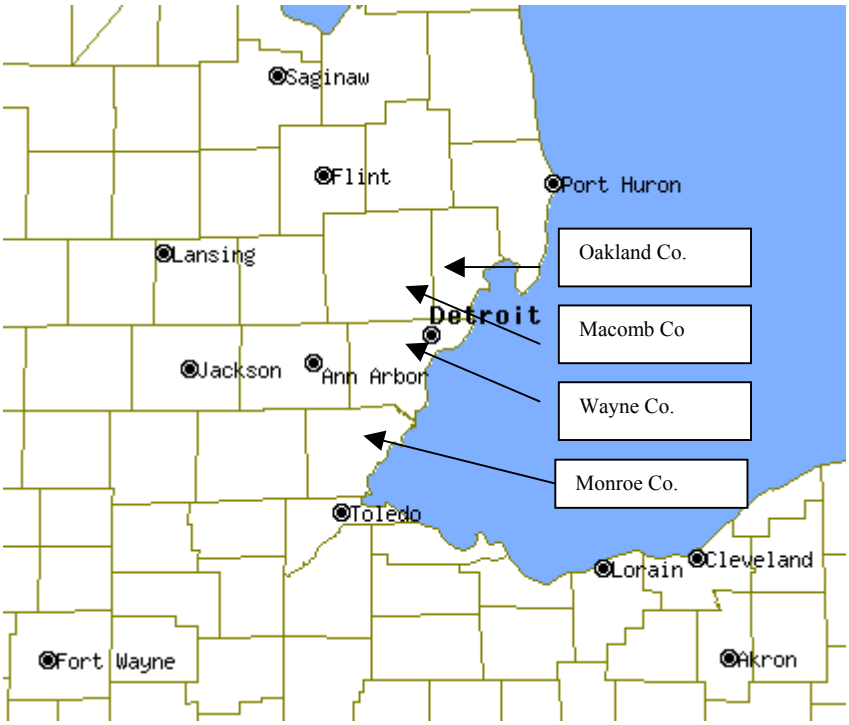

Figure 3-1. Map of southeastern Michigan/northern Ohio.

Wayne County possesses a diversity of ethnicities and housing stock, although their distribution throughout the County is not homogeneous. Based on data from the 2000 U.S. Census, Wayne County is approximately 52% White, 42% Black or African-American and 6% other ethnicities. Hispanics or Latinos comprise only 3.7% of Wayne County. The largest concentrations of African-Americans and Hispanics in Wayne County are located in Detroit (Figure 3-1). African-Americans constitute approximately 82% of Detroit's 950,000 people and are present in greatest numbers in east, west-central and southwest Detroit (>95%). There are other areas of Wayne County that have a high concentration of African-Americans, namely Inkster located in central Wayne County as well as portions of southwest Wayne County. Hispanics are primarily located in southwest Detroit (>40%; Figure 3-1), but only constitute about 5% of Detroit's population. Whites are primarily located in areas outside of Detroit, namely the entire western portion and the northeastern tip of Wayne County, in varying degrees (Figure 3-2). The Asian population is

scattered throughout Wayne County with pockets of concentration in Hamtramck, Dearborn and Plymouth (Figure 3-2).

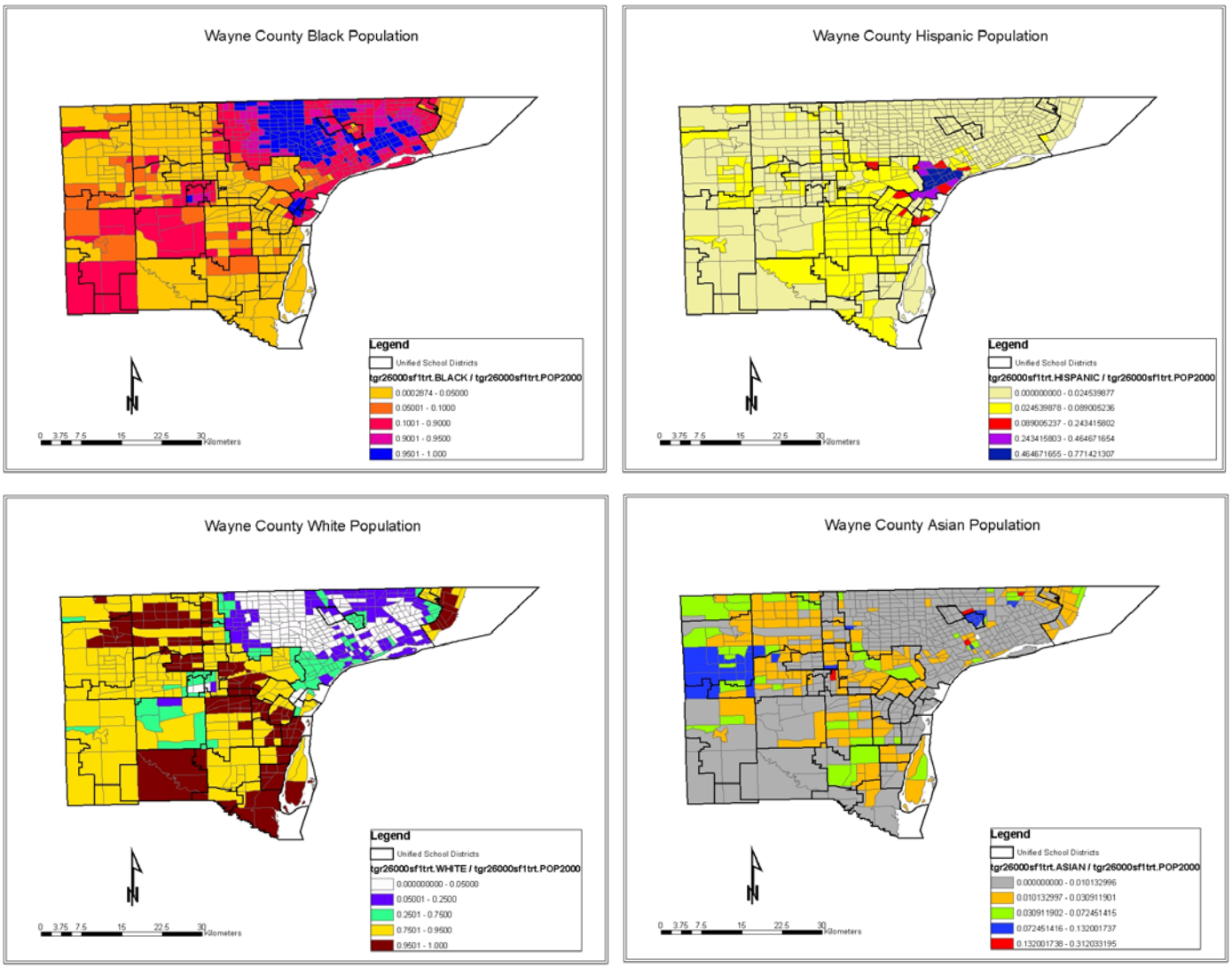

Figure 3-2. Percentage of Black, Hispanic, White and Asian people by census tract in Wayne County.

Housing stock in Wayne County is also not spatially homogeneous with older residences located primarily in Detroit and northeastern Wayne County, and newer construction in the remainder of Wayne County (Figure 3-3). There are, however, isolated census tracts within Detroit and Dearborn that contain a higher percentage of newer construction. In Wayne County there are a total of 826,145 housing units of which 73% are detached or attached single-family homes, 16% are smaller multi-family structures, and 11% are structures with 10 or more housing units. Of all the housing units in Wayne County 62% are owner occupied while 38% are renter occupied.

Owner occupied units had a median of 6.0 rooms per household and a median household size of 2.74 persons while renter occupied units had 4.4 rooms and 2.43 persons.

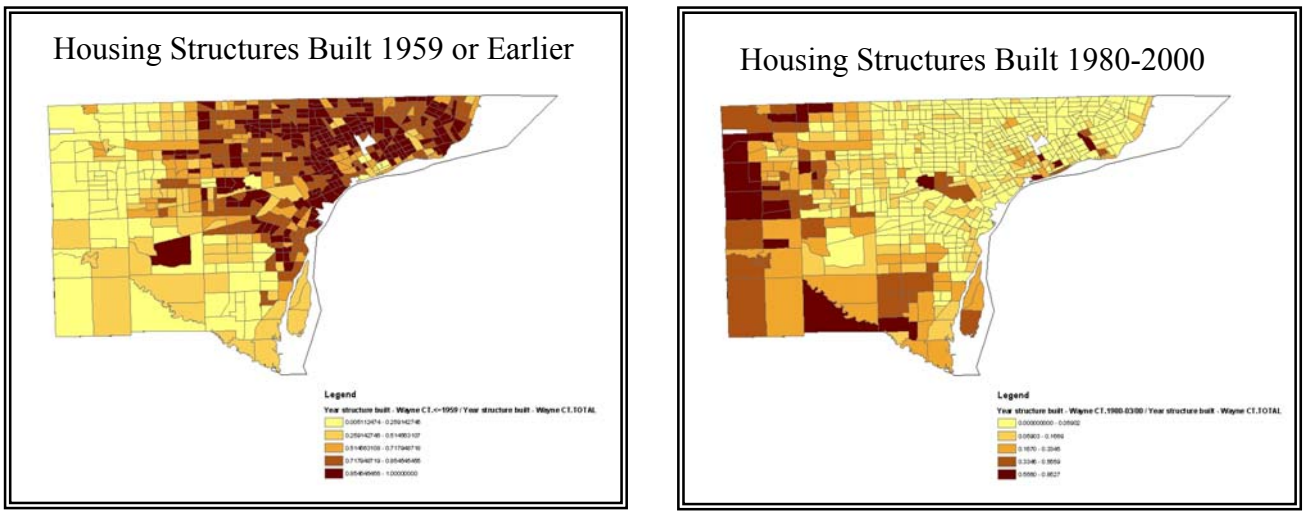

Figure 3-3. Distribution of housing stock in Wayne County by census tract.

Home heating fuel in Wayne County is predominately gas, however, owner occupied units rely more heavily on gas (97%) than do renter occupied units (85%). Electricity follows for home heating fuel use with owners using it in only 2% of units while renters use electricity for heating in nearly 13% of units.

3.2.3 Sources of PM and Air Toxics

There are numerous sources of PM and air toxics in the Wayne County airshed. Stationary sources are located primarily in eastern Wayne County throughout Detroit (Figure 3-4). The heaviest industry is located in southwest Detroit in and around Zug Island, an industrial complex on the Detroit River. Industrial sources here include iron/steel manufacturing, coke ovens, oil refineries, sewage sludge incineration and coal-fired utilities. There are also a number of major interstates and state highways in Wayne County, which converge most notably in southwest Detroit (Figure 3-4). This is also the location of the Ambassador Bridge where nearly 10,000 diesel trucks cross each day. Since 9/11, trucks idle at the border crossing for several hours waiting for inspections and approval to enter/exit the U.S. Still, high traffic volumes are experienced in western Wayne County as well along two east-west corridors (I-96 to the north and I-94 to the south) and one north-south corridor (I-275).

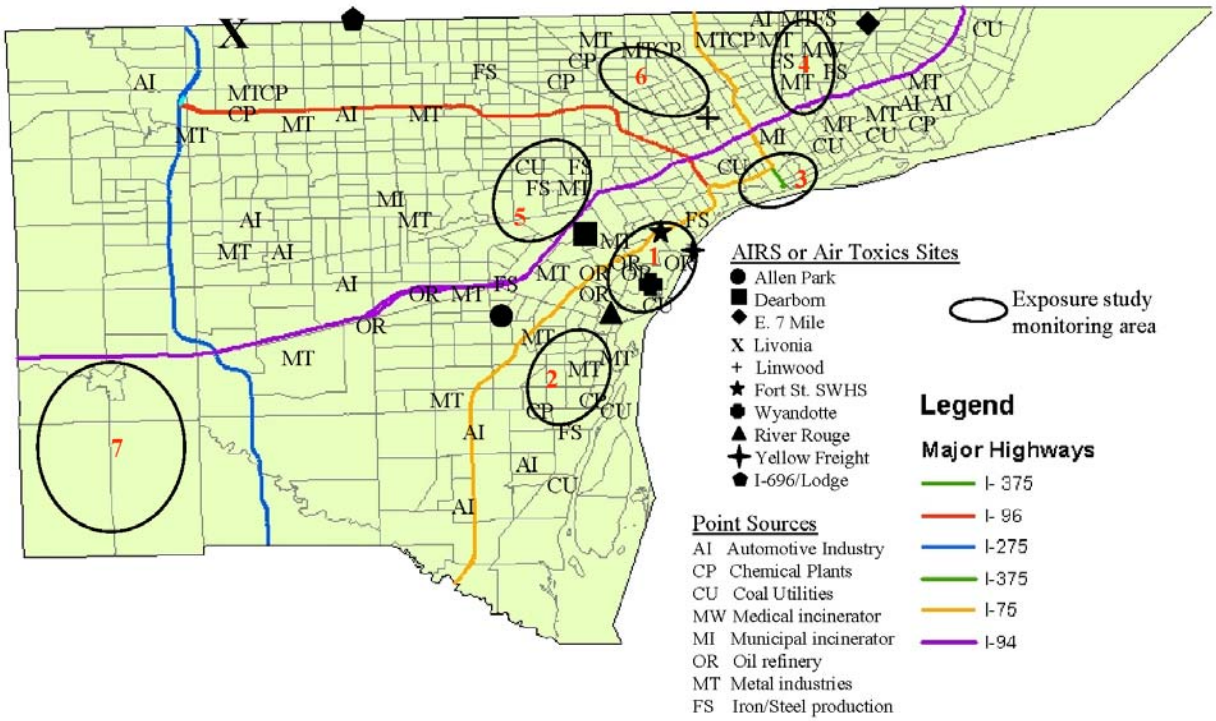

Figure 3-4. Map of Wayne County census tracts showing location of interstates, select industrial point sources and air toxics monitoring sites. Also shown are areas where study participants will be selected for exposure monitoring.

3.2.3.1 Stationary Point Sources of PM and Air Toxics

An analysis of stationary source emissions in the Detroit area was conducted to identify the important sources and source categories for PM and air toxics. There are a number of large PM and air toxics sources outside of Wayne County in the counties of Monroe, Macomb, St. Clair, Oakland, and Washtenaw. PM and air toxics emissions from some sources in adjacent counties may be high enough to impact residential areas in Wayne county, depending on wind direction and other meteorological factors. Emissions data for the six counties in the Detroit area were obtained from the National Emissions Inventory database for 1996 (PM and air toxics) and 1999 (PM only). Air toxics data for 1999 have not been finalized in the NEI database at this time, but are expected to be released during summer 2003. Source emissions data were available by source within each county, so we were able to identify specific sources with large emissions, as well as an inventory for the entire county.

The industrial sectors with the highest $PM_{2.5}$ emissions in the Detroit area are steel production, power plants, and cement/concrete production. Table 3-1 shows annual $PM_{2.5}$ emissions for the largest sources in the Detroit area during 1999 and 1996. The relative emissions of a particular source or source category varied between years probably due to a combination of economic conditions and improved control technology. In 1996 the highest $PM_{2.5}$ emissions were from two industrial sectors; power plants and steel production. The situation was different in 1999 as $PM_{2.5}$ emissions from power plants and steel production decreased relative to other sources including cement/concrete production and glass manufacturing. In 1999, several new casinos were being

built in Detroit, likely resulting in increased production of cement/concrete and glass manufacturing and consequently higher emissions.

County-wide $PM_{2.5}$ emissions inventories show that Wayne County emissions were about 2.3 times higher than Monroe county in 1996, but nearly identical in 1999 (Table 3-2). In general, county-wide emissions varied little between 1996 and 1999 except Oakland County which increased and Wayne and St. Clair counties which decreased between 1996 and 1999.

Major stationary (point) sources of air toxics in the Detroit area have been identified from the 1996 National Toxics Inventories (http://www.epa.gov/air/data/ntidb.html). Table 3-3 shows some of the largest point sources of selected air toxics in the Detroit MSA. The air toxics used to evaluate sources in Detroit were chosen to reflect the types of air toxics that will be measured in this study and included representative compounds from each of the major categories including VOCs (benzene and 1,3-butadiene), aldehydes (formaldehyde, acetaldehyde and acrolein), metals (Ni, Mn, Cr, As, Se and Pb) and PAHs (benzo(a)pyrene and benzo(g,h,i)perylene). For air toxics, the industrial sectors with the highest emissions in the Detroit area are generally power plants, iron/steel production and oil refinement, depending on the air toxic. In contrast to $PM_{2.5}$ emissions, which were highest in Wayne County, emissions of air toxics were generally highest at facilities in the outlying counties. Exceptions to this are for manganese (iron/steel), nickel (oil refinery), benzene (steel production and oil refinery) and 1,3-butadiene (steel production). Most of the area's power plants, which are the largest sources for several air toxics, are located in counties adjacent to Wayne County. The power plant in Monroe County is the largest source of many air toxics for the Detroit MSA, but, combined, the Wayne County power plants are significant sources for several air toxics and are closer to the study population than the Monroe power plant (about 60 km from Detroit). Similarly, there are significant sources for all of the air toxics in Wayne County.

Table 3-1. Largest stationary sources of $PM_{2.5}$ emissions (metric tons) in the Detroit area

Facility	Sector	Location (County)	$PM_{2.5}$ Emissions	
			1999	1996
Power				
J.R. Whiting	Power	Monroe	251	105
Detroit Edison	Power	Monroe	142	890
Trenton Channel	Power	Wayne	86	205
Detroit Edison - Greenwood	Power	St Clair	84	302
Belle River Power Plant	Power	St. Clair	48	303
River Rouge Power Plant	Power	Wayne	47	157
Steel				
National Steel	Steel	Wayne	519	761
Rouge Steel	Steel	Wayne	168	751
DSC LtD.	Steel	Wayne	<27	240
Oil				
Marathon Ashland	Oil	Wayne	163	80
Materials				
Holnam, Inc.	Cement	Monroe	552	36
Guardian Industries Corp	Glass	Monroe	301	224
Angelos Inc	Concrete	Oakland	279	<26
New Haven Foundry	Metals	Macomb	198	212

Table 3-2. County-wide $PM_{2.5}$ Emissions Estimates in 1996 and 1999 for Detroit MSA counties (metric tons).

County	1999	1996
Wayne	1378	2753
Monroe	1211	1187
Oakland	467	103
Macomb	288	302
St. Clair	260	623
Washtenaw	97	84

Table 3-3. Selected stationary sources for hazardous air pollutants in the Detroit area (pounds). * Represents the sum of all power plants located in Wayne County (Trenton Channel, River Rouge, Wyandotte and Mister Sky)

Facility	Sector	Location	Emissions	Amount
National Steel	Steel	Wayne	Benzene	140,863
			Coke Oven	80,596
			Manganese	10,400
BASF	Chemical	Wayne	Formaldehyde	24,450
Marathon Oil	Oil	Wayne	Nickel	4,240
			Chromium	1,499
			Benzene	59,065
Detroit Edison	Power	Monroe	Benzene	12,175
			Formaldehyde	24,771
			Acetaldehyde	5,338
			Manganese	4,592
			Nickel	2,624
			Chromium	2,436
			Arsenic	3,842
			Selenium	12,182
			Lead	3,938
Hancock Peaking Units	Power	Oakland	Formaldehyde	24,584
Chrysler Warren Trucks	Automotive	Macomb	Formaldehyde	15,781
Allied Signal	Tar	Wayne	Benzene	17,933
Wayne Co. Power Plants*	Power	Wayne	Benzene	4,584
			Formaldehyde	1,457
			Acetaldehyde	2,006
			Manganese	1,766
			Nickel	2,128
			Chromium	927
			Arsenic	1,462
			Selenium	4,588
			Lead	1,501

3.2.3.2 Mobile Sources

In Wayne County, the major highway corridors include Interstates 75, 94, 96, and 275 (Figures 3-4 and 3-5). As an example of traffic flows, Interstate 75 has traffic segments in Wayne County with 24-hour annual average vehicle counts that range from 63,900 to 195,000 vehicles per day (Table 3-4). Roadways in Detroit with one-way traffic volume >50,000 vehicles/day are shown in Figure 3-5.

Table 3-4. Traffic Volumes on Wayne County Highways

Traffic volumes (24-hour annual averages in vehicles per day); Wayne County, MI, 2001		
	Minimum Segment	Maximum Segment
Interstate 75	63,900	195,000
Interstate 94	86,200	148,000
Interstate 96	18,600	120,000
Interstate 275	18,100	34,600
U.S. Route 24	14,400	78,400
State Route 1	18,800	23,700
State Route 3	14,600	32,700

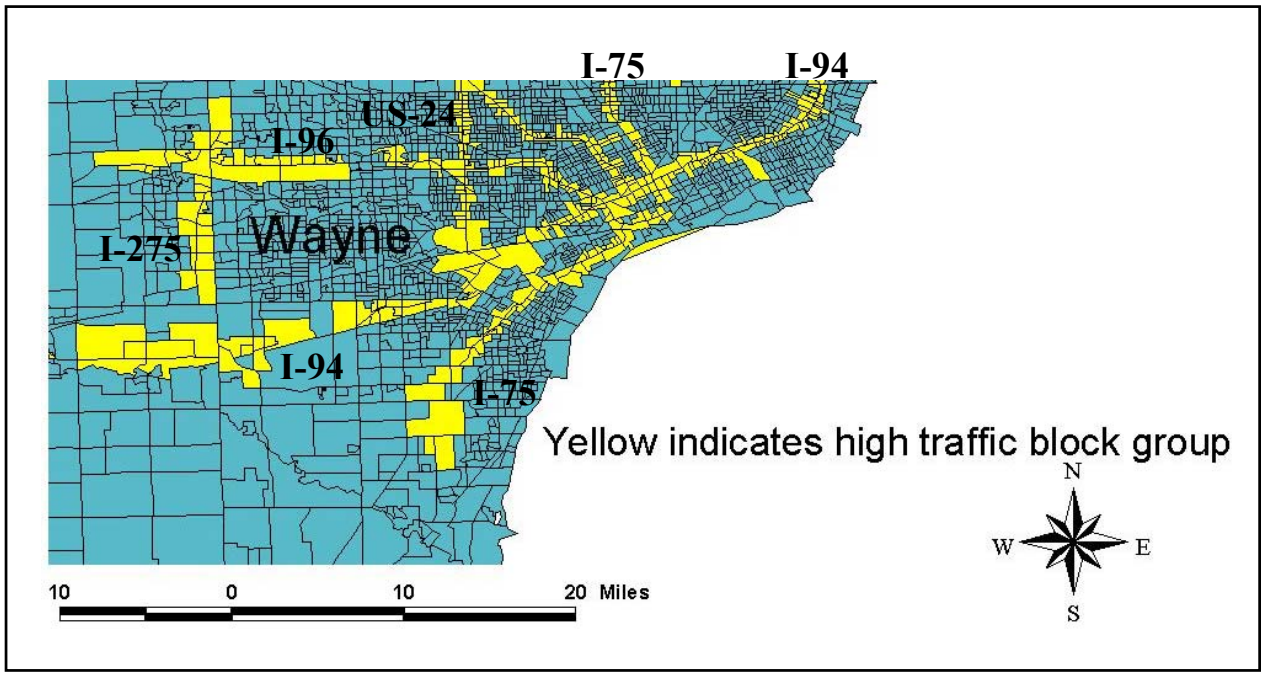

Figure 3-5. Detroit MSA 2000 census block groups intersecting roadways with one-way daily flow in excess of 50,000 vehicles.

In addition to these line sources, the major U.S. / Canada border crossings create major point sources of mobile source emissions as cars and trucks idle at custom stations awaiting inspection. The major crossings are the Ambassador Bridge (U.S. Interstate 75) and the Detroit/Windsor Tunnel (U.S. Highway 12). Transit times through U.S. and Canadian customs and inspections can be lengthy with extended idling times, sometimes for several hours. The high traffic volumes (Table 3-5) and long transit times will make these two border crossings into significant sources of mobile emissions, particularly diesel.

Table 3-5. Traffic volumes across U.S. / Canadian Border

	Vehicles per day		Location	
	Cars	Trucks	Latitude	Longitude
Ambassador Bridge	26,000	10,000	42.3117	-83.0742
Detroit/ Windsor Tunnel	28,000	1,000	42.3244	-83.0403

Several studies have measured the contribution of both organic and elemental carbon to the particulate matter emissions from motor vehicles by operation on chassis dynamometers and measurements in tunnels. The principal components emitted by diesel and gasoline fueled vehicles are organic carbon (OC) and elemental carbon (EC). In general, diesel exhaust is higher in elemental carbon than organic carbon, whereas organic carbon is the dominant species in the exhaust of gasoline fueled vehicles. Per vehicle, total carbon emissions from light and heavy-duty diesel vehicles can range from 1 to 2 orders of magnitude higher than those from gasoline vehicles.

The majority of PM emitted by motor vehicles is in the $PM_{2.5}$ size range. Particles in diesel exhaust are typically trimodal consisting of a nuclei mode (<0.1 μm), an accumulation mode (0.1 – 1.0 μm) and a coarse mode (1 – 10 μm) that are lognormal in form (Kittelson, 1998). More than 90% of the total number of particles are in the nuclei mode, which contains approximately 1 to 20% of the particle mass with a mass median diameter of about 0.02 μm, whereas the accumulation mode (with a mass median diameter of about 0.25 μm) contains most of the mass, with a smaller fraction (5 to 20%) contained in the coarse mode. Kerminin et al. (1997), Bagley et al. (1998), and Kleeman et al. (2000) also have shown that gasoline and diesel fueled vehicles produce particles that are mostly less than 2.0 μm in diameter.

3.2.4 Meteorology

Meteorological conditions for Detroit during 2002 are summarized in Table 3-6 and depicted in Figure 3-6. The summary data show that Detroit has long summer and winter seasons punctuated by shorter fall and spring seasons. Prevailing winds in Detroit are from the southwest during most times of the year except during March and April when winds are from the northwest. The predominant southwest winds separate emissions from the source areas in Detroit with the western part of Wayne County.

Table 3-6 Monthly Summary Statistics for Meteorological Parameters in Detroit during 2002.

	Jan.	Feb.	Mar.	Apr.	May	Jun.	Jul.	Aug.	Sep.	Oct.	Nov.	Dec.
Hi Temp (^{o}F)	30	33	44	58	70	79	83	81	74	62	48	35
Low Temp (^{o}F)	16	18	27	37	47	56	61	60	53	41	32	21
Precip. (in)	2	2	3	3	3	4	3	3	3	2	3	3
Snow (in)	9	9	6	1	0	0	0	0	0	0	2	10
Wind Spd. (mph)	13	12	13	12	11	10	9	9	9	11	12	12
Wind Dir.	SW	SW	WNW	WNW	SW	SW	SW	SW	SW	SW	SW	SW

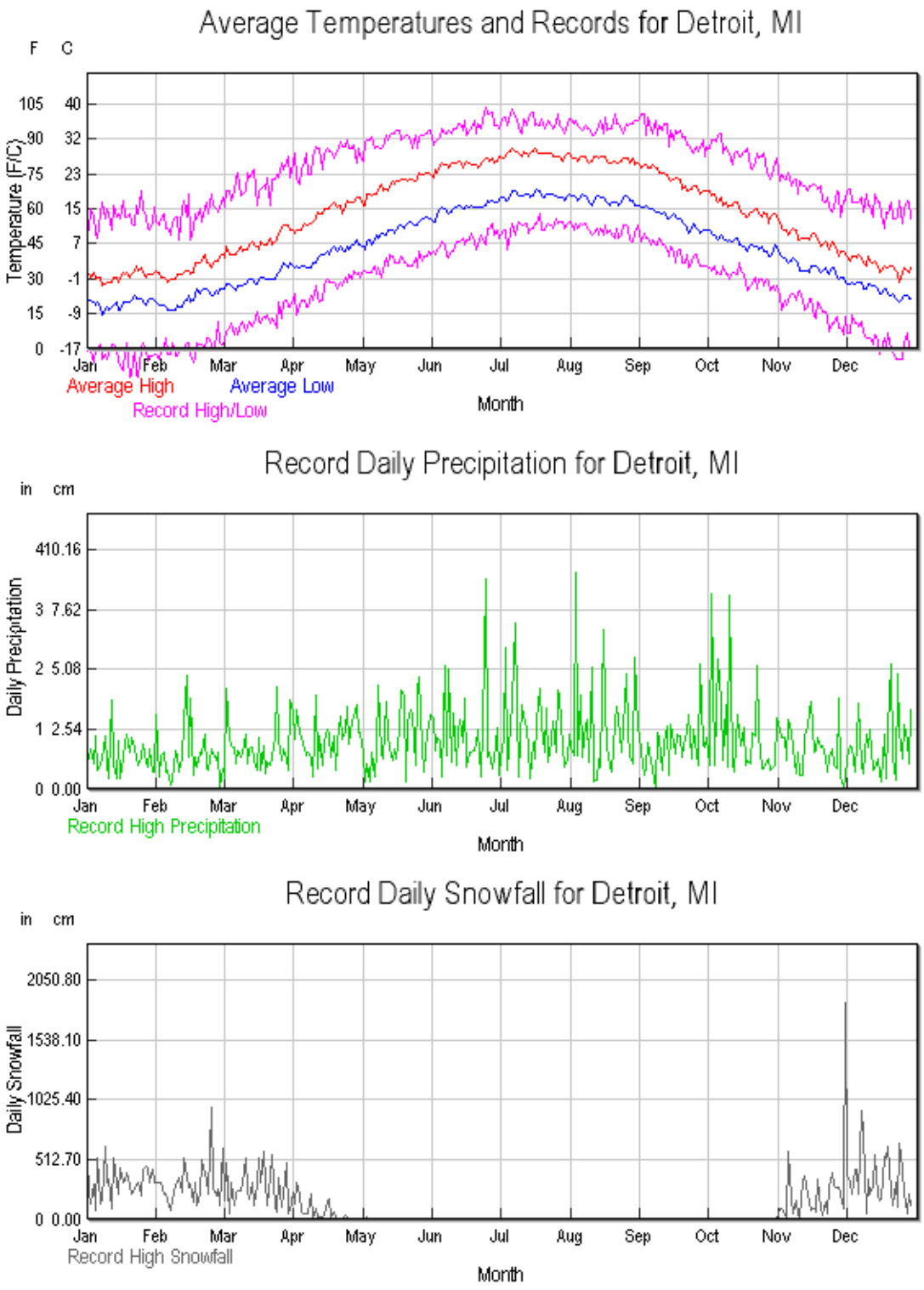

Figure 3-6. Historic weather patterns in Detroit

3.2.5 Available Monitoring Data

There are two sources of monitoring data in the Wayne County area. The first source is the AIRS-AQS database and the second is the Detroit Air Toxics Pilot Study (DATPS). The locations of the monitoring sites within Wayne County are shown in Figure 3-4. Monitoring at the DATPS sites (I-696/Lodge, Allen Park and Yellow Freight) was operated every 6th day for one year (April 2001-April 2002). At these sites, TSP metals, VOCs, carbonyls and PAHs were measured.

Seasonal and annual concentrations of $PM_{2.5}$ at the air toxics sites in Wayne County were in the range of 15-20 µg/m^3 during 2002 (Figure 3-7). Levels of $PM_{2.5}$ were slightly higher at sites located in Detroit (Allen Park, SW HS, Dearborn and Wyandotte) than sites in less industrial parts of Wayne County. $PM_{2.5}$ was consistently higher at Dearborn than at any other site in Wayne County. This is likely due to the close proximity of an automotive production facility near the Dearborn site.

$PM_{2.5}$ concentrations were generally similar between seasons except during summer, which were higher than the rest of the year. At the Dearborn site $PM_{2.5}$ concentrations were similar each season except for winter. The higher $PM_{2.5}$ concentrations during summer is most likely due to secondary photochemical formation of aerosols from mobile source related precursors.

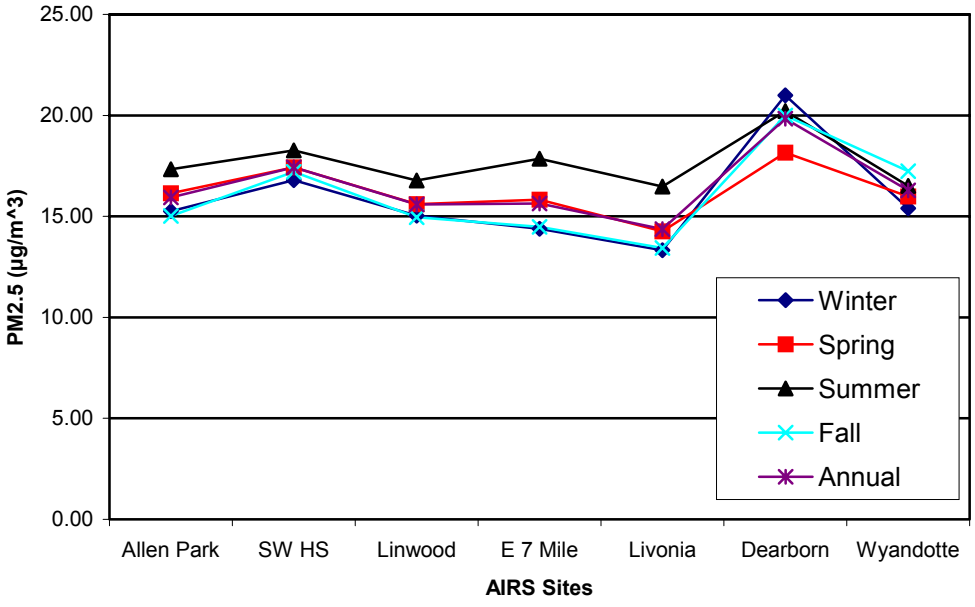

Figure 3-7. Seasonal and annual $PM_{2.5}$ concentrations measured at air toxics sites in Wayne County during 2002.

Composition data of $PM_{2.5}$ at the Allen Park site show that organic carbon, sulfate and nitrate constitute about 90% of $PM_{2.5}$ (Bortnick and Hafner, 2001). Figure 3-8 shows that the distribution of primary species is variable between season with proportions of nitrate varying from 11% in summer to 33% in winter. Organic carbon also varies somewhat from 34% in winter to 44% in fall. Proportions of sulfate remain relatively consistent during each season, but peaks in summer at 33% of the overall $PM_{2.5}$ composition.

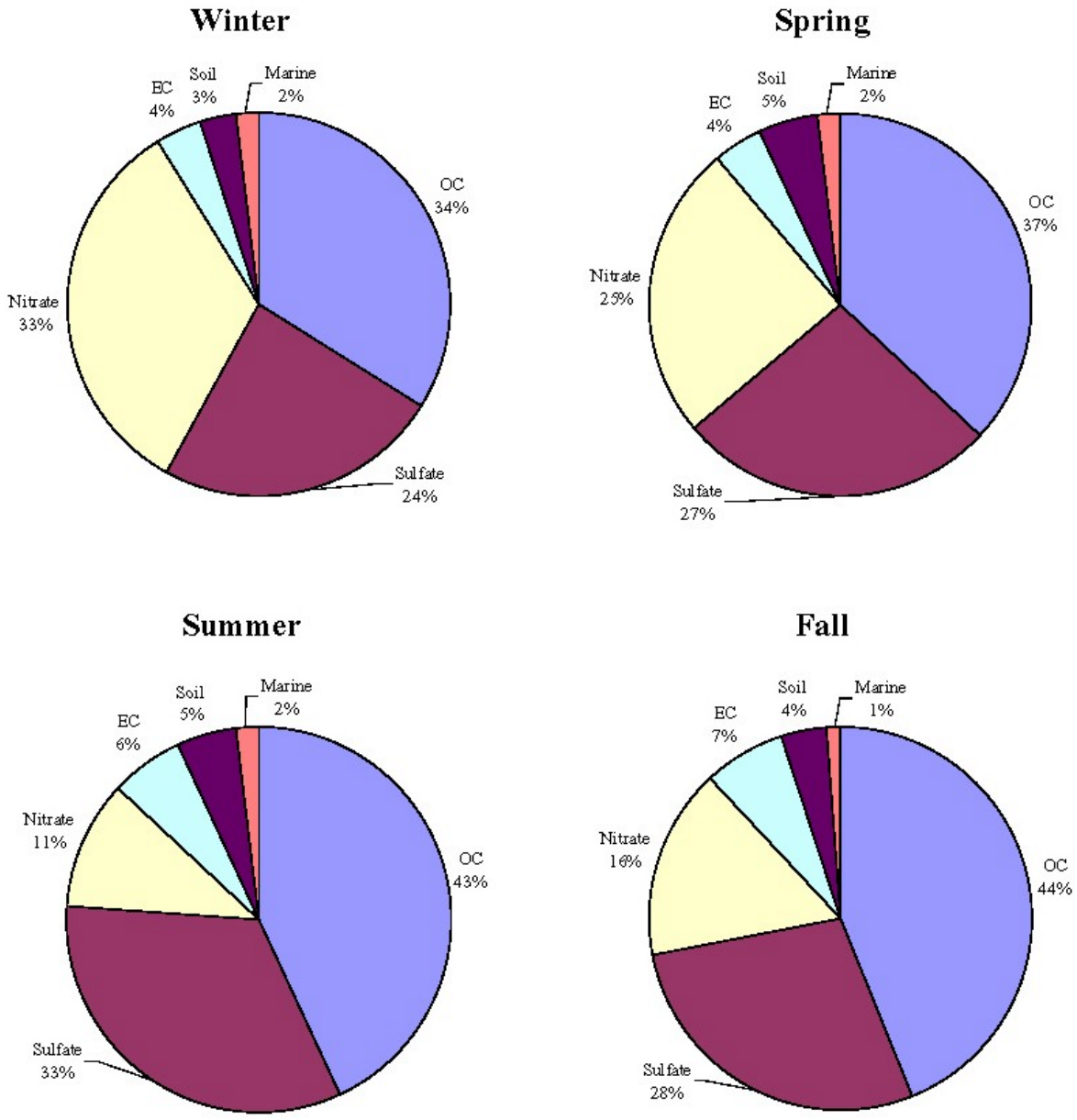

Figure 3-8. Seasonal composition of $PM_{2.5}$ measured at Allen Park.

Preliminary source apportionment of data collected at Allen Park shows several local sources contribute to the $PM_{2.5}$ loading at this site (Bortnick and Hafner, 2001). Table 3-7 shows the regional contribution of $PM_{2.5}$ at the Allen Park site, the single largest loading factor, was 36% with the remainder coming from local point and mobile sources. There were discernible signatures from mobile sources, coal burning, oil and other industrial sources. Although mobile sources accounted for only 11% (motor vehicle and diesel) of $PM_{2.5}$ mass directly, there is a substantial amount related to secondary organic aerosols that are typical of mobile source emissions.

Table 3-7. Preliminary source apportionment results from Allen Park site.

Factor	Mass ($\mu g\ m^3$)	% of Total Mass	Possible Sources	Key Findings
1	1.3	6	Motor vehicle	OC, acetylene, toluene
2	9.6	36	Regional	Almost all NH4 and NO3, 50% of SO4
3	2.0	9	Coal, smelter	50% SO4, many VOCs
4	2.9	14	Oil, industrial	Ni, OC, MEK and chloromethane
5	4.2	19	VOCs	Aldehydes, MEK, toluene; higher in summer
6	2.3	11	Industrial	EC/OC ~0.9, lower on weekends
7	1.2	5	Diesel	EC/OC ~3, high Mn, PAH, Zn, Fe, lower on weekends

3.3 Sampling the Study Population

3.3.1 Identifying and Selecting Study Population

Study participants and households will be identified through a step-wise approach involving identifying census blocks with and without impacts from local point and/or mobile sources based on prevailing wind direction and then targeting individual households for recruitment into the study. Individual census tracts and blocks will be identified by evaluating available data on the location and emissions from various sources, ambient air concentrations and housing stock. Once census tracts/blocks are identified we will work through existing community action groups in Detroit and the surrounding area to assist with contacting individual households. Partnering with community groups will be important in making connections with residents and communicating the purpose of the study.

The process by which we will select study participants and households follows.

Select Census Tracts/Blocks

- Identify the mobile, stationary, and regional sources of PM and air toxics in the Detroit MSA, with the focus on Wayne County and city of Detroit. Evaluate the size of the source and potential magnitude of impact.
- Use available source emissions and air monitoring data to understand the airshed.
- Identify specific geographical areas and census tracts that may be (1) highly impacted and (2) those that may be minimally impacted based on their location relative to sources and meteorology.
- Identify housing characteristics for the identified census tracts.
- Evaluate the potential for participation by residents and security in the census tracts.
- Perform screening measurements in the selected tracts and blocks to assess the potential impact of sources.
- Select census blocks within the tracts that best fit these criteria.

Select Housing Unit

- Develop criteria for inclusion (e.g., non-smoker, non-smoking household, ambulatory, literate, plans to be in the same dwelling for the next 9 months, detached home, and age 18 or older.
- Work with community groups to select homes that fit criteria.

Select Study Participants

- Develop criteria for selection (e.g., age >18 years, working adult, etc.).
- Select participants from those identified by community organizations.

3.3.2 Selecting Census Blocks

Census blocks will be chosen for monitoring based on proximity to stationary point and mobile sources and housing stock (age of structure) within the census block. Areas for participant recruitment and exposure monitoring are shown in Figure 3-4. With predominant southwest winds and numerous point and mobile source areas located in southwest Detroit, we expect the highest exposures to occur in northeastern Wayne County including Detroit and Dearborn. Conversely, we expect the lowest exposures to mobile and point source related PM and air toxics to occur in southwestern Wayne County. Census blocks will be selected in seven different areas to characterize personal exposures and residential indoor and outdoor concentrations.

Exposures related to industrial point sources will be measured in two areas of eastern Wayne County. Census blocks will be selected for monitoring in southwest Detroit near Zug Island (area 1 in Figure 3-4) to characterize exposures from industrial sources (iron/steel and oil refineries). Exposures to industrial sources will also be characterized in census blocks located near the River Rouge area of Wayne County (2 in Figure 3-4). Industry in this area is primarily coal-fired utilities, municipal incinerators and iron/steel. Housing stock in both areas generally consists of older homes built before 1960. There is a larger percentage of homes built after 1980 in the River Rouge area, but the majority of homes were built pre-1960.

Mobile source related exposures will be split between areas impacted primarily by diesel and gasoline powered motor vehicles (i.e., mobile sources). Census blocks will be selected near the Ambassador Bridge in southwest Detroit to characterize exposures to idling diesel (3 in Figure 3-4). Diesel exposures will also be characterized in census blocks along Route 53 near E. 7 mile Rd (4 in Figure 3-4) in Detroit (northeastern Wayne County). This area is more removed from industrial sources than the Ambassador Bridge and will represent exposures to diesel trucks driving on a roadway. Houses in these areas were mostly built before 1960. Areas impacted primarily by gasoline powered motor vehicles in stop-and-go traffic conditions with limited point source impacts are located in Dearborn (5 in Figure 3-4). Housing stock in this area was mostly built after 1980. Census blocks will also be chosen near the John C. Lodge Highway to characterize exposures to stop-and-go traffic (6 in Figure 3-4). Homes in this area were mostly built before 1960. Areas of southwest Wayne County away from the major interstates are expected to be minimally impacted by mobile and point sources and would represent upwind exposures with regional characteristics (7 in Figure 3-4). Housing stock in southwest Wayne County is generally newer with about half of all homes built after 1980.

The number of households targeted for monitoring in each source category was chosen to provide sufficient observations of exposures in each category to determine source impacts on indoor concentrations and personal exposures (Table 3-8). There will be 40 households not impacted by point sources and 80 households that will be impacted by point sources. Similarly, 45 households will not be impacted by mobile sources, 75 households will be impacted by mobile sources (45 gasoline, 30 diesel). Approximately 15 households will be in an area that is expected to be minimally impacted by mobile or point sources. The relatively equal distribution of homes throughout the study domain should provide sufficient data on exposures to the mix of sources in Detroit and surrounding Wayne County. The number of homes in each source category should be adequate to satisfy the objectives and goals of the study.

After individual census blocks are identified as possible monitoring areas, real-time pollutant data will be collected to assess the impact of mobile, point and regional sources on residential neighborhoods. Continuous data will be collected on PAH, CO, and PM concentrations at several locations in the census block. Direct-reading devices will be used which permit fast and efficient determination of combustion-related pollution gradients. This information will be used to determine which neighborhoods best match the selection criteria for a given subject population, such as those that are impacted by gasoline mobile source emissions.

Table 3-8. Proposed matrix of number of households to be selected in each source category. The number in parentheses corresponds to the area in Wayne County indicated in Figure 3-4.

		Mobile Sources				
		None	Gas, stop-and-go	Diesel, driving	Diesel, idling	Point Source Total
Point Sources	None	15 (7)	25 (6)	0	0	40
	Coal, oil, steel, power, incinerator	30 (1, 2)	20 (5)	15 (4)	15 (3)	80
	Mobile Source Total	45	45	15	15	120

3.3.3 Participant Recruitment

Participant recruitment will be achieved by working with community groups to contact occupants in residences previously selected from the census blocks. We will work with existing community level groups such as the Community Action Against Asthma (CAAA) to help facilitate contact with potential study participants. We will employ an approach that will minimize selection bias in recruiting. Participants will be selected from the seven geographical areas that represent various source categories (Figure 3-4, Table 3-8). Households will be selected on the same side of a given line or point source. Information provided to each family identified by the community organization as willing to participate will include a description of the study, inclusion criteria and a brief description of benefits for participating. A detailed recruitment plan will be developed to select a population that minimizes selection bias. Selection criteria for participants are: (1) non-smoker, (2) non-smoking household, (3) ambulatory, (4) able to read and write English, (5) plans to be in the same dwelling for the next 9 months, (6) living in a detached home, and (7) age 18 or older. Individuals expressing interest in participating in the study who meet the inclusion criteria will receive a visit from a study recruiter to further explain the study. At these visits, the study recruiters will obtain informed consent from participants. Recruitment will continue until the number of needed participants has been obtained. Additional homes will be selected as needed. Because the study will be performed over a 3-year period, recruitment and retention of subjects will be performed in the months immediately prior to each of the monitoring seasons. Retention of subjects participating in two seasons will be performed by letter and phone call follow-ups. This follow-up will encourage them to remain in the study and provide them a brief summary of findings to date in their residential area. Approval will be obtained from all appropriate Institutional Review Boards (IRB) including EPA's Human Subjects program, prior to contact with potential participants. Members of Research Triangle Institute, International's IRB will provide the formal review of the recruitment and subject interaction guidelines.

3.3.4 Selecting the Central Site Monitor

The central site monitor for this study should be an existing monitoring site representative of the airshed in which the study will take place and one that reflects community based exposures that

are typically used in epidemiological studies. Residential and personal monitoring will be conducted in areas impacted by point and/or mobile sources and areas with minimal impacts. Therefore, the central site monitor should be situated such that it captures a variety of exposure scenarios depending on meteorological conditions. The Allen Park site will serve as the central monitoring location for this exposure study (Figure 3-4). The Allen Park site is located between two major interstates (I-94 and I-75) providing data on mobile source impacts, in close proximity to but generally not directly downwind of the industrial source region in eastern Wayne County, and centrally located in Wayne County. Current monitoring capabilities at the Allen Park site include:

- Carbon monoxide
- Ozone
- $PM_{2.5}$ and PM_{10} (integrated)
- $PM_{2.5}$ TEOM
- Metals
- PM speciation
- Meteorology

4.0 MEASUREMENT PLAN

4.1 Overview

Field monitoring will be conducted at 120 residences over a three-year period. Measurements of air toxics, PM, PM constituents, and criteria gases will be collected in each home and from each participant for five days during both a winter and summer season for a total of 1200 household-person/days of measurements. Monitoring is anticipated to start in the summer of 2004. A summary of the core personal, residential indoor, residential outdoor, and central site monitoring that will be conducted is given in Table 4-1. A combination of both weekday and weekend sampling will be conducted in order to evaluate expected variations in industrial source emissions, traffic volumes, and personal activities.

Table 4-1. Summary of Collection and Analysis Methods

Parameter	Sample	Collection Method				
		Personal	In Res	Out	Central Site	Rotating Community Site*
$PM_{2.5}$ Mass Elements Sulfate SVOCs EC	$PM_{2.5}$ - Teflon filters	PEM (2L)	HI (10L)	HI (10L)	HI (10L), PEM (2L)	HI (10L), PEM (2L)
PM_{coarse} Mass, Elements	PM_{10} - Teflon filters	--	Sioutas Cascade Impactor (9L)	Sioutas Cascade Impactor (9L)	Sioutas Cascade Impactor (9L)	Sioutas Cascade Impactor (9L)
Fine & Coarse PM		--	--	--	Dichot	Dichot
$PM_{2.5}$	Nephelometer	MIE pDR-1200	MIE pDR-1200	MIE pDR-1200	MIE pDR-1200	MIE pDR-1200
EC-OC	Quartz filters (TOA)	--	PEM (2L)	PEM (2L)	PEM (2L)	PEM (2L)
EC	$PM_{2.5}$ Teflon filter	PEM (2L)	PEM (10 L)	PEM (10L)	PEM (10L)	PEM (10L)
Nitrate	Mini denuder sampler (glass fiber filter)	--	Mini denuder (0.8 L)	Mini denuder (0.8 L)	Mini denuder (0.8 L)	Mini denuder (0.8 L)
Ozone SO_2 NO_2	Ogawa Badge	Passive badge	--	--	Passive badge	Passive badge
Carbonyls	DNSH badge	Passive badge	Passive badge	Passive badge	Passive badge	Passive badge
VOCs	Carbopak-X	Passive badge or tube	Passive badge or tube	Passive badge or tube	Passive badge or tube	Passive badge or tube
Air exchange rate	PFT	--	Passive collector	--	--	--

* One residential outdoor location each week will be designated as a temporary community site . This will only require the additions of a dichotomous sampler and Ogawa criteria gas monitoring badges to the normal residential outdoor monitoring scheme for this home to be consistent with the measures collected at Allen Park.

Measurements will include personal, residential indoor, residential outdoor, and central site monitoring for $PM_{2.5,}$ VOCs and carbonyls. All $PM_{2.5}$ filters will be analyzed for mass, elemental carbon (EC), selected elements, and sulfate as sulfur. Since participants cannot carry a large number of personal monitors, some pollutants will only be measured indoors, outdoors, and at the central monitoring site. These include PM_{coarse}, nitrates, SVOCs, and EC/OC as collected on quartz filters. The criteria gases will only be measured in central site and personal exposure samples. Carbon will be measured by two methods. Elemental carbon on $PM_{2.5}$ filter samples will be estimated based on transmittance of the filter. Using a second approach, elemental and organic carbon will be collected on quartz fiber filters and then their mass concentration determined by thermal optical analysis (TOA). This latter method has shown large positive artifacts for organic carbon in indoor samples; these are speculated to be related to non-ambient sources resulting from the presence of many potential indoor sources (e.g., carpet, furniture, wall coverings). Thus, although organic carbon will be measured in indoor samples, data may not be used unless methods for minimizing the artifact problem can be developed. The indoor samples will be archived and used in the event these artifact problems can be overcome as well as a source of samples potentially useful in the analysis of organic aerosol markers. Passive methods for monitoring VOCs and carbonyls are currently under development. Although these methods have not been finalized, results to date suggest that performance requirements for this study can be met.

The central site monitor is located at Allen Park and is part of the Speciation Trends Network. Monitoring that is ongoing at this site is shown in Section 3.3.4. A dichotomous sampler (Dichot) will be used to collect 24-hour $PM_{2.5}$ and $PM_{10\text{-}2.5.}$ Filters will be analyzed for mass, elemental carbon, selected elements and sulfate as sulfur. A prototype dichotomous sampler is currently being evaluated as an FRM for $PM_{2.5\text{-}10}$ and a FEM for $PM_{2.5.}$ Results from the Dichot will be used to establish bias between personal and residential monitoring methods and standard methods. A Sioutas Cascade Impactor will collect PM_{coarse}. Filters will be analyzed for mass and selected elements.

A rotating community site will be established at one residence weekly. This will permit for a direct comparison between community-based measurements across the metropolitan area. Rotation of the site through each of the seven targeted population areas will permit for a more extensive evaluation of spatial variability and the representativeness of the Allen Park community site. Since a base of residential outdoor measurements will already be taking place for every home incorporated into the study, only the additions of a dichotomous sampler for $PM_{2.5}$ and $PM_{10\text{-}2.5}$ mass measurements and and Ogawa badges for criteria gases and a Sioutas Cascade Impactor for PM_{coarse} will permit a consistent sampling scheme as that being performed at the Allen Park site.

4.2 Target Pollutants

Target pollutants in the following categories will be monitored throughout the study.

- PM mass - $PM_{2.5}$, PM_{coarse}
- PM species - sulfates, nitrates, elemental carbon (carbon black), organic carbon, elements, SVOCs
- Air Toxics –

  - VOCs - 1,3- butadiene, benzene, carbon tetrachloride, 1,2-dichloropropane, tetrachloroethylene, trichloroethylene,
  - Aldehydes -acrolein, acetaldehyde, formaldehyde
  - Metals –cadmium, lead, manganese, nickel
- Criteria Gases - NO_2, SO_2, O_3
- Source Markers – specific markers and their common sources are shown in Table 4-2.

The target compounds were selected based on several criteria. Sulfates, nitrates, and carbon species make up the bulk of the PM mass for the Detroit area. The VOC and carbonyl air toxics are those measured at either the National Air Toxics Trends Sites (NATTS) or the Air Toxics Monitoring Pilot Cities. Currently, acrolein and 1,3 butadiene are not measured at these sites, so this data may be of use for NATA. The Air Toxic metals are those measured at the same sites and which are amenable to XRF analysis. The criteria gases have been associated with PM concentrations and health effects, although the relationship between concentrations of PM and the criteria gases in exposure samples is unclear. We will measure both to determine if the criteria gases are confounders or surrogates of PM exposure and the associated health effects.

A number of physical and chemical properties of PM have been hypothesized as causal agents of PM-induced health effects. Although we have tried to be as inclusive as possible, certain parameters will not be measured. While it is recognized that carbon monoxide represents an important pollutant marker for mobile source emissions, the ability to accurately detect this PM-related pollutant indoors has been shown to be widely problematic. Based upon average ambient 3-4 ppm CO concentrations reported for locations in Wayne County, measurement of this pollutant inside residences (where concentrations might be expected to routinely fall below 1 ppm -- the limit of detection for most portable instrumentation) using portable equipment is not considered feasible at this time. Therefore, carbon monoxide measurements will not be performed during the study at any location or on a personal basis. Other pollutant species already included in the measurement plan are considered adequate for the identification of mobile source impacts.

Ultrafine PM, acid aerosols and biogenic particles have been excluded because of the high burden of measuring these in residences and because previous research has shown poor correlations between indoor and outdoor concentrations. Ultrafines will be collected at the ambient site. Although metals (V, Cu, Fe, Zn, and Ni) will be measured, soluble metals will not. We have chosen to measure SVOCs on the filter samples instead. There are no standard methods for the oxidizing agents such as peroxides, and thus, they are not being measured in this study. Personal methods for measuring coarse PM are not reliable, and also will not be measured in this study. However, we will collect PM_{coarse} inside and outside residences and at the ambient site. Finally, although certain metals including arsenic, chromium (+6), and mercury are listed as air toxics, monitoring for these species is beyond the resources for this study.

Table 4-2. Proposed Source Markers

Class	Compound	Source
Elements	Nickel, vanadium	Oil refineries/oil combustion
	Manganese	Iron/steel production
	Sulfur	Sulfate
	Potassium	Wood
	Iron	Iron/steel production
	Zinc	Incineration
	Silicon, Aluminum	Road dust
Carbon	Elemental	Diesel, mobile sources
	Organic	Mobile sources
Ions	Sulfate	Regional background, coal burning
	Nitrate	Regional background, mobile sources
VOCs	MTBE, benzene, toluene, xylenes, 1,3-butadiene	mobile sources
	Benzene	Steel production, oil refineries, coke ovens
	Ethyl benzene	Mobile sources, oil refineries
	Toluene	Oil refineries, coke ovens
	Xylene	Oil refineries, iron/steel production
Carbonyls	Acrolein	Mobile sources
	Formaldehyde	Mobile sources (outdoors), building materials, furnishings (indoors)
	Acetaldehyde	Mobile sources
SVOCs	Norhopane, hopane, homohopane, bishomohopane	Petroleum/mobile sources
	Levoglucosan	Wood smoke
Alkanes	C27-C30	Mobile sources, wood combustion, natural gas combustion
PAHs	pyrene, chrysene/triphenylenen, benzo[k]fluoranthene, benzo[b]fluoranthene, benzo[a]pyrene,indeno[1,2,3-cd]pyrene, benzo[ghi]perylene	Mobile sources, wood combustion, natural gas combustion

4.3 Field Measurement Protocol

4.3.1 Field Monitoring Daily Timetable

Field monitoring for each participant will be conducted over a 5-day period during two seasons within a year. Activities taking place in each home over the 5-day sampling period are shown in Table 4-3. Sample and data retrieval at each home will take ~ 45 minutes and will occur between 6 am and 10 am. Ideally, sample changeout for all participants should take place at the same time as the central site monitor (8 am) to give a direct comparison of the data. However, this is not practical due to staffing limitations and the need to accommodate the participant's schedules. Rather, samples will be changed as close as reasonably possible to the central site sampling time (projected to be ± 2 hours). Two 3-person teams will conduct sample and data retrieval activities in the participant's homes. Each team will be responsible for monitoring three homes per day. Thus, six homes/participants will be monitored during a 5-day period. Monitoring schedules will be staggered for each home/participant so that both weekday and weekend data collection will occur. Approximately 40 homes/participants will be monitored over a 10-week period each season. New participants will be recruited each year.

4.3.2 Sample Locations

Samples will be collected at an outdoor and indoor residential site at each home. The outdoor site will be on the non-roadway side of the home. For mobile source targeted homes that are surrounded by roads, the outdoor monitor will be located on the least traveled segment/road side of the home. The site will be located using the best available siting criteria relative to the conditions present at each residence (i.e. away from the residence or other buildings, any roadway, parking lots, or other known sources of PM). The indoor residential site will always be the primary living area in each home, which is defined as the non-bedroom area where an individual would spend most of their time. Samples will be collected away from heating vents, known sources of PM, and at least one meter from the wall. Both indoor and outdoor samples will be collected 1 to 1.5 meters above the ground. For personal exposure samples, pumps will be secured in the pocket of a lightweight cloth vest or coat; the sampling heads will be attached to the lapel to collect air from the breathing zone.

Table 4-3. Sample Collection in Each Household

Day	Time	Activity
Pre[a]	10 am to 4 pm (length: ~ 30 minutes)	1. Obtain informed consent 2. Deploy PFT emitters 3. Measure house volume for air exchange rate measurement 4. Administer household questionnaire 5. Confirm schedule for next monitoring day
1	6 to 9 am (length: ~ 45 minutes)	1. Set up indoor/outdoor monitoring equipment 2. Install sampling filters/cartridges in all equipment 3. Measure sampling flow rates 4. Give participant personal monitor in sampling vest 5. Give detailed instructions for personal monitoring and diary completion. 6. Set out PFT collectors 7. Confirm schedule for next monitoring day
2 - 4	6 to 9 am (length: ~ 45 minutes)	1. Measure sampling flow rates in all equipment to end current session 2. Replace sampling filter/cartridges in all equipment 3. Replace PFT collectors 4. Measure sampling flow rates of replacement equipment 5. Give participant personal monitor in sampling vest 6. Download nephelometer data 7. Review daily diary; Administer post monitoring questionnaire 8. Confirm schedule for next monitoring day
5	6 to 9 am (length: ~ 45 minutes)	1. Measure sampling flow rates in all equipment 2. Remove sampling filter/cartridges; disassemble all equipment 3. Collect PFT collectors and emitters 4. Download nephelometer data 5. Review daily diary; Administer post monitoring questionnaire 6. Pay incentive

[a] Will be performed 24 to 48 hours prior to field monitoring

4.3.3 Sample Management

All samples (field sample, field duplicate, laboratory blank, field blank, etc.) will be given a unique bar code label at the time of preparation. Specific identifiers will be used for each set of samples so that different size fractions, collection instruments, collection dates and other key identifiers will be easily discernable to avoid operator error during sample collection and analysis. Data log sheets and an electronic sample collection "shell" will be used in conjunction with the bar codes to track sample collection (collection schedule, recovery, and transport). Field notebooks will be used to record sampling information regarding instrument audits, flow verifications and other factors ensuring the integrity of the data collection. Senior field staff will be responsible for daily archival of samples, their handling, and transport back to the laboratory.

Collected samples will be stored in the field at $\leq 0^{o}$C after their collection. Samples will be shipped to the laboratory weekly via overnight carrier in chilled containers. Dedicated study

freezers of ≤ -20°C will be used to store samples prior to analysis. Chain of custody forms will accompany all samples from the time of their collection through storage, analysis, and archiving. Individuals handling the samples will acknowledge this action by signature and date on the chain of custody forms that will remain with the samples until the time of their disposal. Copies of this record will be maintained in the study files at all times.

4.4 Monitoring Methods

4.4.1 $PM_{2.5}$ and PM_{coarse}

4.4.1.1 Collection

$PM_{2.5}$ will be collected using inertial impactor samplers. All of these samples will be collected for 24 hours onto pre-weighed 37 mm Teflon filters (Teflo®-Gelman Sciences, Ann Arbor, Michigan). The Harvard Impactor (HI) samplers will be used at the central site, outdoors at the home, and indoors at the home to collect $PM_{2.5}$ at flow rates of 10 LPM. The HI particle sampler uses an oiled porous impactor plate to minimize particle bounce off while also providing a sharp cut point. The PEM (Personal Environmental Monitor, MSP model 100) $PM_{2.5}$ sampler operates at a flow rate of 2 LPM and will be used to collect personal air samples. The PEMs inlets have been modified with a 4 μm "scalper plate" (MSP#PEM-019) to reduce large particle burden upon the impactor during the sample collection. PEMs sampling pumps have been modified to permit extended operation as well as temperature, flow rate, and motion sensing data logging. $PM_{2.5}$ mass concentrations collected on PEM samplers have been shown to be equivalent to those produced by collocated FRM samplers in earlier NERL panel studies (Williams et al., 2000).

The Sioutas Cascade Impactor (SKC, PA) will be used to collect five size ranges of PM for a PM_{coarse} measurement. The Sioutas Cascade Impactor consists of four impaction stages and a final filter that allows the separation and collection of airborne particles in five size ranges. It uses a flow rate of 9 LPM. Particles above each cut-point are collected on a 25-mm PTFE filter in the appropriate stage. Particles below the 0.25 μm cut-point of the last stage are collected on a 37-mm PTFE final filter. Size-fractionated samples can be analyzed gravimetrically, chemically, and microscopically.

4.4.1.2 Analysis

Mass, EC, elements, and SVOCs collected on the filter samples will be analyzed using gravimetric, XRF, optical transmission, and GC-MS methods, respectively. The first three methods are nondestructive thus multiple analyses can be performed. For SVOCs, filters must be extracted prior to GC/MS analysis. Once this is done no additional analyses can be performed.

Mass. Gravimetric analysis of PM filter samples will be performed following 24-hour equilibration at 25°C (± 5°) and 40% relative humidity (± 5%). The method quantitation limit for the PEM PM monitoring has been determined to be 1.91 $\mu g/m^3$ with a collocation precision (root mean square) of ± 4.3 $\mu g/m^3$. These historical values indicate that the gravimetric method to be used will be sufficient under even the most stringent conditions (low volume personal

monitoring). Lawless and Rodes (1999) have reported upon the gravimetric procedures required to accurately measure filter mass loadings from low volume air sampling.

Elemental Carbon. An optical transmittance procedure, currently being evaluated using data from two recent EPA studies (the RTP Panel Study and the Tampa Children's Study) will be used to establish EC concentrations. Data from our recent studies is being used to compare results between this method and the thermal optical analysis (TOA) method to determine bias.

Elements. Multiple elements can be measured using X-ray Fluorescence techniques on samples collected using Teflon filters. The filter is irradiated with x-rays and then elemental fluorescence is quantified. The detector is placed at an appropriate angle to measure the intensity at a certain wavelength. The elements of primary interest and their estimated detection limits are given in Table 4-4. As the table shows, although samples will be analyzed for all of these elements, some of them may not be at sufficiently high concentrations to be detected. Precision ranges from 6-9 % depending on the concentration, while accuracy is +/- 10%.

Table 4-4. Elemental detection limits and environmental concentrations (ng/m^3).

ng/m^3	Detection Limits		Environmental Concentrations	
Element	PEM (2LPM)	HI (10 LPM)	Detroit, MI*	Detroit AT Study
Al	82.8	16.6	25	--
Si	28.3	5.7	110	--
S	12.5	2.5	1500	--
K	6.6	1.3	78	--
Ca	4.1	0.8	69	6.7
V	2.0	0.4	2	--
Cr	1.3	0.3	2	2.9
Mn	1.5	0.3	4	100.0
Fe	4.1	0.8	120	--
Ni	1.9	0.4	2	2.3
Cu	2.0	0.4	6	--
Zn	1.3	0.3	25	--
Cd	5.2	1.0	--	--
Pb	3.2	0.6	6	--

* Speciation Trends Network data from October, 2001 to September, 2002 (Fourth External Review Draft of PM Criteria Document, 2003).

SVOCs. A variety of particle phase SVOCs will be analyzed including PAHs, alkanes, hopanes and steranes, organic acids, and sugars. Specific target compounds for this study include

- Alkanes – C_{23} to C_{34}
- Petroleum biomarkers – norhopane, hopane
- PAHs - pyrene, chrysene/triphenylenen, benzo[k]fluoranthene, benzo[b]fluoranthene, benzo[a]pyrene,indeno[1,2,3-cd]pyrene, benzo[ghi]perylene
- Organic acids – hexadecanoic acid, hexadecenoic acid, octadecanoic acid, octadecenoic acid
- Sugars – levoglucosan

SVOCs will be removed from the filter using a validated solvent extraction technique, such as sonication. The NERL is currently validating extraction, recovery and analysis procedures for particulate phase SVOCs collected onto teflon filter media of various sizes, sampling volumes, and sampling locations (community, residential indoors, residential outdoors). This work is being performed in the NERL's Organic Analysis Laboratory. Organic acids and sugars will be derivatized prior to analysis (using diazomethane or a similar compound). Filters will be spiked with several deuterated internal standards prior to extraction to check recoveries in all samples.

SVOCs in sample extracts will be measured using GC-MS. Specific ion monitoring (SIM) of select mass fragments for each of the target compounds will permit the establishment of calibrated response factors. SIM is a fundamentally more sensitive approach in comparison to total ion methods. Detection limits on the column should be 0.02 ng or less with estimated detection limits of ~0.01 ng/m^3 (10 LPM for 24 hr sample) with 10-15% precision. Extraction recoveries will be at least 80% and likely higher. Estimated performance results for selected SVOCs are shown in Table 4-5.

Table 4-5. Estimated Performance Results for Select PAH SVOCs

Analyte	MQL (ng/m^3)	Mean Analytical Precision (%)	Analytical Recovery %
Pyrene	$\leq$ 0.30	13	$\geq$ 85
Chrysene	$\leq$ 0.10	10	$\geq$ 95
Benzo(a)anthracene	$\leq$ 0.10	11	$\geq$ 95
Benzo(k)fluoranthene	$\leq$ 0.10	12	$\geq$ 95
Benzo(b)fluoranthene	$\leq$ 0.10	10	$\geq$ 95
Benzo(a)pyrene	$\leq$ 0.04	9.0	$\geq$ 95
Indeno(1,2,3-cd)pyrene	$\leq$ 0.07	11	$\geq$ 95
Benzo(ghi)perylene	$\leq$ 0.12	9.0	$\geq$ 95

Table 4-6 shows typical ranges in air concentrations for the target analytes. Method performance studies are currently under way. Standard operating procedures for this effort will be developed once the method is finalized.

Table 4-6. Estimated air concentrations for selected target SVOCs.

SVOC	Compound Class	24-hr Environmental Concentrations (ng/m^3)	
Season		Schauer & Cass 2000 winter	Zheng et al., 2002 spring, summer, winter, fall
Location		rural, suburban, and urban sites in the San Joaquin Valley, CA	rural/suburban and urban cites in AL, GA, MS, FL
nC23	Alkane		
nC24	Alkane	2.04-42.3	0.02-3.97
nC25	Alkane	2.44-41.2	0.13-4.00
nC26	Alkane	1.49-29.9	0.30-2.87
nC27	Alkane	1.50-25.0	0.46-1.88
nC28	Alkane	0.78-12.3	0.28-1.41
nC29	Alkane	3.22-40.4	0.62-2.65
nC30	Alkane	0.28-7.39	0.16-1.08
nC31	Alkane	0.68-16.1	0.68-2.10
nC32	Alkane	1.98-3.71	0.06-0.42
nC33	Alkane	3.31-5.02	0.12-0.51
nC34	Alkane		
norhopane	Petroleum biomarker		0.01-0.57
hopane	Petroleum biomarker		0.01-0.59
pyrene	PAH	0.05-3.28	0.02-0.56
chrysene/triphenylenen	PAH	1.2-7.7	0.03-2.55
benzo[k]fluoranthene	PAH	0.04-8.69	0.06-2.75
benzo[b]fluoranthene	PAH	0.10-10.7	0.06-2.40
benzo[a]pyrene	PAH	1.77-8.23	0.03-2.50
indeno[1,2,3-cd]pyrene	PAH	2.56-6.84	0.04-1.58
benzo[ghi]perylene	PAH	3.49-9.75	0.04-2.18
hexadecanoic acid	Organic acid		
hexadecenoic acid	Organic acid		0.1-1.7
octadecanoic acid	Organic acid		
octadecenoic acid	Organic acid		
levoglucosan	Sugar	22.5-7590	166-358

4.4.2 Nephelometer (personal DataRam®)

The NERL has successfully employed the MIE pDR (personal DataRam) nephelometer (MIE Inc., Bedford, MA) in well over 1000 sample collections in four major field studies. In this study, the pDR-1200 will be operated concurrently with personal, indoor, and ambient impaction samplers. It is a portable optics-based monitoring instrument that uses light scattering to estimate PM mass concentrations. The MIE provides real-time PM mass concentrations (~0.5 to 8 μm) on a one-minute basis. It will be operated using a 2.5 μm size selective inlet and relative humidity

controls to permit more optimized operation. The nephelometers will be calibrated by the manufacturer using 2.5 μm dust particles; no additional field calibration will be performed. Samplers will be zeroed prior to use and daily in the field. For personal monitoring, the MIE will be placed in the sampling vests along with the PEM samplers. The nephelometers correlate well (r> 0.8) with the PEM monitors (Tampa Asthmatic Children's Study).

This device has been determined to be robust and highly portable. Even though the instrument is capable of reporting 0.01 $\mu g/m^3$ of PM mass concentration, field evidence has indicated that a practical limit of detection of 1.0 $\mu g/m^3$ be used with a limit of quantification of 2.0 $\mu g/m^3$. Precision of the instruments has been shown to be ± 10% from collocated instruments under a variety of environmental conditions. Stored digital data will be recovered using the unit's RS232 data port and the manufacturer's software. Data will be recovered automatically from the instrument using a macro-imbedded software program (Excel). The output from this spreadsheet will provide minute-by-minute $PM_{2.5}$ nephelometer mass concentration data ready for input into the final analysis program (SAS).

The nephelometer data will be used to examine personal and indoor sources of PM and the influence that personal activities have on PM exposures. Recovery of MIE data during each measurement day will permit study technicians to investigate time-activity diary patterns with episodes of observed high PM concentrations (5 highest peaks). Comparison of data from the real-time devices (central site, personal and residential) will permit the direct determination of source influence upon personal and residential settings). Infiltration factors and source strengths will be calculated from the resulting data.

4.4.3 Elemental Carbon/Organic Carbon

Personal, indoor, outdoor, and central site samples will be collected for elemental carbon (EC). $PM_{2.5}$ EC will be collected onto Teflon filters at flow rates ranging from 2 to 16.6 LPM for 24 hours. These will be the same filters used to collect PM mass and SVOCs. An optical transmittance procedure, currently being evaluated using data from two recent EPA studies (the RTP Panel Study and the Tampa Children's Study) will be used to establish these EC concentrations. The optical method is nondestructive, therefore allowing for these filters to be used for the other primary analyses (e.g., extracted for SVOC content). Values of 0.11 and 0.34 $\mu g/m^3$ have been established for the LOD and MQL respectively, for the optical procedure.

The thermal optical analysis (TOA) method is a destructive EC and OC (Organic Carbon) analysis technique. This technique is well established and typically involves the collection of $PM_{2.5}$ carbonaceous matter on a pre-fired quartz filter. The NERL is currently evaluating experimental data pertaining to the collection, retention, and recovery of OC species on quartz, teflon, and other media (e.g., denuders, electrostatic precipitators). The need for back-up collection media associated with a primary quartz filter for OC is being evaluated based upon some reported literature findings about this technique. It is anticipated that data from these experiments will be fully evaluated prior to the start of field data collection. Based upon the results of these ongoing studies, modifications to the basic quartz filter-based OC collection may be required.

The TOA procedure calls for a known portion of the used filter area to be recovered and placed into a combustion chamber where it undergoes increased thermal heating under a controlled state. Typically an inert gas, such as helium is used in this process. As a result of the thermal heating, various carbonaceous fractions, such as the OC1 peak containing many of the more volatile organic carbon species, are released from the combustion chamber, converted to simple hydrocarbons and ultimately detected by an in-line flame ionization detector. The detection limits for OC, OC1, and EC by TOA are listed in Table 4-7. Historical data has shown that well over 95% of even the low volume (2 lpm) personal samples can meet this limit. Artifact issues have been raised on the use of quartz filters for organic and total carbon for personal and indoor exposure assessment (Landis et al., 2001). These artifacts have been associated with indoor SVOC sources not related to ambient infiltration.

Table 4-7 EC-OC Detection limits

Measure	Analysis Technique	MDL ($\mu g/m^3$)	MQL ($\mu g/m^3$)
OC	TOA	1.18	3.53
OC1	TOA	0.48	1.43
EC	TOA	0.54	1.61
Optical EC	Optical transmittance	0.11	0.34

4.4.4 Criteria Gases

Personal and central site gaseous O_3, SO_2, and NO_2 will be collected using Ogawa diffusion badges. The Ogawa badge is a passive sampler containing a coated filter supported by two screens with a diffusion barrier end cap (Koutrakis et al., 1994). The filter substrate will be coated with either sodium nitrate to retain O_3 or triethanolamine to retain SO_2 or NO_2 where the criteria gas stoichometrically reacts with the substrate and is retained.

All of the Ogawa badges are then extracted and the resulting ion quantified using ion chromatography. This methodology has been used by the NERL for almost 10 years and has been highly successful in the passive determination of ambient gas concentrations. Details on detection limits, quantitation limits, blanks and duplicates for these gases in the TACS are shown in Table 4-8. These badges have been shown to be highly successful in the collection of ambient samples, however, low personal exposure concentrations, especially for ozone, often result in concentrations at or below the limit of detection. It is expected that similarly low concentrations may occur in the Detroit homes. Indoor concentrations are expected to be highly dependent upon the participant's use of natural ventilation.

Table 4-8. NO_2, SO_2 and O_3 completeness and detection limits (ppb) from the TACS.

Analyte	Completeness (%)	MDQ	MQL	Blanks			Duplicates		
				N	mean	SD	N	Mean	SD
NO_2	97	0.3	1	3	8.3	0.4	2	0.9	0.5
SO_2	97	7.6	22.8	4	6.3	10.8	2	6.4	8.1
O_3	97	8	24	4	24.1	16.6	2	-1.4	3.4

4.4.5 Nitrate

Nitrate in central site, outdoor, and indoor air samples will be collected using mini denuder samplers. Samples are collected on 15 mm diameter sodium carbonate-coated glass fiber filter at 0.8 LPM. To minimize the effect of acidic ambient gases upon the PM collected on the filter, the sample stream first passes through two sodium carbonate-coated mini-denuders. These samplers have been successfully used as part of the ChemPass Model 3400 Personal Sampling System developed by the Harvard School of Public Health and available through Rupprecht & Patashnick Co., Inc. (Albany, NY).

Nitrate is extracted from the filter samples with distilled deionized water for 30 min. The extract is then analyzed by ion chromatography. The reported method detection limit, 0.26 $\mu g/m^3$, is well below reported ambient air concentrations in Detroit. Precision ranges from 11.3-14.6% for the method based on analysis of duplicate samples.

4.4.6 Carbonyls

Formaldehyde, acetaldehyde, and acrolein will be collected using a passive aldehyde and ketone sampler known as PAKS. This method is a diffusion based passive sampler originally developed for use in the RIOPA study for the analysis of formaldehyde and other aldehydes and ketones. The PAKS method is currently being optimized for the collection and analysis of acrolein while maintaining its effectiveness for the measurement of other carbonyls such as formaldehyde and acetaldehyde. The PAKS method utilizes a C_{18} cartridge coated with dansylhydrazine (DNSH) which reacts with carbonyls to form stable DNSH-carbonyl derivatives. The DNSH-carbonyl derivatives are solvent extracted with acetonitrile and analyzed by HPLC with fluorescence detection. A sampling time period of 24 hours is targeted for this study. Preliminary quality assurance results from its use in the Tampa Asthmatic Children's Study are presented in Table 4-9.

Table 4-9. Performance Results for the PAKS Method from the Tampa Asthmatic Children's Study (48-hr samples).

Analyte	MDL ($\mu g/m^3$)	Analytical Precision	Source	Analytical Recovery (%)
Formaldehyde	0.19	5.4%	lab, field blanks; duplicate samples	101 ± 4
Acetaldehyde	0.46	5.3%	lab, field blanks; duplicate samples	87 ± 3
Acrolein	0.47	9.5%	lab, field blanks; duplicate samples	60 ± 2

4.4.7 VOCs

The method proposed for VOC collection and analysis is currently under development at EPA. It uses passive samplers containing Carbopack X as the collection media and thermal desorption/GC/MS as the analysis method. This method is intended to overcome performance issues for 1,3-butadiene that have been observed with other passive methods. Preliminary testing indicates that the Carbopack X (graphitized carbon black) sorbent will be suitable for the collection of 1,3-butadiene as well as the other listed target VOCs over the proposed 24 hour monitoring period.

Several configurations of the diffusion based passive samplers are being investigated including: Perkin-Elmer stainless steel tubes, SKC Ultra Badges, and a "Monsanto" large surface badge. Primary differences in these devices are effective sampling (or diffusion) rates and the procedure for thermal desorption. Perkin-Elmer tubes exhibit the lowest effective sampling rates (~ 1 mL/min or less) and are directly desorbed using a Perkin-Elmer thermal desorption interface. The SKC badges exhibit a moderate effective sampling rate (~10 to 15 mL/min) and require that the sorbent be transferred from the sampling badge to a Perkin-Elmer tube and then analyzed using the Perkin-Elmer interface. The "Monsanto" badge effective sampling rate has not been determined but is expected to be the highest. Monsanto badges are directly desorbed and injected into the GC/MS using a specifically designed and built interface for these badges.

The three collection devices will be evaluated and the method selected will be based on a number of parameters including sensitivity, recovery, effective sampling rates, precision for the target compounds, ease of use and analysis, ruggedness, and availability of materials. A representative number of collocated canister samples will be employed to assist in the validation of this new methodology.

Typical urban concentrations of 1,3-butadiene are in the range of 0.3 to 1.6 $\mu g/m^3$. The limit of detection for this method is currently being defined but is expected to be in the sub $\mu g/m^3$ range.

4.4.8 Air Exchange Rate Measurements

Air exchange will be measured in each study residence to determine the integrated air infiltration rate during the monitoring period. Residences will be treated as a well-mixed one-compartment model. Permeation devices (emitters) containing a perfluorocarbon compound will be placed throughout each residence 24 hours before monitoring. The perfluorocarbon tracer (PFT) is emitted at a known rate that can be adjusted for temperature. The tracer mixes with the indoor air in the house. Tracer gas concentrations measured in residences will be dependent on the house volume and the rate of outdoor air infiltration. The tracer compound will be collected by diffusion using capillary adsorbent tube samplers (CATS) placed inside the residences for monitoring. One CAT will be placed in the central living area. Samples are collected when the adsorbent tube is uncapped. At the end of each 24-h monitoring period, the samplers will be capped, stored in sealed aluminum cans at room temperature, and shipped to Brookhaven National Laboratory for analysis. The amount of adsorbed PFT is determined by gas chromatography with electron capture detection. The PFT concentration (C) will be derived

from the amount of adsorbed PFT (A), the sampling period (t), and the adsorption rate of PFT by the collectors (r):

$$C(pft) = A/rt.$$

The air infiltration rate (E) will be calculated from the volume of the residence (V), the source emission rate (S) and the number of sources (N):

$$E\ (1/h) = NS\ r\ t/A\ V.$$

These samplers have been shown to be reliable with low blanks and minimal contamination. Recovery of spiked controls is usually close to 100% with 4% precision.

Historical limits of detection for this procedure have been on the order of 0.2 exchanges/h with a precision of ± 10%. Extensive use of this technique in recent NERL panel studies (approximately 1000 measurements), have resulted in a sample QA completeness record (environmental samples above the quantification limit) of over 95%.

4.4.9 Supplemental Central Site Monitoring

Air sampling at the central site monitor will include a suite of monitors to collect additional samples for ultrafines, PAHs, trace metals, and soluble metals. Ultrafine PM will be collected using a SMPS/LASX sampling system. Additional filters will be collected for trace metal analysis, soluble metal analysis, and PAH analysis.

Particulate mass at the central monitoring site will also be collected using a Sierra Anderson dichotomous sampler (or comparable monitor). The dichotomous design provides both fine and coarse measures. The dichotomous sampler draws air at a flow rate of 16.70 LPM through 37 mm Teflon filters. Ninety percent of the air (15.03 LPM) flows through the fine particulate filter and the remaining 10 percent (1.67 LPM) flows through the coarse particulate filter. The dichotomous sampler uses a virtual impactor (region of stagnant air) to segregate the air sample into two fractions. The virtual impactor particle separator accelerates the air sample through a nozzle and then deflects the air at a right angle. Most particles smaller than 2.5 μm (fine fraction) will follow the higher air flow path and collect on a fine particulate filter. Particles between 2.5 and 10 μm (coarse fraction) have sufficient inertia to impact into the chamber below the nozzle and are collected on a coarse particulate filter. Ten percent of the sample air flows through the coarse particulate filter and because of this approximately 1/10 of the fine particles are collected on the coarse particulate filter. Both coarse and fine particle samples will be analyzed for mass, EC, elements, and SVOCs as described in Section 4.4. 1.

Meteorological variables will be collected daily at the central monitoring site. These data include real-time wind speed, wind direction, relative humidity, and temperature for each measurement day. Historic weather data from the central site is available so that common weather regimes can be defined to permit extrapolation of study data. Back-trajectories will be established using the

meteorological data to match personal or residential source findings with known industrial facilities or other pollutant sources.

4.4.10 Supplemental Traffic Counts

Vehicle activity on the road can have profound effects on the concentrations and characteristics of pollutant impacts in the vicinity of the roadway. To account for vehicle activity on temporal variability of the pollutant measurements, daily traffic count data will be obtained from the Detroit transportation planning agency (SEMCOG) and the Michigan Department of Transportation (MDOT) for all available roads in the vicinity of the monitoring sites. An attempt will be made to install traffic count monitors on the nearby major roadways during residential monitoring periods, as available through the MDOT. When direct vehicle count measurements are not available, traffic volumes will be estimated by traffic demand modeling conducted by SEMCOG personnel. Fleet characteristics (percent light-duty and percent heavy-duty) data will also be obtained from SEMCOG for these roadways. When feasible, video recordings will be conducted on all major roads in the vicinity of the monitoring sites. The video will be analyzed to determine flow rates and fleet mixes on the roadway during sampling events. Software will be used to interpret the video images for eight vehicle class categories (including light, medium and heavy duty diesel and gasoline vehicles).

4.5 Survey Instruments

Five questionnaires will be developed and submitted to the Office of Management and Budget (OMB) for approval. These include a Participant Characteristic Survey, a Technician Survey collecting data on household characteristics and local ambient sources, a Daily Follow-up questionnaire concerning that day's activities, particularly those related to sources as determined from a real-time examination of the continuous MIE results, and a Time Activity Diary that the participant carries with him and fills out regarding his location and activities (Table 4-10). The draft questionnaires and survey instruments are attached in Appendix D. These instruments have been developed as a result of field use in a series of EPA human exposure panel studies. The Time Activity Diary and the Technician Surveys are similar documents used in previous studies. These are scannable forms which allows fast recovery of the raw data and improvements in the data validation. These documents and the corresponding data have been shown to be practical, low burden, and directly applicable to the stated goals of this project. The Daily Follow-up Questionnaire is now in a computer assisted survey format to reduce data collection errors and decrease data recovery time. Evaluation of the survey instruments from the Tampa Asthmatics Children's Study version has resulted in additional improvements in the form that should further improve data quality (reduced missing entries, increased valid entries, and consistent responses).

Table 4-10. Survey Instruments

Instrument Type	Administered	Information Gathered
Participant Characteristic Survey	Completed at the time of informed consent by study technician	Basic information such as the age, height, weight, and occupational status of the participant
Technician Surveys (2)	At start of monitoring period each season; completed by technician observation	Housing location, building characteristics, ventilation, cooking/fuel characteristics, room characteristics, local ambient sources
Daily Follow-up questionnaire	Daily at end of monitoring period; technician administers to participant	Potential sources (i.e., cooking, smoking, cleaning) in home each day, ventilation characteristics
Time Activity Diary	Participant completes diary throughout the day; technician reviews and verifies entries daily at the end of the monitoring period. Output from the personal nephelometers will be downloaded each day; participant will be queried about locations and activities during times that show high PM exposures.	Information on participant's location, activities, and proximity to smoking or cooking, recorded for every 15 minutes

4.6 Pilot Study

A 9-person pilot study was performed in Tampa, FL. Households were monitored for four consecutive days using many of the techniques that will be employed in the DEARS. (Certain new methods such as those for the VOCs and SVOCs will be extensively tested in the laboratory and to some degree in the field before adopting them for use in the DEARS.) The Tampa study data are still being analyzed. However, the methods have been evaluated and found for the most part to be well suited for use in field studies. A limited period (2-3 days) of technician field training will be employed at the start of the study that will involve collection of field samples. This will provide the RTI, International staff the opportunity to become fully familiar with any of the new sample collection methodologies employed since their last EPA assignment (e.g., new survey questionnaires).

5.0 STATISTICAL ANALYSIS PLAN

Our analysis approach will be tied to the objectives discussed in Section 2 above. That is, univariate statistics to compare personal, indoor, residential outdoor and central site concentrations of the target pollutants, together with testing to if determine differences exist between locations. Cross-sectional correlations between types of measurements (e.g., personal vs. central site) will be determined, as will correlations among target pollutants within one type of measurement. Longitudinal correlations (e.g., personal vs. indoor, indoor vs. outdoor, etc.) will be determined for each subject across the 5 days per season, and, if appropriate, across the full 10 days covering both seasons. The infiltration factor governing the fraction of outdoor aerosol or air toxic contributing to the indoor concentrations will be determined using one of several methods (See below).

Following elemental and chemical analyses of the speciated compounds, source apportionment models such as PMF and/or UNMIX will be run in an attempt to identify the major source types affecting exposure using personal PM and SVOC measurements. Relationships between personal exposure and indoor/outdoor concentrations identified from the study results will provide information needed for development of predictive models of exposure such as NERL's Stochastic Human Exposure and Dose Simulation (SHEDS) models for PM and air toxics. Exposure model algorithms and inputs for PM constituents and air toxics will be improved and refined based on the DEARS measurements and data analysis results.

5.1 Preliminary analysis of existing monitoring data

5.1.1 Sources of monitoring data

PM, air toxics, and related environmental data collected from recent (non-DEARS), measurements will be analyzed to strengthen the current study design while awaiting OMB study clearance. Data sets included in this analysis will include those from Wayne County air toxics sites, select Detroit Air Toxic Pilot Study measurement and potentially data from local academic research groups (e.g., such as that of Gerald Keeler, University of Michigan). Analyses performed upon this data (simple univariate statistics as well as more sophisticated factor analysis) will be used to strengthen the ultimate selection of census tracts incorporated into the participant recruitment process by helping to further establish the dominant pollutant source(s) in a given area. It will also serve to help guide any additional measurement needs prior to field data collections (e.g., the value of a specific VOC relative to cluster analysis).

5.2 Analysis of study data

Univariate statistics will be employed for the personal, indoor, residential outdoor, and central community site data. Assuming lognormal distributions, the geometric means of the different sites, population groups, and types of samples (e.g., indoor vs. outdoor) will be compared using Student's t test or other appropriate metric. Non-parametric tests will be used if the distribution is not lognormal.

Correlations (both Spearman and Pearson) of personal exposure and indoor concentrations with the residential outdoor and central site data will be performed. A mixed-effects regression model will be performed to identify the influence of community sources on indoor and personal exposures, the infiltration factor as a function of measured air exchange, and the attenuation factor affecting personal exposures based on activities.

Using the PM, gases, carbonyls, VOC, elements, and SVOC data, a source apportionment model or models (PMF and/or UNMIX) will be run to identify and quantify the contributions to and uncertainties associated with ambient and indoor air of various types of sources, including diesel, gasoline exhaust, possibly gasoline vapor, stationary source combustion, resuspended crustal material, and other major sources. Two ways to extend the types of models analyzed are to use transformed variables and/or non-identity link functions in the generalized linear models.

The data analysis plan is summarized in Table 5-1 where each study question is restated, the data that will be available to address the objective are summarized, and the data analysis approach is given.

Data analysis will occur in four different phases.

Phase 1. This phase will focus primarily on calculating descriptive statistics (i.e. means, standard deviations, etc.) along with frequency histograms and box plots of personal, indoor, outdoor, central site measurements for $PM_{2.5}$, PM_{coarse}, gases, elements, VOCs, SVOCs, and EC-OC and the other pollutants. This will serve not only to summarize the data but provide an exploratory approach to look for anomalies in the data as well. In addition to determining univariate summary statistics, time series plots of the results will also be used for preliminary analysis.

Relationships between paired daily personal PM exposures and indoor, outdoor, and ambient concentrations will be evaluated for each metric and participant using simple linear regression, and Pearson correlations. For these analyses, data will be stratified by season and geographical location. Ratios, xy-plots along with correlation tables will be used to examine potential relationships.

General linear and mixed models will be used to determine the influence of activity patterns and housing characteristics on relationships of residential indoor/outdoor particle/gas concentrations to those measured at the central site monitor. Information from these analyses will be used to identify important variables for possible inclusion in linear models. Random coefficients models will not be appropriate since individuals selected for the study are not chosen at random; however data on subjects can be included as fixed effects in the model. General linear mixed models will be use to account for possible correlations in residuals that result from the repeated measurements. Homogeneity of variance will be examined before pooling the data for analysis using general linear models**.**

Table 5-1. Data analysis plan for the DEARS.

Analysis Objective	Measurements Required	Proposed Analysis	Analysis Results and Interpretations
1. Determine the associations between concentrations measured at central site monitors and outdoor residential, and indoor residential and personal exposures for selected air toxics, PM constituents and PM from specific sources.	Personal, indoor, outdoor, and central site measurements for $PM_{2.5}$, PM_{coarse}, elements, air toxics, and other pollutant variables. Stratified by site, season, housing stock, geographical location, and primary source influence.	Univariate statistics on personal exposures, indoor, outdoor, and central site data by site, season, subpopulation. Box plots, histograms, time series plots, and other preliminary and exploratory data analyses and graphics methods. Linear regressions, correlations (Pearson/Spearman), ratios between personal, indoor, outdoor, central site data.	Box plots and histograms show visual evidence of skewness and other departures from normality, relative shapes, central tendencies and dispersions of the measurements collected under different spatial and temporal conditions. Cartesian xy-plots show relative scatter and potential linear relationships, and correlations between indoor, outdoor and ambient data. Differences between mean values hypothesized to be equal under the null hypothesis (H_o) and between slopes/intercepts or correlations (assumed to be zero) will be tested for statistical significance ($p < .05$) using appropriate statistical tests.

2. Describe the physical and chemical factors that affect the relationship between central site monitors and outdoor residential and indoor residential concentrations, including those that affect ambient source impacts.	Covariate measurements consisting of meteorology data, community sources, air exchange rates, house structure, house ventilation parameters, indoor sources, participant locations, participant activities, real-time personal, indoor and outdoor PM data, elements, and air toxics.	General linear and/or mixed models will be used to examine the effects and influences of external sources of variation on personal exposure relationships with indoor/outdoor and central site measurements.	Significant ($p < .05$) differences in linear slopes and intercepts due to subjects, sites, seasons, or uncontrolled external influences hypothesized (under H_o) to be zero will be determined by appropriate statistical tests. Tests for positive auto-correlation between repeated measurements resulting in under-estimation of slopes in linear relationships will be dealt with using SAS mixed model analysis and/or testing at more conservative p-levels (e.g. by setting $p < .01$).
3. Identify the human activity factors that influence personal exposures to selected PM constituents and air toxics.	Personal, indoor, outdoor, and central site measurements for $PM_{2.5}$, PM_{coarse}, elements, air toxics, and other pollutant variables. Stratified by site, season, housing stock, geographical location, and primary source influence. Time activity and household surveys.	General linear models will be used to establish relationships following integration of exposure and time activity databases.	Results will be based on least squares and maximum likelihood methods to fit linear models and test the statistical significance ($p <.05$) of the difference between slopes that are hypothesized to be equal under H_o regardless of the individual activity patterns.

4. Improve and evaluate models used to characterize and estimate residential concentrations of and human exposures to selected air toxics, PM constituents, and PM from specific sources.	Personal, indoor, outdoor, and central site measurements for $PM_{2.5}$, PM_{coarse}, elements, air toxics, and other pollutant variables. Stratified by site, season, housing stock, geographical location, and primary source influence. Time activity and household surveys.	Development of models, such as the SHEDS-PM, using actual field data as primary inputs.	Validation, maintenance, and fit of population exposure models using output from emissions-based atmospheric dispersion models will be determined on the basis of statistical significance ($p < .05$) of changes in (R^2); the percent personal exposures variation explained as a result of the of the reduction in pollutant emissions.
5. Investigate and apply source apportionment models to evaluate the relationships for PM from specific sources and to determine the contribution of specific ambient sources to residential concentrations and personal exposures to PM constituents and air toxics.	Personal, indoor, outdoor, and central site PM measurements. Incorporation of VOC, SVOC, carbonyl, elemental and other speciated data.	Mass-balance/Source apportionment models such as UNMIX or PMF.	Source impacts and contributions will be quantified with effective variance least squares chemical mass balance results. The most probable combination of sources that significantly account for percent variation of mass explained will be determined interactively by adding or subtracting variables in multiple regression and/or by changing the number of principal components in factor analysis.

6. Determine the associations between central site concentrations of criteria gases (O_3, NO_2, and SO_2) and personal exposures for these gases as well as personal exposures to air toxics, PM constituents and PM from specific sources.	Personal, residential and central site criteria gas concentrations. Personal, indoor, outdoor, and central site PM measurements along with VOC, SVOC, carbonyl, elemental and other speciated data.	Univariate statistics on personal exposures, indoor, outdoor, and central site data by site and season. Box plots, histograms, time series plots, regression analyses, other preliminary and exploratory data analyses and graphics methods. Linear regressions, correlations (Pearson/Spearman), ratios between personal, indoor, outdoor, central site data. Mixed models will be used to examine the relationships between concentrations and exposures of criteria gases and $PM_{2.5}$, PM constituents, and air toxics.	Box plots and histograms show visual evidence of skewness and other departures from normality, relative shapes, central tendencies and dispersions of the measurements collected under different spatial and temporal conditions. Significant correlations between indoor, outdoor and central site data will be used to determine the important parameters for mixed model anlaysis. Differences between mean values hypothesized to be zero under the null hypothesis (H_o) and between slopes/intercepts and correlations will be tested for significance ($p < .05$) using appropriate statistical tests. Tests for positive auto-correlation between repeated measurements will be tested using appropriate statistical tests.

Phase 2. To achieve adequate predictions for indoor concentrations, factors affecting outdoor levels must be considered. The effects of different site locations, local weather conditions, traffic patterns, and local outdoor sources along with air exchange rates can be used to categorize the data. T-tests can be used to determine if homes in categories with sources are different from homes with no direct influence from known outdoor or indoor sources. General linear and mixed models will be used to determine the influence of activity patterns and housing characteristics on relationships of personal particle/gas exposures to indoor/outdoor concentrations. Information from these analyses will be used to identify important variables for possible inclusion in linear models. Random coefficients models will not be appropriate since individuals selected for the study are not chosen at random; however subjects can be included as fixed effects in the model. General linear mixed models will be used to account for possible correlations in residuals that result from the repeated measurements. Homogeneity of variance will be examined before pooling the data for analysis using general linear models**.**

Phase 3. Validated data and information gained in phases 1-2 will be incorporated into the PM human exposure modeling development. This will be an ongoing and iterative process over the course of the full study period. New datasets will be incorporated as they become available and the model refined at each step. Existing models, such as the SHEDS-PM, will be used as the foundation of the initial effort. A goal of this phase will be to develop a model that will be versatile and translatable to other metropolitan sites.

Phase 4. Data obtained in phases 1-2, along with original environmental measurements will be used to perform source apportionment modeling. The goal of this phase will be to establish the dominant sources that impacted personal, residential and central site-based measurements and the contribution of each primary source upon each spatial setting. Modeling tools such as UNMIX and PMF will be used to support this phase.

5.2.1 General Linear Models Analysis

The specific type of statistical analysis to be performed on the data is dictated by the objectives of the study, and the study design. One fundamental objective of the study is to examine the relationships (with respect to mobile and stationary sources), between residential (household) PM/gas measurements and central site concentrations of PM/gas over a 5-day period (or a 10-day period covering two seasons) for each of the 120 homes monitored. Assuming this is a linear relationship, these relationships can be expressed in terms of a simple linear regression model as follows:

$$Y_{ij} = \alpha_i + \beta_i X_j + \varepsilon_{ij} , \qquad (1)$$

$i = 1,2, ...,120 \quad j = 1, 2, ...10$ days,

where

Y_{ij} = personal PM/gas measurement for the i^{th} household on the j^{th} day ,
α_i = intercept for the i^{th} household,
β_i = slope for the i^{th} household,
X_j = either the indoor, or outdoor, or central site PM/gas concentration on the j^{th} day, and
ε_{ij} = residual error (random) for the i^{th} household on the j^{th} day.

This basic regression model will be fitted for each individual participant, home, and for each of the PM/gas metrics. The coefficient of determination (R^2) along with the p-values to examine the significance of the intercepts and slopes, will be calculated for each relationship. Measurements on the same person taken sequentially and close in time may be correlated. This results in a correlation between residuals on adjacent days. This will be accounted for in the error variance matrix and will primarily involve first order correlation ρ, between residuals one day apart, with correlations of ρ^2, ρ^3, etc. for residuals 2, 3, etc. days apart. Ignoring a positive autocorrelation will lead to an intra-personal variance that is too large. In general (see discussion below), the variance between groups of participants (i.e., inter-personal variance) will be underestimated and the variance across time will be overestimated. Correlated outcomes must be addressed to obtain valid estimates.

As discussed earlier, there will be a total of at least 120 households being measured repeatedly for 5 days in each of 2 seasons. Along with possible seasonal effects on the relationships, other factors that may or may not statistically affect the relationships are related to the categories in which the participants will be placed based source impacted categories. Other factors included in the study design that possibly could affect the relationships are distance from source, meteorology, and residential type (apartment, stand-alone homes). As illustrated above, the regressions will be calculated separately for each individual participant within each season. Statistical comparisons of slopes and intercepts within and between distance from sources, seasons, housing types, etc. will be performed by comparing individual results on a case-by-case basis within groups and by comparing household group averages of intercepts and slopes using pooled within-group estimates of intra-household variation as the basis for statistically testing and measuring the separation between group means. A more general linear model that combines both categorical variables as well as continuous variables into a single comprehensive model and allows for multiple comparisons with the aid of analysis of variance techniques is given as follows:

$$Y_{ijkl} = \mu + S_i + P_j + X_k + S*P_{ij} + S*X_{ik} + P*X_{jk} + S*P*X_{ijk} + E_{ijk} , \qquad (2)$$

$$i = 1,...,2 \qquad j = 1,.....,120 \qquad k = 1,.....,5$$

where
Y_{ijkl} = personal PM/gas exposure for the j^{th} participant on the k^{th} day during the i^{th} season,
μ = overall mean,
S_i = effect of i^{th} season (fixed),

P_j = effect of j^{th} household (fixed),
X_k = PM/gas measurement at indoor, or outdoor, or central site on the k^{th} day
$S*P_{ij}$ = effect of interaction of the i^{th} season with the j^{th} participant (fixed),
$S*X_{ik}$ = effect of the i^{th} season on the slope (fixed),
$P*X_{jk}$ = effect of the j^{th} participant on the slope fixed),
$S*P*X_{ijk}$ = effect of the i^{th} season and j^{th} participant on slope (fixed), and
E_{ijk} = random error on the k^{th} day during the i^{th} season for the j^{th} participant.

Using SAS, appropriate sums of squares will be generated to test hypotheses linked to the study objectives. For example, the term P*X provides an additional sum of squares due to the different regression coefficients for the individual homes. Since individuals are not chosen at random, the slopes for X_k are considered to be fixed effects. Therefore, a random coefficients analysis using mixed models is not appropriate. However a mixed model procedure will be used to account for intra-personal correlations due to repeat measures across time within seasons. Homogeneity of variance across seasons and households will be examined and appropriate adjustments and assumptions made.

The test corresponding to the sum of squares provided by the source of variation **X**, is a test of the significance of the regression of PM/gas personal exposure (Y) onto X (indoor, outdoor, or central site concentrations) ignoring groupings due to seasons or households. Other tests include a test of whether or not seasons significantly alter the regression coefficients, a test to detect inter-personal variability, and a test on proximity to mobile or stationary sources. As stated above, homogeneity of variance should be examined as well as the possibility of autocorrelation among residuals due to repeat measurements on the same individual over time. Since relationships will be determined for central site versus indoor, central site versus outdoor, central site versus personal, indoor versus outdoor, and indoor versus personal, the results of these relationships can be compared.

An over-parameterized linear model is require to further examine the effects of household grouping by potential source impacts, residential types, proximity to sources, and meteorology and the effects these factors have on the linear relationships as well as seasonal effects, and various interactions between factors in a single analysis. These additional groupings apply to the original 120 households so the size of the study is not increased. This may lead to an imbalance in the design. The number of males versus females or the number of households from a particular housing stock will be determined during recruitment. A general linear model that incorporates these categorical groupings as main effects and 2-way interactions along with the continuous variable X (i.e. indoor, outdoor, and community PM/gas) and its interactions with the main effects and 2-way categorical interactions is given below.

$$Y_{ijklmn} = \mu + S_i + C_j + G_k + H_l + R_m + X_n + S*C_{ij} + S*G_{ik} + S*H_{il} + S*R_{im} + C*G_{jk} + C*H_{jl} + C*R_{jm} + G*H_{kl} + G*R_{km} + S*X_{in} + C*X_{jn} + G*X_{kn} + H*X_{ln} + R*X_{mn} + (\text{all 3-way interactions involving X}) + \text{Residual},$$

i = 1,..,2 j = 1,4 k = 1,2 l = 1,2 m = 1,2 n = 1,...,5,

where

Y_{ijklmn} = personal PM/gas exposure during i^{th} season, in j^{th} cohort, for k^{th} housing stock, with l^{th} source proximity, m^{th} residential status, on n^{th} day,
μ = overall mean
S_i = effect of i^{th} season,
C_j = effect of j^{th} cohort,
G_k = effect of k^{th} housing stock,
H_l = effect of l^{th} source proximity,
R_m = effect of m^{th} residential type,
X_n = indoor, outdoor, or central site PM/gas measurement on the n^{th} day,
$S*C_{ij}$ = effect of interaction between i^{th} season and j^{th} cohort,
$S*G_{ik}$ = effect of interaction between i^{th} season and k^{th} household group,
$S*H_{il}$ = effect of interaction between i^{th} season and l^{th} source proximity,

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(plus other 2-way and 3-way interactions with the continuous variable X), and

Residual = variation due to all higher order interactions + model error.

Variations of this more general model could include other factors or covariates such as spatial groupings of participants or temperature ranges to group measurements instead of seasons or activity patterns. Using SAS, model terms such as $R*X_{mn}$ will generate appropriate statistics for estimating and testing different regressions of Y onto X for different groups. For this particular term, the relationships between central site exposures and either indoor, outdoor, or personal PM/gas measurements will be compared for different residential types. The effect of different factors on relationships can be examined by testing the change in R^2. To do this the amount of variation in the Y variable accounted for by fitting the full model is compared to the amount of variation in the Y variable accounted for by fitting a reduced model by leaving out the term in question. The contribution of these "left out" effects to the full model is determined statistically with the F-distribution. Graphical plots and visual inspection of the trends and outliers in the data and model residuals will also be used to evaluate the model fit.

This approach applies to any of the above linear models. Statistical significance is a necessary but not sufficient condition for practical significance. Statistical significance indicates that something other than chance is operating, but does it really matter from a practical standpoint? Research studies such as this one with large sample sizes often do find statistically significant results that are not of practical importance. Since R^2 is not affected by sample size it should also be reported as a measure of adequacy in fitting each term in the linear model.

The summary in Table 5-1 is included to ensure that the data collection plan and data analyses are linked to study objectives and hypotheses. Analysis of the study data will not necessarily be limited to those described in the Table. As in any study, additional analysis needs and methods will be identified as the study is performed and the data are examined.

5.3 Exposure Modeling

The measurement data collected during this study and the results of the data analysis described above will provide critical new information needed for further development of population exposure models for PM and air toxics. Population exposure models provide a method for estimating exposures for a population of interest when no measurements or limited measurements of personal exposure exist. These models are useful tools for predicting the range in exposures across a population and the likelihood of exposures above a particular level. When output from an emissions-based atmospheric dispersion model is used as input, population exposure models are also useful for estimating the reductions in exposures that would occur as a result of reductions in pollutant emissions.

The development of algorithms and databases needed to predict exposures requires as much information on the factors that influence exposures as is available. The DEARS will provide new information on the factors that influence the relationships between outdoor and indoor concentrations of and human exposure to PM constituents and air toxics. This information will be used to improve and refine the algorithms and input databases for population exposure models. The measurements of personal exposures collected during the DEARS will also allow for evaluation of the exposure model predictions. The outcome will be refined and evaluated exposure models for PM and air toxics that are available for application to other metropolitan areas.

NERL's Stochastic Human Exposure and Dose Simulation (SHEDS) models use a probabilistic approach to predict the distribution of exposure and dose for a specified population. The SHEDS models for PM and air toxics estimate this distribution by simulating the time series of exposure and dose for individuals that demographically represent the population of interest. US Census data are used to build the simulation population, and human activity pattern data are assigned to each simulated individual to account for the way people interact with their environment. Pollutant concentrations in the microenvironments that people spend time in (e.g., home, car, office, school, restaurant, etc.) are calculated based on relationships between central site outdoor and indoor or in vehicle concentrations obtained from measurement study data. Each individual's exposure and dose profile is estimated from the time spent in each location, the concentration in that location, and the activity-specific inhalation rate while in that location. Daily-averaged exposure and dose for each individual are calculated and combined to provide a distribution of exposure and dose for the population. Statistical methods for incorporating both variability and uncertainty in the model input parameters are utilized to obtain the predicted population distribution and the uncertainty associated with the predicted distribution.

The exposure modeling component of the DEARS will include three phases. Initial model development will occur prior to the completion of the measurement study and case study applications will be performed with the SHEDS models using available data on PM components from the Detroit Speciation Trends Network (STN) sites and on air toxics from the Detroit Pilot Air Toxics Network sites. Results from NERL's DEARS will then be used to improve and refine the algorithms and input databases. In this second phase, the exposure model predictions for the DEARS will be evaluated against the measurements of personal exposure for PM components and air toxics. Lastly, further model development will involve incorporation of source apportionment results into the exposure models so that the SHEDS models will be able to predict population exposures to specific sources.

5.4 Source Apportionment Modeling

Data from the Particle Total Exposure Assessment Methodology (PTEAM) study and the 1998 Baltimore Particulate Matter Epidemiology-Exposure Study (BPMEES) were analyzed with advanced receptor models to identify and quantify the sources of particulate matter collected on personal, indoor, and outdoor samples. In addition, the planned receptor model development and analysis of the Research Triangle Park Exposure Study data (NERL) will evaluate the approach that will be used in the DEARS. A discussion of these studies and the modeling approach are given in Appendix C.

5.4.1 Detroit Receptor Modeling

A modified 2-way factor analytic model will be used to analyze the DEARS data. The Multilinear Engine technique (the program ME-2) will be used, incorporating the following additions to the standard non-negative PMF 2-way model:

1. Divide the factors in two groups, so that there is a group of "indoor only" factors that only explain indoor and personal data, not outdoor or ambient. (This technique was used in the Baltimore study.)
2. The factor analytic model should not attempt to explain irregular non-recurrent features of the data. Elements like Cu and Zn displayed such behavior in the Baltimore study, but there may be other such elements. Such elements should be fitted with a dynamic reweighting according to the following scheme:
    - data points with residuals more positive than a chosen cutoff limit (+0.5 sigma, say) are considered "irregular" and downweighted analogously to the PMF outlier downweighting scheme (Paatero 1999)
    - all other data points would be given the standard PMF/ME-2 weighting.

This scheme will fit regular behavior of the chosen elements in the normal way. However, stray positive excursions will not influence the factorization. Such excursions will be inspected separately, outside of the factor analytic model. The purpose of the suggested technique is that a reasonably good fit is achieved with a small number of factors. Otherwise, additional factors would be needed in order to fit the irregular behavior, and these extra factors would introduce undesirable rotational ambiguity in the factor analytic model. It should be emphasized that the

irregular positive excursions are not considered "bad data." Instead, it is recognized that such data values cannot be successfully analyzed within a factor analytic framework.

3. Implement closure constraints in the model so that the sum of all identified mass (including oxygen etc. in certain compounds) cannot exceed the mass coefficient in any factor. (This technique was used in the Baltimore study.)
4. Include constraints that prevent some specific factor(s) from explaining certain individuals/regions/times. This approach depends on the interpretation of factors and may not be possible at all. An example is a localized source that only affects individuals living in a certain suburb. Such a factor should be excluded from explaining individuals who do not live (or visit) in the suburb in question. The use of constraints should be coupled to supplementary information about the activities of the participants, such as traveling by car, being indoors only, vacuuming/cooking/etc, wherever possible.

Receptor models will be developed to determine and quantify the sources of PM and air toxics in DEARS samples (community, outdoor home, indoor, and personal) at the central site, homes near roadways (< 50 m), and homes farther away from roadways (> 150 m). In addition, the relationship (correlation and attenuation) between central site source contribution estimates and personal exposure to the central site source contributions will be determined.

6.0 QUALITY ASSURANCE

A Quality Assurance Project Plan (QAPP) will be prepared that describes quality assurance goals and the methods that will be used to meet these goals. The plan will address both the overall DEARS analysis plan as well as the individual sample collection and analysis procedures that will provide the data. Critical elements of the Quality Assurance Project Plan include (U.S. NAMS 005/80):

- Project description
- Project organization
- QA objectives
- Sampling procedures
- Sample custody
- Calibration frequency and procedures
- Analytical procedures
- Data reduction, validation and reporting
- Internal quality control checks
- Performance and system audits
- Preventative maintenance
- Routine procedures to assess data quality
- Corrective actions
- QA reports to management
- Standard Operating Protocols (sops) for sample collection and analysis

The Quality Assurance Plan prepared for the RTP PM Exposure Panel Studies will serve as the basis for this plan. Selected components will be revised and/or updated including the project description, sampling populations, cited instruments, data quality objectives, analytical procedures and the calibration frequency of listed equipment. The plan will be reviewed and revised as needed before measurements are performed. All work will be conducted as described in the approved QA Plan.

In addition to developing an approved QA Plan, NERL will be responsible for demonstrating compliance to the plan. Field and laboratory audits will be conducted during selected portions of the work. For these audits, NERL QA specialists will review and observe EPA and contract field staff during sample collection, calibration of test equipment, documentation of sampling/field efforts, and data reduction and reporting, etc. Formal audit reports will be prepared and released to the NERL-HEASD Quality Assurance Director. A QA report will be prepared by the NERL QA staff at the end of the project

7.0 MANAGEMENT

7.1 Schedule

Table 7-1 reports the anticipated timeline of the study and pertinent events. In general, field activities are anticipated to begin in the summer of 2004. This will be followed by a winter monitoring program in early 2005 and then a repetition of summer and winter sampling for two additional years. Laboratory and statistical analysis of seasonal samples/data will be performed throughout the initial three years of the field study and continuing until goals 1-6 have been achieved (approximately the fall of 2007). Databases will be expanded as additional validated data becomes available.

Table 7-1 Anticipated DEARS Timeline

Task	Projected Date
Complete study design peer review	July 2003
Receive OMB approval for data collection	April 2004
Receive IRB and EPA study approval involving human subjects	May 2004
Start first summer field monitoring session	June 2004
Start first winter field monitoring session	January 2005
Begin development of validated datasets	May 2005
Perform initial analyses	June 2005
Start second summer field monitoring session	June 2005
Start second winter field monitoring session	January 2006
Start third summer field monitoring session	June 2006
Start third winter field monitoring session	January 2007
Complete all laboratory analyses	December 2007
Validate all data and datasets	December 2008
Complete all analyses and report summary findings	December 2009

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APPENDIX A: SOURCE-RELATED EXPOSURE FINDINGS

Janssen et al. (2001) studied the exposure to traffic related air pollution of children attending schools near motorways. 24 schools within 400 m of motorways in the Netherlands were monitored for $PM_{2.5}$, NO_2, EC (reflectance) and benzene. Particles and benzene were averaged over school hours only during one week. NO_2 was averaged over a full week. Each school was measured 5-10 times over 16 weeks. Main findings were that fine particles and EC significantly increased with increasing truck traffic and significantly decreased with increasing distance from the motorway. NO_2 significantly increased with car traffic. Soot, benzene, and NO_2 had a 2.5-fold range, but $PM_{2.5}$ had only about 1.5-fold.

Roorda-Knape et al. (1998) measured air pollution from traffic in city districts near major motorways. Ambient and indoor measurements were made in 12 schools in six cities. Outdoor measurements of PM_{10}, $PM_{2.5}$, black smoke (BS), benzene, and NO2 were made in two cities; NO2 alone in four more cities. Indoor measurements included weekly averages of PM_{10}, benzene, and NO_2**.** The main findings from this study were that EC and NO_2 declined with increasing distance from roadways up to 100-150 m. No distance effect was noted for particles or benzene. PM_{10} outdoors was 32 $\mu g/m^3$ at the two cities, but 50-165 $\mu g/m^3$ indoors at the 12 schools. BS was 12-15 $\mu g/m^3$ outdoors, 5-20 $\mu g/m^3$ indoors.

Van Vliet et al (1997) studied motor vehicle exhaust and chronic respiratory symptoms in children living near freeways. Ambient and indoor measurements of PM_{10}, $PM_{2.5}$, black smoke, and NO_2 were made in 13 schools within 1000m of major freeways in South Holland. Lung function and chronic respiratory symptoms were ascertained for 1500 children 7-12 years old (1068 usable questionnaires). Home addresses were plotted as distance from roadway. The main result was that girls (but not boys) living less than 100m from the roadways had higher symptoms.

Kingham et al., (2000) studied spatial variations in the concentrations of traffic-related pollutants in indoor and outdoor air in Huddersfield, England. Outdoor and indoor measurements were made in paired homes <50 m and >50 m from roadways. 24-h daily averages of PM_{10}, $PM_{2.5}$, EC by reflectance, benzene, benzo-a-pyrene (Bap) and other PAHs were made over two 2-week periods. A total of 49 homes (23 near, 26 far) took part. No spatial gradients were apparent for any pollutants.

Fischer et al., (2000) studied traffic-related differences in outdoor and indoor concentrations of particles and volatile organic compounds in Amsterdam. Outdoor and indoor measurements were made in paired homes: major streets vs. quiet streets. 24-h daily averages of PM_{10}, $PM_{2.5}$, EC by reflectance, 15 PAHs, and 8 VOCs were collected in 36 homes (18 & 18). Results indicated that outdoor particle levels were only slightly (15-20%) higher in the homes near major streets, but 100% increases for BaP, total PAH, EC, benzene, and total VOC were documented. Indoor levels also increased for the latter except for the VOCs.

Kousa et al., (2002) studied associations between ambient fixed site, residential outdoor, indoor, workplace, and personal exposures to $PM_{2.5}$ in four European cities in the EXPOLIS study. Central-site, back yard, indoor, workplace, and personal measurements were made using a

probability sample of working-age populations (N = 50 in most cities, 200 in Helsinki) in six cities. Measurements were made using 48-h personal PM2.5 (PEMs) with two filters, one for work and the other for non-work time; 48-h microenvironmental monitors (MEMs) at home, outdoors, and workplaces, programmed to run during expected times at home and work; and 24-h monitors at central sites. Results included low personal-ambient correlations (r = 0.25-0.37) for the full Helsinki population; slightly higher personal-ambient correlations (r = 0.34-0.47) for non-ETS exposed population. Indoor-ambient correlations (r = 0.22-0.41) were also low. High correlations were noted between residential outdoor and central-site measurements.

Edwards et al., (2001) reported on VOC concentrations in in EXPOLIS in Helsinki. Outdoor, indoor, workplace, and personal 48-h VOC measurements were made for 183 persons in a probability sample of working-age populations**.** Results showed that 21 of 22 compounds had indoor mean concentrations 2-7 times higher than concurrent outdoor levels. ETS-exposed subjects (40% of total) had twice the BTEX and styrene levels of non-ETS exposed.

Edwards et al. (2001) extended the previous study to identify VOC sources for a subset (N=111) of the non-ETS-exposed participants. Four factors were identified, associated with traffic emissions, cleaning products, emissions from trees (a-pinene), and long-range transport.

Hoek et al., (2002) studied the spatial variability of fine particle concentrations in three European areas PM2.5 and EC were monitored at 40 sites for four 14-day averages. Continuous monitoring at a central site in each of three areas was carried out to remove bias from temporal variation. Annual averages ranged from 11-20 $\mu g/m^3$ in Munich, 8-16 $\mu g/m^3$ in Stockholm, and 14-26 $\mu g/m^3$ in the Netherlands. Traffic-related sites were 17-18% higher than background sites in PM2.5 but 31-55% higher in EC.

Ruuskanen et al. (2001) studied concentrations of ultrafine, fine and PM2.5 particles in three European cities. One site in each of three cities in the Netherlands, Germany, and Finland Was monitored during winter only. 10-minute average measurements were takenof ultrafines, total particle number, PM2.5, and EC.$PM_{2.5}$ was poorly correlated with ultrafines, but well correlated with accumulation mode number concentration. Principle Components Analysis (PCA) indicated two components, one associated with number, the other with mass.

Houthuijs et al (2001) measured 24-hour average concentrations of PM_{10} and $PM_{2.5}$ in 24 study areas in six countries in Central and Eastern Europe. Samples were collected every sixth day. Additional sampling at one or two urban background sties within each area took place for one or two months. $PM_{2.5}$ increased significantly in the heating season (from 18-45 $\mu g/m^3$) with no increase in coarse particles. Within-area spatial variation was much smaller than between-area variation (but the sample size for within-area comparisons was limited).

Van der Zee et al., (1998) characterized particulate air pollution in urban and non-urban areas in the Netherlands Daily measurements of PM_{10}, EC, sulfate, nitrate, ammonium, acidity, elements took place overthree consecutive winters in three urban and three non-urban areas. $PM_{2.5}$ was added in the third year. PM_{10} and EC were 13-19% higher in urban areas; $PM_{2.5}$ was not higher. All elements significantly higher except for Si.

Hoek et al. (2002) found that cardiopulmonary mortality was associated with living near a major road for a Dutch cohort.

Delfino (2002) published a review of epidemiological studies of the relationship between asthma prevalence or morbidity and traffic-related exposures. Delfino concluded that his review "gives the overall impression that asthma, related respiratory symptoms, lung function deficits, and atopy are higher among people living near busy traffic."

Other studies including Shao et al. (2002), Buckeridge et al. (2002), and Brauer et al. (2002) indicate the potential for adverse respiratory outcomes among populations living near roadways. Pearson et al. (2000) associated distance-weighted traffic counts in Colorado on the nearest road to children's residence with increased odds of childhood cancer, including leukemia using a case-control design, although other studies have found no associations (Langholz et al., 2002; Reynolds et al., 2002).

Appendix B: Power calculation

One of the primary objectives of the Detroit study is to evaluate and describe the physical and chemical factors that determine the impact of various ambient sources on outdoor and indoor residential concentrations and the spatial variability of these concentrations in relation to the source locations. As detailed in section 5.2.1, this can be accomplished based on a simple linear relationship in which residential (outdoor or indoor) concentrations (i.e., the dependent y-variable) are regressed onto the ambient concentrations (i.e., the independent x-variable).| The strength of such a linear relationship is measured by R^2 = SSR/SST ,the coefficient of determination, where SSR is sum of squares due to regression, and SST is the total sum of squares in the y-variable corrected for mean. R^2 can be interpreted as the amount of variation in the y-variable accounted for by regression onto the x-variable. For a simple linear regression, R^2 is the square of the correlation between xy pairs. Regressions will be calculated for each cross-sectional group of source and location categories as discussed in section 3.3.2 and shown in Table 3-8 in order to compare the effects of different point sources, mobile sources, and locations on the slopes and intercepts.

Our study calls for a sample size of 5 days/season/ household. The study will be conducted over 3 years; two seasons (winter and summer) per year. Approximately 40 households will be chosen each year giving a total of N = 3*40*2*5=1200 measurements. However, since regressions will calculated for groups of different sample sizes Figure A-1 shows power curves relating N, R^2, and power at the alpha = 0.05 significance level. Each curve shows the probability (i.e., power) of detecting $R^2 > 0$ for different sample sizes. Each value of R^2 represents a different alternative to the null hypothesis of zero. These curves were derived using the noncentral F-distribution in SAS and allowing N to range from 5 to 30 and R^2 from zero to 1.0 at the alpha=0.05 significance level. For example, for a single household with N = 5 days per season, the R^2 for a simple linear regression needs to be at least 0.78 to have a high probability (0.80) of being significantly different from zero at the alpha = .05 significance level. If the results from two seasons within a year can be pooled (N = 10), the R^2 can be as low as 0.50 and still be significant. Rejecting a null hypothesis of a zero slope is equivalent to rejecting a null hypothesis of $R^2 = 0$. Pooling measurements across residences within source groups may provide a larger sample size on which to base regression estimates, however the calculated R^2 will not necessarily increase with more samples, but rather tends to settle down around the population value.

Because of the inherent relationship between linear regression, correlation, and R^2, the relative difference between independent regression slopes can be detected at a specified probability level (power) using the appropriate sample sizes and R^2 values. For example, Table A-2 shows that for a correlation of r = 0.90 (R^2 = .81), a sample size of N = 45 per group is required in order to have a probability of at least .78 of detecting a relative difference of * = ± 20% (± 0.20) between estimated slopes at the *alpha* = .05 significance level. The highlighted probabilities correspond to combinations of sample sizes (n1,n2) and R^2 values (rs1, rs2) that provide a better than 50:50 chance of detecting a specified relative difference at the alpha = .05 significance level. A comparison of intercepts is valid only if the slopes are equal.

Figure A-1

Table A-2. Power of t-test to detect differences of magnitude D(%) between two slopes for two different source groups: under the null hypothesis of no difference and an alpha=.05 significance level

n1=40 n2=80

D(%)	rs1=.49 rs2=.49	rs1=.49 rs2=.64	rs1=.49 rs2=.81	rs1=.64 rs2=.49	rs1=.64 rs2=.64	rs1=.64 rs2=.81	rs1=.81 rs2=.49	rs1=.81 rs2=.64	rs1=.81 rs2=.81
10	0.07	0.07	0.08	0.08	0.09	0.10	0.10	0.13	0.16
20	0.14	0.15	0.16	0.19	0.22	0.25	0.27	0.35	0.45
30	0.23	0.25	0.27	0.34	0.39	0.44	0.50	0.62	0.74
40	0.34	0.37	0.39	0.50	0.56	0.61	0.71	0.82	0.90
50	0.45	0.48	0.50	0.64	0.70	0.75	0.86	0.93	0.97

n1=45 n2=30

D(%)	rs1=.49 rs2=.49	rs1=.49 rs2=.64	rs1=.49 rs2=.81	rs1=.64 rs2=.49	rs1=.64 rs2=.64	rs1=.64 rs2=.81	rs1=.81 rs2=.49	rs1=.81 rs2=.64	rs1=.81 rs2=.81
10	0.06	0.07	0.07	0.06	0.08	0.09	0.07	0.09	0.13
20	0.11	0.13	0.16	0.13	0.17	0.23	0.15	0.22	0.35
30	0.19	0.23	0.27	0.23	0.31	0.41	0.29	0.42	0.62
40	0.28	0.34	0.40	0.36	0.47	0.58	0.45	0.63	0.83
50	0.38	0.45	0.52	0.49	0.61	0.73	0.61	0.79	0.94

n1=45 n2=45

D(%)	rs1=.49 rs2=.49	rs1=.49 rs2=.64	rs1=.49 rs2=.81	rs1=.64 rs2=.49	rs1=.64 rs2=.64	rs1=.64 rs2=.81	rs1=.81 rs2=.49	rs1=.81 rs2=.64	rs1=.81 rs2=.81
10	0.06	0.07	0.08	0.07	0.09	0.10	0.08	0.11	0.15
20	0.13	0.15	0.17	0.16	0.20	0.25	0.20	0.28	0.41
30	0.22	0.25	0.29	0.29	0.36	0.44	0.38	0.52	0.70
40	0.32	0.37	0.41	0.43	0.53	0.62	0.57	0.74	0.88
50	0.43	0.49	0.53	0.58	0.68	0.76	0.74	0.88	0.96

Appendix C: Source Apportionment

Sampling Designs

The PTEAM study was the first large-scale probability-based study of personal exposure to particles conducted in fall of 1991 in Riverside, California (Özkaynak et al., 1994a - b). Personal exposure to PM_{10} was measured for 178 participants selected for study based on socioeconomic stratification, after examining their screening interviews. Sampling continued during 49 days: from September 22 to November 9, 1991. Each participant wore personal exposure monitors (PEM) for two consecutive 12-hour periods. The PEM design is discussed in detail by Özkaynak et al. (1996). PM_{10} and $PM_{2.5}$ samples were also collected with stationary indoor monitors (SIM) and stationary ambient monitors (SAM) at each home for every observation period. This resulted in 10 samples per household (day and night samples from PEM_{10}, SIM_{10}, $SIM_{2.5}$, SAM_{10} and $SAM_{2.5}$). A central outdoor site was maintained during the entire period, where PM was measured by two high-volume PM_{10} samplers , two dichotomous PM_{10} and $PM_{2.5}$ samplers, one PEM, and one SAM. Following each of the two 12 hour monitoring periods, the participants answered an interviewer-administered questionnaire concerning their activities that might involve the exposure to increased particle level (nearby smoking, cooking, gardening, etc.) and locations during monitoring time. All of the filters were weighed on-site in a van with controlled temperature, humidity and protection from vibration. Elemental concentrations were determined by X-ray fluorescence (XRF). Fourteen elements (Al, Si, S, Cl, K, Ca, Ti, Mn, Fe, Cu, Zn, Sr, Br, Pb) were present in measurable quantities on a majority of the filters.

In a recent study by EPA, Williams et al. (2000a, 2000b) evaluated the relationship between $PM_{2.5}$ mass measured at a stationary outdoor community location in central Baltimore county (Towson, Maryland) and personal exposure observations from July 26 to August 22, 1998 at an 18-story retirement facility 11 km away. The study was designed to minimize the microenvironmental activities that were previously shown to contribute to elevated exposures, and to evaluate $PM_{2.5}$ exposure of susceptible populations that might have higher risk factors than other segments of the population.

Daily personal and every other day apartment $PM_{2.5}$ samples were collected on 37 mm Teflon filter media using inertial impactor Personal Exposure Monitors (PEMs) manufactured by MSP Inc., sampling at 2 liters per minute. The PEM samplers were refurbished daily and randomly reassigned to avoid any potential bias. PEM samplers were also collocated with a Versatile Air Pollutant Sampler (VAPS) at the indoor, outdoor, and central community sites (Clifton Park Golf Course). The VAPS is a modified dichotomus sampler which collects $PM_{2.5}$ on both Teflon and quartz filter media (15 LPM), and also collects concentrated coarse ($PM_{8.0}$ - $PM_{2.5}$) particulate matter on a Nucleopore® filter (2 LPM) (Pinto et al., 1998). Outdoor sampling at the 18-story retirement facility was conducted with another VAPS on the rooftop of an attached three-story retirement facility. Centralized indoor sampling was conducted with a third VAPS in an unoccupied fifth floor apartment of the retirement facility. The windows in this location remained closed, but the apartment door was kept open to the facility's central hallway.

The Teflon filters from the VAPS and PEM samplers were analyzed using XRF. The VAPS Teflon and Nylon filters were subsequently extracted in deionized water and analyzed for major ions (NH, SO, and NO) and Na^+ by ion chromatography (IC) (Stevens et al., 1978). The NH, SO, and Na^+ determinations were made from the Teflon filter extracts and the NO results are the sum of the Teflon and Nylon filter extract determinations. The VAPS quartz filters were manually split half was used for organic carbon (OC) and elemental carbon (EC) analysis by thermal optical analysis (TOA) (Birch and Cary, 1996).

Receptor Modeling Approaches

It is suggested by the following, that there is no single model that will be generally applicable to all situations. For the PTEAM data, a three-way analysis was employed. The model is written as

$$x_{ijk} = \sum_{h=1}^{p} a_{ih} b_{jh} c_{kh} + e_{ijk} \qquad (1)$$

In this equation, a_{ih} is the h*th* source contribution for each i*th* sampling interval, b_{jh} contains the jth concentration in the hth source profile, and c_{kh} indicates the k*th* sample type (community, indoor, or outdoor) such that the product of a_{ih} and c_{kh} provides the source contributions for each sample within the data set. This equation can be rewritten in matrix form as

$$X = A \cdot B \cdot C + E \qquad (2)$$

The three modes used in the analysis were (1) 18 elemental concentrations, (2) 303 samples, and (3) sample type (personal PM_{10}, indoor PM_{10}, outdoor PM_{10}, indoor $PM_{2.5}$, outdoor $PM_{2.5}$). This analysis provided the factor profiles, their contributions, and the distribution of each source between personal, indoor, outdoor fine and coarse (PM_{10} - $PM_{2.5}$) PM. Fit to the data was reasonably good and the individual factors could be readily interpreted.

For the Baltimore Exposure Panel Study (BPMEES), this three-way model was used to analyze the fixed site monitors at the community site, outside and inside the residence facility (Hopke et al., 2002). In this case, the indoor sources could be readily distinguished from the clearly outdoor sources. It was also possible to estimate the fraction of indoor particulate matter concentration that was largely due to the presence of outdoor particles in the indoor environment.

However, in the case of the personal and apartment particulate matter concentrations measured in the BPMEES (Hopke et al., 2003), it was found that a three-way model (Equation 1) did not provide a good fit to these data. After further exploration, a more complex model was developed that did provide an appropriate fit to the data and interpretable factors. In this new model, two different categories of factors are defined. In one category are external factors that contribute to all four types of environments (indoor, outdoor, personal, and apartment) and have the same composition profiles for all of the types of samples. The other category consists of internal factors that contribute mass to only the personal and apartment samples and thus, attempt to represent sources specific to this subset of samples. This model is then used to fit all of the different types of samples. A number of factors (*N*) are used for fitting all samples, while *P* factors are used only to fit the personal and apartment samples. The set of *P* factors forms an ordinary 2-way factor model. The *N* factors of the first set must fit the indoor and outdoor data,

and thus they form a basis set for them. This basis set is used for fitting the personal and apartment data, together with the additional 2-way model. The equations of the model are relatively simple:

$$x_{ijdt} = \sum_{p=1}^{N} a_{ipdt} b_{jp} + \sum_{p=N+1}^{N+P} a_{ipdt} b_{jp} \qquad (i = 1,\ldots,15, \quad t = \text{pers/apt data})$$
$$x_{ijdt} = \sum_{p=1}^{N} a_{ipdt} b_{jp} \qquad (i = 16, \quad t = \text{indoor/outdoor data}) \tag{3}$$

where a_{ipdt} is the contribution of the pth source to sample type t collected on day d and b_{jp} is the concentration of the jth chemical species in particulate matter from the pth source. Such a model can be fitted using the multilinear engine (ME) (Paatero, 1999).

The three modes used in the analysis were (1) 42 elemental concentrations plus unknown mass ($PM_{2.5}$ - sum of the oxides of the crustal elements - sulfur as ammonium sulfate), (2) 391 PEM samples, and (3) sample type (personal $PM_{2.5}$, apartment $PM_{2.5}$, indoor $PM_{2.5}$, outdoor $PM_{2.5}$). This analysis provided the factor profiles, their contributions, and the distribution of each source for both external and internal factors between personal, apartment samples. In addition, the external source contributions to outdoor and indoor were reported. In general, a reasonable understanding of the particle sources was obtained and specific unusual samples could be readily identified by evaluating each subjects external and internal source contribution estimates. From these results, it appears that there is no single model that can be routinely applied to the range of possible measurement plans. The model must be structured based on the measurement plan. If a standard protocol were developed that was employed in repeated studies, it may be possible to develop a model that would be applicable to all such studies.

Interpretation of Results

For the PTEAM data, reasonable fits could be obtained using the three-way model in equation (2) where the A matrix provides the series of source contributions for each individual, B contains the source profiles, and C indicates the sample type such that the product of A and C provides the source contributions for each sample within the data set. Using this approach, major particle sources of personal PM_{10} exposure in Southern California in Fall 1991 were resuspension of the soil particles, generation of the particles by personal activities, penetration of fine particles from the outdoors, such as emissions from oil combustion or nonferrous metal operations and motor vehicle exhaust. Sources of coarse particles such as sea-salt or ambient soil did not directly influence indoor air conditions and personal exposure to PM_{10}. Sources of fine particles such as secondary sulfate, oil combustion, nonferrous metal operations and motor vehicle exhaust had similar impacts on outdoor, indoor and personal PM_{10}. In addition, good agreement was obtained between information about daily personal activities and values of factor scores. Smoking, indoor cooking, vacuuming resulted in elevated "personal activities", "indoor soil", and "resuspended indoor soil" factor scores, and did not affect "secondary sulfate" and "nonferrous metal operations and motor vehicle exhaust."

From the results of the BPMEES data analysis (Hopke *et al.*, 2002), it appears that there are severe limits to how much interpretation can be applied to the results for individual personal or apartment samples. Because of the uncertainties in input data, the limited number of samples,

and the apparent unique behavior of specific individuals that cannot be reflected in a factor analysis, it is necessary to recognize the limitations on how much information can be gleaned on all of the sources leading to the exposure of individuals to airborne particulate matter. There are difficult decisions to be made by the data analyst as to whether or not to retain unusual values in the analysis since in some cases, such as person 5 in the BPMEES data base, it is clear that sources are producing exposure to the specific individual and apartment that are unrelated to any other participant in the study.

EPA United States Environmental Protection Agency

Daily follow-up Questionnaire

(technician administered)

Participant ID ☐☐☐☐ Start Date (yesterday's date) ☐☐/☐☐/☐☐

A A B C D M M D D Y Y

A = Participant ID
B = Cohort
C = Cycle

This form is a hard copy version of a computer driven questionnaire. The overall format and layout of the computer questionnaire may differ significantly, but this hard copy version provides an accurate representation of the questions and overall extent of the information we wish to capture in the follow-up interview with the participant. The tables in the computer version can capture many more events/occurrences of an activity than is indicated in this hard copy version. The use of '24 hours' throughout the questionnaire refers to the previous monitoring period in its entirety.

1. Did you or anyone smoke (cigarette, cigar or pipe) around you within the last 24 hours at any location? Y ☐ / N ☐

If yes, please indicate in the following table where and when smoking occurred. If you smoked or your exposure to smoke occurred repeatedly over time in one location, please only indicate a single start time and duration for the entire time you smoked or were exposed to smoke in each location.

Location[1]	Time (start)	Duration (minutes)	Comments

1 - (IH) Indoors at Home, (IO) Indoors at other, (O) Outdoors, (C) Car or other vehicle

2. Did you or anyone smoke (cigarette, cigar or pipe) inside your home within the last 24 hours? Y ☐ / N ☐

If yes, please indicate in the following table when smoking occurred inside your home. If smoking occurred repeatedly over time, please only indicate a single start time and duration for the entire time you or someone else smoked inside your home.

Time (start)	Duration (minutes)	Comments

3. Did you cook or were you around when someone else was cooking during the last 24 hours? Y ☐ / N ☐

If yes, please fill out the following table.

Your Location[1]	Cooker Type[2]	Type of cooking[3]	Time (start)	Duration (minutes)	Smoke Produced[4]	Exhaust Fan[5]	Comments

1 - (IH) Indoors at Home, (IO) Indoors at other, (O)utdoors, (C)ar or other vehicle
2 - (S) Stove, (M) Microwave, (O) Oven, (G) Grill
3 - (FG) Frying or grilling, (BB) Baking or broiling, (TO) Toasting, (BO) Boiling, (OT) Other, please specify
4 - Was anything burned while cooking that produced visible smoke? (Y, N, Don't Know)
5 - Was an exhaust fan used that was vented outdoors? (Y, N, Don't Know)

4. Were you around burning candles or incense at any location during the last 24 hours? Y ☐ / N ☐

If yes, please indicate in the following table your location and when you were around burning candles or incense.

Your Location[1]	Time (start)	Duration (minutes)	Type[2]	Metal Wick[3]	Comments

1 - (IH) Indoors at Home, (IO) Indoors at other, (O)utdoors, (C)ar or other vehicle
2 - (C) Candles or (I) Incense
3 - Applies to candles only. Did the candle have a metal wick?

5. Did you use a humidifier in your home in the last 24 hours? Y ☐ / N ☐

If yes, please fill out the following table.

Time (start)	Duration (minutes)	Humidifier Type[1]	Water Type[2]	Additives[3]	Comments

1 - (E) Evaporative, (U) Ultrasonic cool-mist, (H) Heated, (O) Other
2 - (T) Tap, (D) Distilled, (B) Bottled, (O) Other
3 - Specify additives including mentholatum, etc.

6. Was your primary heater (furnace, etc.) used in your home during the last 24 hours? Y ☐ N ☐

7. Were any other heating devices used in your home during the last 24 hours? Y ☐ N ☐

If yes, please indicate in the following table when and what type of device was used.

Time (start)	Duration (minutes)	Device Type[1]	Smoke or fuel smelled?	Door open[2]	Comments

1 - (WF) Wood burning fireplace, (GF) Gas logs fireplace, (WS) Wood burning stove, (KE) Kerosene space heater, (O) Other, please specify.
2 - Applies to a wood stove only. Other than to add wood, was the door left open on the wood stove while it was in operation?

8. Was an air conditioner run during the last 24 hours in your home? Y ☐ N☐

9. Were any windows open in your home in the last 24 hours? Y ☐ N☐

If yes, please indicate in the following table when, the number of windows and how many were open wider than 6 inches.

Time (start)	Duration (minutes)	# Windows Open	# Open > 6"	Comments

10. Were any exterior doors left open for more than five minutes or were screen doors used for ventilation in your home during the the last 24 hours? Y ☐ N ☐

If yes, please indicate in the following table when exterior doors were open.

Time (start)	Duration (minutes)	Comments

11. Was an air cleaner or air filter used in your home in the last 24 hours? Y ☐ / N ☐

If yes, please indicate in the following table when and the type of air filter/cleaner.

Time (start)	Duration (minutes)	Type[1]	Comments

1 - (H) HEPA filter, (Z) Ozonator, (E) Electrostatic precipitator, (O) Other, please specify

12. Were housecleaning chores performed by you or someone else in your home during the last 24 hours? Y ☐ / N ☐

If yes, please indicate in the following table when and the type of cleaning that took place.

Time (start)	Duration (minutes)	Type of cleaning[1]	Comments

1 - (V) Vacuuming, (S) Sweeping, (D) Dusting, (O) Other, please specify.

13. Were cleaning products used in your home within the last 24 hours? Y ☐ / N ☐

If yes, please indicate in the following table when and the cleaning product(s) used. Please do not include bleach, ammonia based cleaners (e.g. Windex), vinegar, baking soda, dishwashing detergent, laundry detergent

Time (start)	Duration (minutes)	Cleaning Product	Comments

14. Were any of the following aerosol spray products used in the home within the last 24 hours? Air freshener, spray perfume or cologne, hair spray, spray deodorant.

Time (start)	Duration (minutes)	Type[1]	Comments

1 - (AF) Air freshner, (PC) Perfume or cologne, (HS) Hair spray, (SD) Spray deodorant, (OT) Other, please specify.

15. Were any petroleum based solvents, paints or glues used in or around your home during the last 24 hours? Petroleum based solvents include paint thinner, paint stripper, etc. Paints may include oil based and latex or acrylic paint. Y ☐ / N ☐

If yes, please indicate type of solvent or paint used during the last 24 hours _____
_____

16. Were any dry-cleaned items (clothes, etc.) brought into your home during the last 24 hours? Y ☐ / N ☐

17. Did you smell smoke or any other unusual chemical smells in or around your home within the last 24 hours that you have not already identified? Y ☐ / N ☐

If yes, please indicate in the following table when and the type and source of odor, if known.

Location[1]	Time (start)	Duration (minutes)	Type and source of odor	Comments

1 - (I) Indoors, (O) Outdoors

18. Were household or lawn chemicals used around your home in the last 24 hours? Y ☐ N ☐

If yes, please indicate in the following table where, when and the type of chemical(s) used.

Location[1]	Time (start)	Duration (minutes)	Type[2]	Comments

1 - (I) Indoor, (O) Outdoors
2 - (H) Herbicides, (P) Pesticides, (O) Other, please specify

19. Were lawn mowers and/or other small engines used around your home in the last 24 hours?
Y ☐ N ☐

If yes, please indicate in the following table when and the type used.

Time (start)	Duration (minutes)	Type[1]	Comments

1 - (L) Lawn mower, (W) Weed eater, (B) Blower, (O) Other, please specify.

20. Did you drive or were you a passenger in a motor vehicle of any type within the last 24 hours? Y ☐ N ☐

If yes, please indicate in the following table when and what type of vehicle.

Time (start)	Duration (minutes)	Vehicle Type[1]	Comments

1 - (C) Car, (T) Truck, (B) Bus, (M) Motorcycle, (O) Other, please specify

21. Did you put gas in a vehicle or were you in a vehicle while it was being refueled in the last 24 hours? Y ☐ / N ☐

22. If a garage is connected to the home, did anyone leave a vehicle or a small engine appliance (e.g. lawnmower, weed-wacker, etc.) running in the garage longer than 30 seconds during the last 24 hours? Y ☐ / N ☐

23. If a garage is connected to the home, did anyone drive a vehicle into the garage after it was running for more than 5 minutes? Y ☐ / N ☐

If yes to either **22.** or **23.**, please indicate in the following table when either of these events occurred.

Time (start)	Duration (minutes)	Description[1]	Garage door[2]	Comments

1 - Indicate whether (A) vehicle/other engine was running in garage longer than 30 seconds or (B) a vehicle was driven into the garage after the engine was running for more than 5 minutes

2 - Indicate whether the garage door was (O) Open or (C) Closed during this activity. Was the garage door closed just after a vehicle was driven into the garage?

Neighborhood Source Survey Instructions

Purpose: To identify sources that may impact a participant's residence influencing indoor concentrations of and exposure to PM/Air Toxics.

Identify any potential sources of PM and Air Toxics (ATs) within a 1 km radius in all directions from the participant's residence. Potential sources are those that may alter concentrations of PM/ATs at the residential or local level. Examples of source categories include (type of pollutant indicated in parentheses):

Roads – the distance from the residence to the nearest roadway should be noted in the comments column along with the GPS coordinates. Any other roadways within the 1 km radius that could impact the residence should be noted as well. (PM, PAH, VOCs)
Construction – building (PM, diesel) and road (PM, diesel, asphalt/PAHs)
Diesel trucks/buses – warehouse/distribution hub, bus terminal, bus line/bus stop nearby, railroad (PM, diesel)
Gas stations – indicate if diesel is served there? (BTEX, diesel vehicles)
Manufacturing/industry – type/description of facility (i.e. what is produced there – need idea of what may be emitted in terms of PM/ATs)
Restaurant – meat cooking, wood burning, (PM, PAH)
Dry cleaner – solvents (VOCs)
Lumber yard – plywood, particle board, pressed wood (formaldehyde)
Automotive repair/body shop – sanding, solvents (PM, VOCs)
Janitorial services – cleaners/solvents (VOCs)
Photofinishing – solvents (chlorinated VOCs)

Neighborhood Source Survey

Subject ID

Source Information

GPS Coordinates of Source

			.					N
			.					W

Source Description - Category

Comments

Source Information

GPS Coordinates of Source

			.					N
			.					W

Source Description - Category

Comments

Source Information

GPS Coordinates of Source

			.					N
			.					W

Source Description - Category

Comments

Date

		/			/				

Draft

Participant Survey

Subject ID

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1. What is this person's sex?

- ⬭ Male
- ⬭ Female

2. What is this person's height in inches?

3. What is this person's weight in pounds?

4. What is this person's date of birth?

		/			/				

Note: Please answer BOTH Questions 5 and 6.

5. Is this person Spanish/Hispanic/Latino?

- ⬭ No, not Spanish/Hispanic/Latino
- ⬭ Yes, Mexican, Mexican Am., Chicano
- ⬭ Yes, Puerto Rican
- ⬭ Yes, Cuban
- ⬭ Yes, other Spanish/Hispanic/Latino:

6. What is this person's race?

- ⬭ White
- ⬭ Black, African Am., or Negro
- ⬭ American Indian or Alaska native
- ⬭ Asian Indian
- ⬭ Chinese
- ⬭ Filipino
- ⬭ Japanese
- ⬭ Korean
- ⬭ Vietnamese
- ⬭ Native Hawaiian
- ⬭ Guamanian or Chamorro
- ⬭ Samoan
- ⬭ Some other race:

7. LAST WEEK, did this person do ANY work for either pay or profit? *Mark the "Y" oval even if the person worked only 1 hour, or helped without pay in a family business or farm for 15 hours or more, or was on active duty in the Armed Forces.*

- ⬭ (Y)
- ⬭ (N) → Skip to **12**

8. At what location did this person work LAST WEEK? *If this person worked at more than one location, print where he or she worked most last week..*

a. Address (Number and street name)

b. Name of city, town, or post office

c. Name of U.S. state or foreign country

d. ZIP Code

					-				

9. How did this person usually get to work LAST WEEK? *If this person usually used more than one method of transportation during the trip, mark the oval of the one used for most of the distance.*

- ⬭ Car, truck, or van
- ⬭ Bus or trolley bus
- ⬭ Streetcar or trolleycar
- ⬭ Subway or elevated
- ⬭ Railroad
- ⬭ Ferryboat
- ⬭ Taxicab
- ⬭ Motorcycle
- ⬭ Bicycle
- ⬭ Walked
- ⬭ Worked at home
- ⬭ Other method

10. What time did this person usually leave home to go work last week?

		:				AM/PM

Date

		/			/			

Draft

Participant Survey

Subject ID

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11. Approximately how long doe it usually take this person to commute one-way to work?

hr min

12. Approximately how many hours each day are you away from home? | | |

13. Approximately how many hours each day do you spend outside? | |

14. Do you have any hobbies that involve the use of solvents, paint, glue or textiles? (Y) (N)

Survey Approved
OMB Control No. 2080-0058
Approval Expires 7/31/2002

1

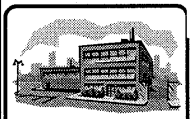
Indoors at Home

Outside at or Near Home

In Transit

At Work away from Home

Outside away from Home



Indoors away from Home

Activity Description								*Smoker Nearby* (minutes)	*Cooking* (minutes) Self	Other
AM 6:00										
:15										
:30										
:45										
7:00										
:15										
:30										
:45										
8:00										
:15										
:30										
:45										
9:00										
:15										
:30										
:45										
10:00										
:15										
:30										
:45										
11:00										
:15										
:30										
:45										

Tech. Init. ☐☐

Subject ID ☐☐☐☐☐

Start Date ☐☐ / ☐☐ / ☐☐
M M / D D / Y Y

50268

2

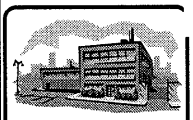

Indoors at Home


Outside at or Near Home


In Transit


At Work away from Home


Outside away from Home


Indoors away from Home

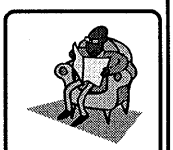
Activity Description							*Smoker Nearby* (minutes)	*Cooking* (minutes) Self	Other
PM 12:00									
:15									
:30									
:45									
1:00									
:15									
:30									
:45									
2:00									
:15									
:30									
:45									
3:00									
:15									
:30									
:45									
4:00									
:15									
:30									
:45									
5:00									
:15									
:30									
:45									

Subject ID | **Start Date**

50307

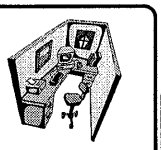
** Same as previous page **

3

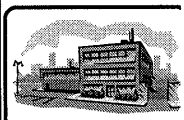

Indoors at Home


Outside at or Near Home


In Transit


At Work away from Home


Outside away from Home


Indoors away from Home

Activity Description								*Smoker Nearby* (minutes)	*Cooking* (minutes) Self	Other
PM 6:00										
:15										
:30										
:45										
7:00										
:15										
:30										
:45										
8:00										
:15										
:30										
:45										
9:00										
:15										
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:45										
10:00										
:15										
:30										
:45										
11:00										
:15										
:30										
:45										

Subject ID □□□□□ **Start Date** □□ / □□ / □□

50327

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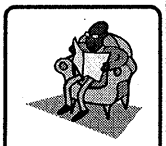
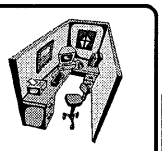
4

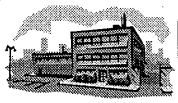

Indoors at Home


Outside at or Near Home


In Transit


At Work away from Home


Outside away from Home


Indoors away from Home

Activity Description							*Smoker Nearby* (minutes)	*Cooking* (minutes) Self	Other
AM 12:00									
1:00									
2:00									
3:00									
4:00									
5:00									
** Resume 15 min. intervals **									
6:00									
:15									
:30									
:45									
7:00									
:15									
:30									
:45									
8:00									
:15									
:30									
:45									
9:00									
:15									
:30									
:45									

Subject ID **Start Date**

_____ __ / __ / __

50346

** Same as previous page **

Residence Survey

Subject ID

Home and Vehicle Characteristics

GPS Coordinates of Residence

			.								

N **W**

1. Which best describes this building? *Include all apartments, flats, etc., even if vacant.*

- ⬭ A mobile home
- ⬭ A one-family house detached from any other house
- ⬭ A one-family house attached to one or more houses
- ⬭ A building with 2 or more apartments
- ⬭ Boat, RV, van, etc.
- ⬭ Other, please specify:

2. Approximate age of building (years): | | | |

3. How many people usually reside in this home? | |

4. How many children (<18 years old) usually reside in this home? | |

4. a. What are the ages of the children in this home?

3. What type of garage, if any, is there associated with the dwelling?

⬭ None, detached, or separate carport ⬭ Attached ⬭ Underneath

3. a. Is this garage used for:

⬭ Parking one car ⬭ Parking two cars ⬭ Parking more than two cars ⬭ Storage only

3. b. Indicate any small gasoline engine appliances stored in the garage:

- ⬭ Lawnmower
- ⬭ Weedwacker
- ⬭ Leaf blower
- ⬭ Chain saw
- ⬭ Other, please specify:
- ⬭ None

3.c. Are gasoline or other petroleum based solvents stored in the garage? ⓨ ⓝ

3.d. Is there a door leading from the garage into the dwelling? ⓨ ⓝ

4. Indicate below information about the vehicle(s) used for transportation:

Primary vehicle used for transportation:

Make	Model	Year

Secondary vehicle used for transportation:

Make	Model	Year

Date

		/			/				

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Residence Survey

Subject ID

Heating, Cooling and Ventilation Characteristics

1. How many separate central AC or window/wall units are in the home?

☐ Central AC units ☐ Window/wall AC units

2. What are the heating sources in the home?

- ⬭ Forced air gas
- ⬭ Forced air oil
- ⬭ Forced air electric
- ⬭ Forced water, radiator
- ⬭ Heat pump
- ⬭ Wood burning stove
- ⬭ Fireplace, gas
- ⬭ Fireplace, wood
- ⬭ Gas space heater
- ⬭ Kerosene space heater
- ⬭ Electric space heater
- ⬭ Open stove/oven
- ⬭ Other, please specify:

2.a. Indicate which heating source is NOT vented to the outside:

- ⬭ None
- ⬭ Forced air gas
- ⬭ Forced air oil
- ⬭ Forced air electric
- ⬭ Forced water, radiator
- ⬭ Heat pump
- ⬭ Wood burning stove
- ⬭ Fireplace, gas
- ⬭ Fireplace, wood
- ⬭ Gas space heater
- ⬭ Kerosene space heater
- ⬭ Electric space heater
- ⬭ Open stove/oven
- ⬭ Other, please specify:

2.b. Indicate which heating source has an external fresh-air source:

- ⬭ None
- ⬭ Forced air gas
- ⬭ Forced air oil
- ⬭ Forced air electric
- ⬭ Forced water, radiator
- ⬭ Heat pump
- ⬭ Wood burning stove
- ⬭ Fireplace, gas
- ⬭ Fireplace, wood
- ⬭ Gas space heater
- ⬭ Kerosene space heater
- ⬭ Electric space heater
- ⬭ Open stove/oven
- ⬭ Other, please specify:

3. Is there a whole-house or attic fan? ⬭ Y ⬭ N

4. Are there storm windows? ⬭ Y ⬭ N

Residence Survey

Subject ID

Cooking, Cleaning and Home Characteristics

1. What type of cooking fuel is used?

○ Gas ○ Electric ○ Other, please specify: _____

2. Is there an exhaust fan for the stove, range, oven, or elsewhere in the kitchen area? (Y) (N)

2. a. How does this fan work?

○ Kitchen exhaust vented outside ○ Other, please specify: _____

○ Recirculation of indoor air ○ Don't know

○ Charcoal filter

3. Is there a clothes dryer? (Y) (N)

3. a. Is the clothes dryer vented out of the dwelling? (Y) (N)

4. Is there a continuously burning pilot light on a:

○ Gas range ○ Oven ○ Clothes dryer ○ Water heater ○ Furnace

5. Have freshly dry cleaned clothes been brought into the house during the last week? (Y) (N)

5. a. If yes, how many days ago? ☐

6. Does anyone living here smoke inside your home? (Y) (N)

If yes:

6. a. How many persons living here smoke inside your home? ☐

6. b. How many persons living here smoke cigarettes inside your home? ☐

6. c. How many persons living here smoke cigars or pipes inside your home? ☐

7. Does anyone use a humidifier in your home? (Y) (N)

7. a. If yes, what type of humidifier?

○ Ultrasonic ○ Other, please specify:

○ Evaporative ○ Don't know

Brand Name and Model _____

Residence Survey

Subject ID

Recent Construction and Painting

1. Have you painted in or around your home during the last 7 days or will you have any painting done during the monitoring period? (Y) (N)

2. Do you have any new furniture that has been in your home less than 1 month? (Y) (N)

3. Have you had any new construction to your home during the last 6 months that involved plywood or particle board, including cabinets, or any other pressed wood products? (Y) (N)

4. Have you had any new carpet installed in your home during the last 6 months? (Y) (N)

5. Have you had any new linoleum installed in your home during the last 6 months? (Y) (N)

Residence Survey

Subject ID

Room Characteristics

Draw a floor plan of the home (each level, if applicable) in the space provided below. Please label the rooms using the following format - KI (Kitchen), LR (Living Room), DR (Dining Room), FR (Family Room), BR# (Bed Room 1,2, etc.), BA# (Bathroom 1,2,etc.). Use the same names on the following page.

Residence Survey

Subject ID

Use the same name for each room as used on the previous page.
Measurements of length and width are in feet.

	Room	Length	Width	Floor Coverage %	Presence of molds, mildew, water damage	Dust factor for room Clean Dusty
1					Y N	○ ○ ○ ○ ○ ○
2					Y N	○ ○ ○ ○ ○ ○
3					Y N	○ ○ ○ ○ ○ ○
4					Y N	○ ○ ○ ○ ○ ○
5					Y N	○ ○ ○ ○ ○ ○
6					Y N	○ ○ ○ ○ ○ ○
7					Y N	○ ○ ○ ○ ○ ○
8					Y N	○ ○ ○ ○ ○ ○
9					Y N	○ ○ ○ ○ ○ ○
10					Y N	○ ○ ○ ○ ○ ○
11					Y N	○ ○ ○ ○ ○ ○
12					Y N	○ ○ ○ ○ ○ ○
13					Y N	○ ○ ○ ○ ○ ○
14					Y N	○ ○ ○ ○ ○ ○
15					Y N	○ ○ ○ ○ ○ ○