



Sonoma Technology, Inc.

1360 Redwood Way, Suite C
Petaluma, CA 94954-1169
707/665-9900
FAX 707/665-9800
www.sonomatech.com

**A TECHNICAL APPROACH USING AMBIENT DATA
TO TRACK AND EVALUATE AIR QUALITY
PROGRAMS**

**Draft Final Report
STI-905213.02-2918-DFR**

**By:
Hilary R. Hafner
Paul T. Roberts**

**Sonoma Technology, Inc.
1360 Redwood Way, Suite C
Petaluma, CA 94954-1169**

**Prepared for:
Dennis Doll
Work Assignment Manager
Emissions, Monitoring and Analysis Division
Ambient Air Monitoring Group
Office of Air Quality Planning and Standards
U.S. Environmental Protection Agency
Research Triangle Park, NC 27711**

March 13, 2006

TABLE OF CONTENTS

<u>Section</u>	<u>Page</u>
LIST OF FIGURES	v
LIST OF TABLES	vii
1. INTRODUCTION.....	1-1
1.1 Background and Objective	1-1
1.2 Overview of the Technical Approach.....	1-1
2. BUILDING A CONCEPTUAL MODEL	2-1
2.1 Decision Trees	2-1
2.2 Identify Spatial Scale and Timing of Emissions.....	2-3
2.3 Identify Spatial Scale and Timing of Control Implementation	2-6
2.4 Identify Pollutant-Specific Temporal and Spatial Scales	2-8
2.5 Identify Availability of Ambient Data.....	2-9
3. PERFORM STATISTICAL AND GRAPHICAL ANALYSES	3-1
3.1 Basic Data Analyses	3-1
3.2 Assessing Trends	3-3
3.2.1 Metrics for Consideration: Ozone	3-3
3.2.2 Metrics for Consideration: PM _{2.5} Mass	3-4
3.2.3 Trends Analysis.....	3-5
3.3 Adjusting for Meteorological Impacts.....	3-6
3.3.1 Mathematical Techniques for the Meteorological Adjustment of Air Quality Trends.....	3-7
3.4 Corroborative Analyses	3-8
3.4.1 Transport and Trajectories.....	3-8
3.4.2 Source Apportionment	3-11
3.4.3 Estimating Relative Emission Contributions	3-13
3.4.4 Other Corroborative Evidence.....	3-16
4. SELECTING EXAMPLES FOR CASE STUDIES.....	4-1
4.1 Overview.....	4-1
4.2 NO _x and SO ₂ Reductions	4-1
4.3 Multiple Pollutant Reductions	4-2
4.4 Diesel Emissions Reductions.....	4-4
4.5 Air Toxics Reductions	4-5
4.6 Summary.....	4-7
5. REFERENCES	5-1

LIST OF FIGURES

<u>Figure</u>	<u>Page</u>
2-1 Decision tree for assessing trends starting with known emission changes or control strategy implementation.....	2-2
2-2 Decision tree for assessing trends starting with an observed trend in an ambient pollutant concentration over time	2-3
3-1 Annual average PM _{2.5} mass concentrations at sites in the Detroit area.....	3-2
3-2 Annual average reconstructed PM _{2.5} mass components at sites in the Detroit area	3-2
3-3 Preliminary map of annual average benzene concentration trends from 1990-2005 by EPA region.....	3-6
3-4 Spatial probability density plots for all samples and for samples with high contributions from a mining factor identified in source apportionment of PM _{2.5} data collected in Phoenix, Arizona, during 2001-2003.....	3-10
3-5 Average VOC mass apportioned to mobile and non-mobile sources at Azusa in Los Angeles, California, in 1987 using CMB and in 2001-2003 using PMF.....	3-12
3-6 Spatial probability density plots for Fresno, California, for the summers of 2002 and 2004.....	3-14
3-7 Transport pattern differences between the summers of 2002 and 2004 for Fresno, California	3-15
3-8 Geographic distributions of SO ₂ and (b) NO _x EIP for the 20%-worst visibility days and 20%-best visibility days observed at Hercules-Glades, Missouri.....	3-16
3-9 Corroborative information including vehicle miles traveled, gasoline composition, and controls to investigate changes in PM _{2.5} components in Washington, DC	3-17
4-1 Clinton Drive site near the Houston Ship Channel.....	4-4
4-2 Annual average perc concentrations at three sites in Los Angeles, California, and the NEI estimates of perc emissions.	4-6

LIST OF TABLES

<u>Table</u>		<u>Page</u>
2-1	Example summary of conceptual model information for SO ₂	2-4
2-2	Example summary of conceptual model information for NO _x	2-5
2-3	Summary of control information for RFG.	2-7
2-4	Summary of information to consider for ozone.....	2-8
2-5	Summary of information to consider for PM _{2.5} mass.	2-9
2-6	Important issues about data availability.....	2-10

1. INTRODUCTION

1.1 BACKGROUND AND OBJECTIVE

The U.S. Environmental Protection Agency (EPA) has promulgated and begun implementing several regional-scale programs to reduce concentrations of ozone and particulate matter less than 2.5 microns (PM_{2.5}). The oxides of nitrogen (NO_x) multi-state implementation plan (SIP) call has resulted in significant reductions in NO_x emissions from large utilities and boilers over the past several years in eastern parts of the United States. The EPA documented improvements in summertime ozone air quality in rural and urban areas, especially between 2002 and 2005, and linked these improvements to the emissions reductions (U.S. Environmental Protection Agency, 2005). While regional-scale improvements in ozone have been noted, the EPA wants to develop refined methodologies to correlate emissions controls to changes in ambient air quality that can be applied not only to other regional-scale pollutants, such as PM_{2.5}, but also to urban (local) scales where local controls should noticeably impact air quality.

The goal of this document is to provide a technical approach to assess the link between emissions controls and air quality changes with a focus on local and urban scales. The focus of this work is primarily ozone and PM_{2.5}; however, techniques developed may be applicable to other pollutants such as air toxics.

1.2 OVERVIEW OF THE TECHNICAL APPROACH

This technical approach was developed to guide analysts in their evaluation of the impacts of regional control programs (e.g., those that affect multiple states) and local control programs (e.g., those that affect an urban area) on air quality. This evaluation is complicated and will be stepwise and site- and pollutant-specific. A major challenge is the scale of influence of a control and of the impact of that control on air quality. Previous investigations of ambient air quality changes encountered the confounding influences of multiple controls applied within similar time frames and at different spatial scales. One goal of this technical approach is to provide a more detailed process that will improve confidence in identifying and quantifying the cause and effect of changes in ambient pollutant concentrations (i.e., accountability).

While the focus of the technical approach is retrospective, the thought process, and some of the results, could be used to assess future controls and their potential impact on air pollutants. For example, if a future control scenario resembles a past control application, the technical approach in this document could be used to estimate future changes in ambient air quality based on the changes observed in the past.

The technical approach in this document provides a structure or road map for analysts. A key consideration is that consensus among several analyses gives the analyst more confidence in the cause and effect of a selected control on ambient data changes. The technical approach comprises the development of a conceptual model for the analysis (Section 2), discussion of statistical and graphical analysis tools or techniques and their applicability to different pollutants (Section 3), and a straw plan for an example application (Section 4). The example application will be addressed in the next task of this work assignment.

2. BUILDING A CONCEPTUAL MODEL

The first step in the analysis is to develop a conceptual model to help the analyst define the parameters of interest in the analysis, set up expectations, and identify the potential limitations and confounding issues for the analysis. The conceptual model aids the analyst in developing hypotheses to test, which include null hypotheses (e.g., comparing air quality trends in areas with and without the selected control). This section discusses the issues in assessing pollutant trends over time with respect to emissions controls and changes.

2.1 DECISION TREES

A decision matrix or tree provides an overview of required steps for evaluating various control scenarios. A decision matrix guides an analyst through the thought process of an analysis. Two approaches to a trends analysis can be taken depending on the availability of information: an emission control approach and an ambient data approach. Starting with the control measure, **Figure 2-1** illustrates a possible approach to an analysis:

- Select a control measure.
- Identify the air pollutants expected to be affected and the available data, other controls that might have affected the pollutants, and other pollutants that may have been affected.
- Consider the spatial scale, or zone of influence (ZOI), of the control measure. Was the control applied at a single facility (monitor-specific or fence line), at an urban scale (Metropolitan Statistical Area [MSA]-wide), regional scale (e.g., multi-state NO_x SIP call), national scale (e.g., 49-state automobile emission rules), or global scale (e.g., Montreal protocol)?
- Determine the timing and magnitude of the changes. Was the control phased in over a period of time, applied to specific emitters?
- Consider the magnitude of the expected air quality changes relative to the variability in the ambient data. If the inherent variability in the ambient data is very large, a small change in emissions may not be observable.
- Select the appropriate statistical metrics or approach for the analysis. Data treatments may help reduce the variability in the data so that trends can be observed.
- Develop hypotheses of expected changes, identify supporting evidence of changes, and investigate corroborative evidence of the changes. It is often helpful to test for changes in data sets or pollutants in which changes were not expected (i.e., check the null hypothesis).

An example of this approach is the selection of reformulated gasoline (RFG) introduction. The EPA Phase I RFG was implemented in 1995 with targeted benzene reductions of up to 43% in gasoline. Models predicted significant benzene reductions in gasoline vehicle exhaust caused by the use of RFG. Model predictions also indicated reductions in total aromatic hydrocarbons and 1,3-butadiene and increases in formaldehyde. Investigations of ambient benzene concentrations, weight fractions, and ratios showed that statistically significant

reductions were observed at Northeastern and Mid-Atlantic cities and that these reductions could be tied to gasoline composition changes (Main, 1998). Benzene reductions were not observed in ambient data in cities that did not introduce RFG. Similarly, hydrocarbons not expected to be affected by RFG, such as isoprene, did not change.

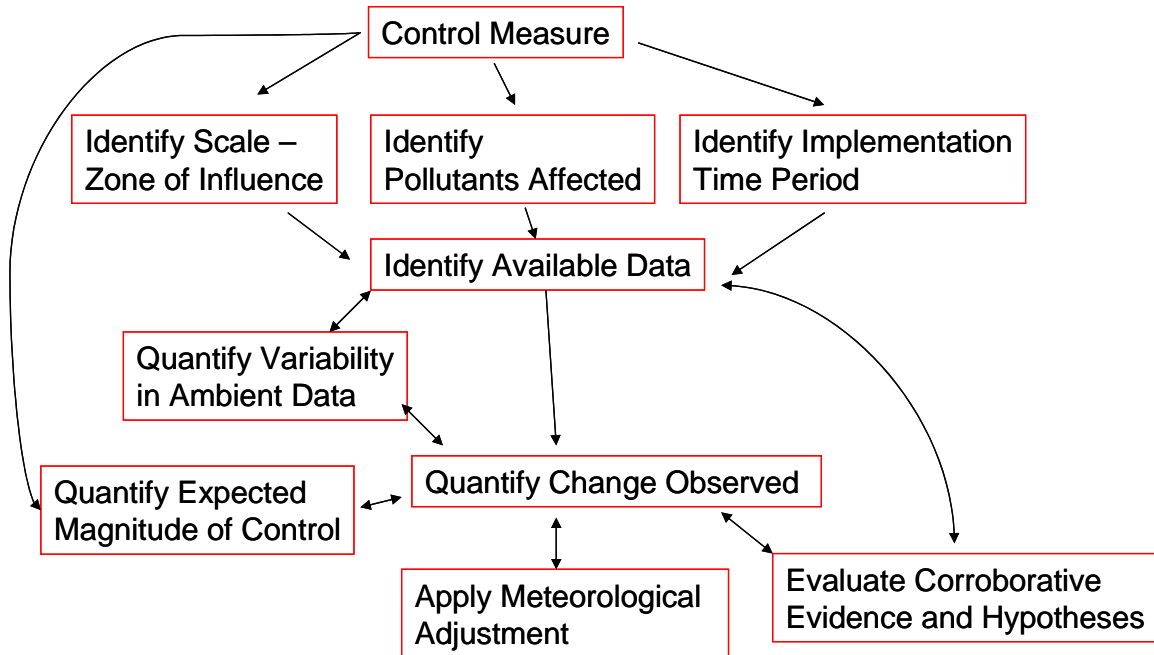


Figure 2-1. Decision tree for assessing trends starting with known emission changes or control strategy implementation.

Starting with an observed trend in ambient pollutant concentrations, **Figure 2-2** illustrates a possible approach to the analysis:

- Quantify the change observed in the ambient data. This approach could also be applied to a pollutant in which a change was not observed but expected.
- Apply meteorological adjustments to the pollutant trend. The goal is to reduce the effect of meteorology on the ambient concentrations so that the underlying trend in emissions can be more readily observed.
- Identify and assess other data sets and sites that may have also been affected by a similar control measure or emission change to understand the spatial scale of the ambient change. If the control was applied across a broad area, changes at additional sites might be expected.
- Identify potential emissions changes or control measures that could have contributed to the ambient trends. Local knowledge is often a key component of this part of the analysis.
- Compare the control measure implementation schedule with the ambient trends. Do the timing of the control implementation and the change in ambient concentrations coincide?

- Investigate corroborative evidence of the change and test for changes in pollutants in which a change was not expected. It is important not to over-interpret changes in ambient data.

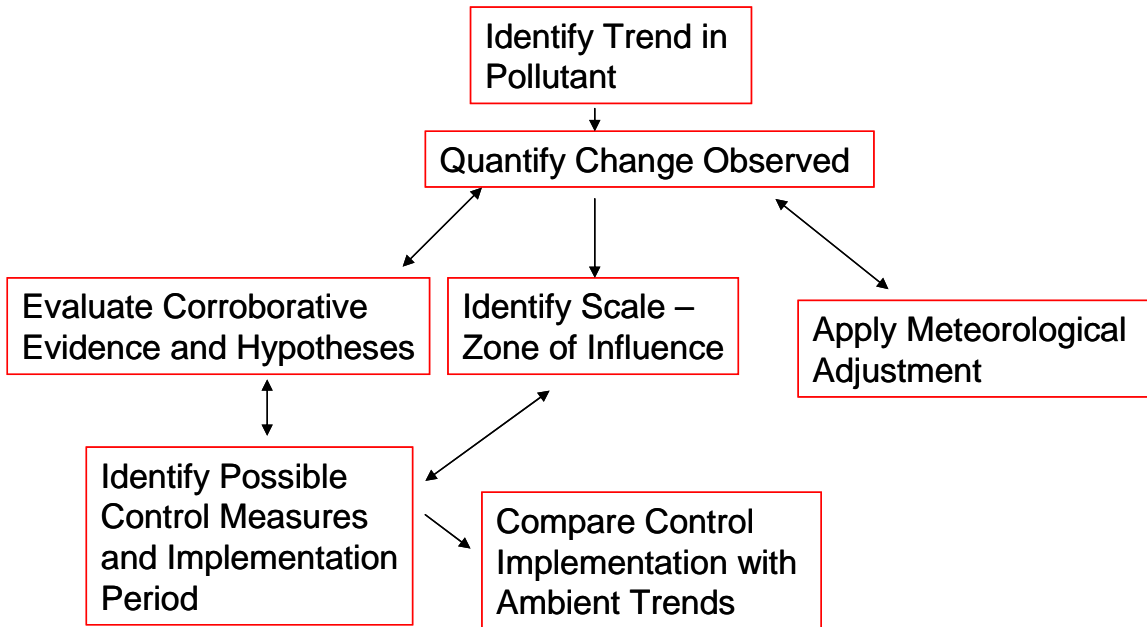


Figure 2-2. Decision tree for assessing trends starting with an observed trend in an ambient pollutant concentration over time.

An example of starting with the data was illustrated in recent air toxics work in which a site with significant declines in ambient lead concentrations were observed in Philadelphia, Pennsylvania. Local contacts confirmed that an industrial facility near the monitoring site had implemented PM₁₀ controls that also resulted in reduced lead emissions (Hafner et al., 2004).

The following sections provide a guide to develop the conceptual model. The conceptual model helps the analyst form expectations, develop hypotheses to build the case for accountability, and identify an analytical procedure. Example tables are used to illustrate emissions, controls, ambient data, and monitoring site considerations.

2.2 IDENTIFY SPATIAL SCALE AND TIMING OF EMISSIONS

Issues to consider about emissions include spatial distribution of emissions and typical surrogates for emissions density (e.g., population density), data sources (is it important to understand the projections used over time periods?), spatial maps (determine resolution needed), source areas (e.g., Ohio River Valley), expected changes from emission to receptor and time scale of these changes (e.g., conversion of sulfur dioxide [SO₂] to sulfate), and biogenic versus anthropogenic sources. Primary emissions for consideration include PM_{2.5} mass, carbon monoxide (CO), organic carbon (OC), elemental carbon (EC)/ black carbon (BC), nitrogen oxide (NO), volatile organic compounds (VOCs), SO₂, ammonia (NH₃), metals, and some toxics. Examples are provided for SO₂ (**Table 2-1**) and NO_x (**Table 2-2**).

Table 2-1. Example summary of conceptual model information for SO₂.

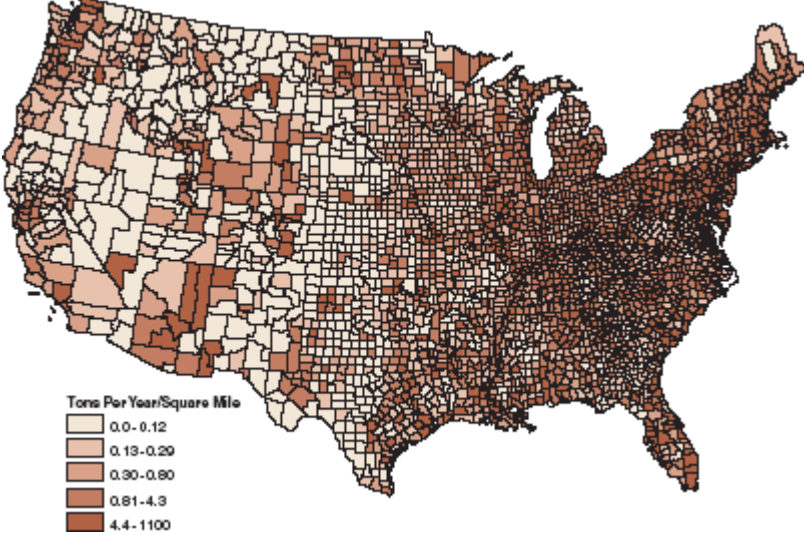
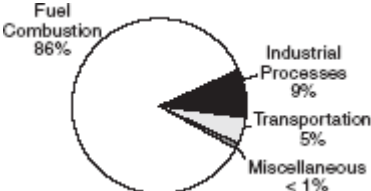
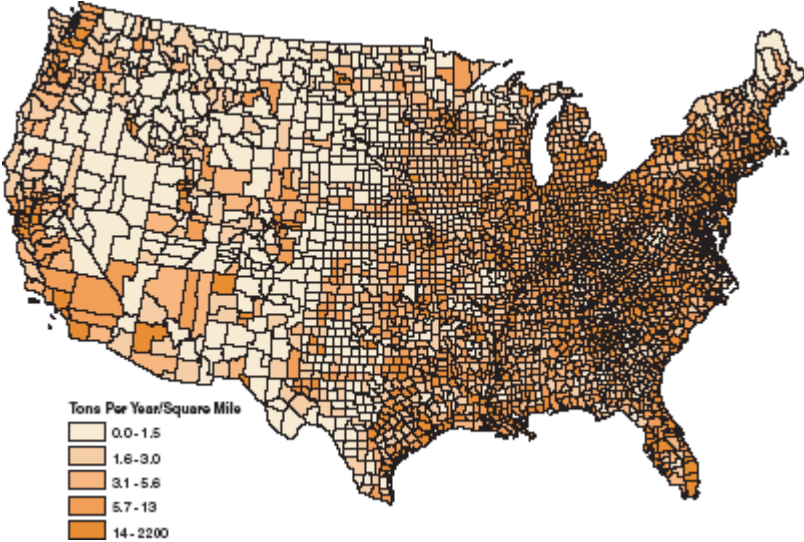
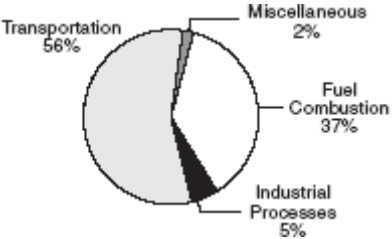
<p>Geographic distribution</p>	<p>2001 SO₂ emissions by county. Higher SO₂ emission rates occur to the east of the Mississippi River and in urban locations.</p> 
<p>Atmospheric residence time</p>	<p>One to one-and-one-half days. It takes about a day to form sulfate PM.</p>
<p>Major emission source categories</p>	<p>Fuel combustion, principally point sources</p>
<p>Emission inventory for the locations of concern</p>	<p>Compare a given location to the United States as a whole (2001):</p> 
<p>Seasonal variability</p>	<p>SO₂ -to-sulfate transformation rates peak in the summer because of enhanced summertime photochemical oxidation and SO₂ oxidation in clouds. Sulfate concentrations vary between seasons by at least a factor of two.</p>
<p>Biogenic sources?</p>	<p>No.</p>

Table 2-2. Example summary of conceptual model information for NO_x.

<p>Geographic distribution</p>	<p>2001 NO_x emissions by county. Emissions are typically higher in urban areas.</p> 
<p>Atmospheric residence time</p>	<p>NO_x can contribute to PM nitrate formation at night and during the day; daytime photochemistry also forms ozone.</p>
<p>Major emission source categories</p>	<p>Mobile sources, power generation</p>
<p>Emission inventory for the locations of concern</p>	<p>Compare a given location to the United States as a whole (2001):</p> 
<p>Seasonal variability</p>	<p>NO_x -to-nitrate transformation rates are favored in winter. It is estimated that about one-third of anthropogenic NO_x emissions in the United States are removed by wet deposition. Nitrate concentrations vary between seasons by at least a factor of two.</p>
<p>Biogenic sources?</p>	<p>Yes, natural sources include lightning, biological and abiological processes in soil, and stratospheric intrusion. Ammonia and other nitrogen compounds produced naturally are important in the cycling of nitrogen through the ecosystem.</p>

These tables help illustrate the spatial scales encountered for emissions. SO₂ emissions are dominated on a national scale by power generation, while NO_x emissions at a national scale are a combination of transportation and power generation. Thus, for NO_x, it is difficult to demonstrate the effect of regional trends in power plant NO_x controls on regional ozone changes because ozone downwind of an urban area has been influenced more by urban NO_x than by NO_x from a regional power plant.

2.3 IDENTIFY SPATIAL SCALE AND TIMING OF CONTROL IMPLEMENTATION

For changes in emissions and emission control implementation, the analyst needs to know as much as possible about the spatial scale and timing of control implementation in order to determine effective analyses of the ambient data. Information useful to the analyst includes where and when changes were implemented, model predictions of affected pollutants and magnitude of change in ambient air, the relative magnitude of change in emissions over space and time, what ambient pollutants or precursors were targeted, what other species could be affected, the availability of corroborative evidence (e.g., gasoline information), potential conflicting issues, the need for, and availability of, local knowledge, method of control enforcement (i.e., voluntary versus fine system), and the uniformity in space and time of the control implementation.

An example summary of control implementation information is provided in **Table 2-3** for the introduction of RFG at a national level. The table summarizes the types of information useful in developing a conceptual model of emissions controls:

- *Spatial scale.* The graphic provided in Table 2-3 illustrates the inherent spatial complexity in some of the control programs. A wide range of gasoline regulations, which would have a range of emissions changes, is shown.
- *Implementation period.* Some areas have separate winter and summer gasoline requirements. Changes in gasoline formulation often occurred relatively abruptly (short phase-in periods), and some areas did not have to implement RFG—or opted to implement RFG even though they were not required to do so. Changes in gasoline formulation also occurred over time. RFG was implemented with set seasons indicating that subsets of annual data, rather than annual averages, would need to be investigated.
- *Targeted pollutants.* While specific pollutants were targeted (i.e., reduce NO_x, ozone, benzene), changes in other pollutants were likely to occur, such as reductions in 1,3-butadiene and increases in carbonyl compounds. Other considerations for the introduction of RFG include the changes in total aromatic hydrocarbon content (i.e., could secondary organic aerosol formation be affected?), VOC and NO_x emissions, and sulfur content (i.e., would sulfate concentrations be affected?).
- *Magnitude of control.* The estimated changes in emissions because of RFG were large (i.e., more than 40% reduction in gasoline benzene content), which indicated that changes could likely be detected in the ambient data.
- *Corroborative evidence and hypothesis testing.* Gasoline composition data were available for most areas implementing RFG (although gasoline composition data were not readily available prior to the change) to help explain potential year-to-year differences in ambient benzene concentrations. In addition, some areas of the country did not implement RFG so that the benzene trends in those areas could be compared to the areas with RFG.

Table 2-3. Summary of control information for RFG.

<p>Implementation geographically^a</p>	<p>U.S. Gasoline Requirements</p> <p>Legend:</p> <ul style="list-style-type: none"> RFG - North RFG - South Oxygenated Fuels CA CBG RFG/CA CBG AZ CBG City Fuels/7.8 RVP City Fuels/7.0 RVP City Fuels/6.5 RVP RFG w/Ethanol NV CBG 7.2 RVP 7.6 RVP 7.8 RVP, MTBE No Increase 7.8 RVP 7.0 RVP, 150 ppm S 300 ppm S 800 ppm S <p><small>This map is not intended to provide legal advice or to be used as guidance for state and/or federal fuel requirements, including but not limited to use for RFG compliance requirements. Consult local air departments or attorneys, experts or otherwise, as to the accuracy or completeness of this map.</small></p> <p><small>K.W. Gerner RVP requirements are applicable in the Summer.</small></p>
<p>Major emission source categories</p>	<p>Gasoline-fueled mobile sources</p>
<p>Implementation schedule (incomplete)</p>	<ul style="list-style-type: none"> ● Phase 1 simple model introduced in 1995 ● California RFG (California only) introduced in 1996 ● Phase 1 complex model introduced in 1998 ● Phase 2 complex model introduced in 2000
<p>Targeted pollutants</p>	<ul style="list-style-type: none"> ● Phase 1 (1995): lower Reid Vapor Pressure (RVP) (e.g., butanes), and benzene; add oxygen ● California RFG (1996): lower RVP, benzene, aromatics, olefins, and sulfur ● Phase 1 (1998): lower VOC, toxics; add oxygen ● Phase 2 (2000): lower VOC, toxics, and NO_x; add oxygen
<p>Magnitude of change expected</p>	<ul style="list-style-type: none"> ● > 40% reduction in gasoline benzene content ● Phase 1 (1998): 15% VOC reduction ● Phase 2 (2000): 25% VOC reduction; 6.8% NO_x reduction
<p>Model predictions</p>	<ul style="list-style-type: none"> ● 20% reduction in 1,3-butadiene emissions ● 20% increase in formaldehyde emissions ● Increase in i-butene (if methyl tert-butyl ether [MTBE] used as oxygenate)
<p>Seasonality</p>	<ul style="list-style-type: none"> ● A summer-only program was implemented for RFG. ● A winter oxygenate program was also implemented in some areas.
<p>Scope of program (market penetration)</p>	<p>The California program is statewide; the program is federally mandated for areas with severe ozone problems; some other areas opted in.</p>
<p>Other considerations</p>	<ul style="list-style-type: none"> ● Could reduction in aromatics content lead to change in OC? ● More information is needed about parallel sulfur reduction.
<p>Corroborative data</p>	<p>Gasoline composition information is available for RFG cities from 1995+; pre-RFG gasoline content data are less available.</p>
<p>Other considerations</p>	<p>While vehicle emissions might be reduced by the control, increases in population or vehicle miles traveled could potentially offset the reductions.</p>

^a Graphic courtesy of <http://www.eia.doe.gov/oiaf/servicert/fuel/ozone.html>.

2.4 IDENTIFY POLLUTANT-SPECIFIC TEMPORAL AND SPATIAL SCALES

Different data analysis methods will be required for primary and secondary pollutants because of differences in formation and removal rates and in spatial and temporal variation in concentrations and emissions. These differences complicate the scale (local, regional) of the cause and effect relationship between emissions and the receptor. The pollutant list includes

- Primary: PM_{2.5} mass, CO, OC, EC/BC, NO, VOCs, SO₂, NH₃, metals, and some toxics.
- Secondary: ozone, nitrogen dioxide (NO₂), some portion of PM_{2.5} mass, ammonium sulfate, nitric acid (HNO₃), ammonium nitrate, OC, VOCs, and some toxics.

For the conceptual model, the analyst must identify the key temporal and spatial scales so that appropriate analyses can be selected later. **Tables 2-4 and 2-5** summarize information for ozone and PM_{2.5} mass as examples. Understanding the formation, transport, seasonality, variability, and regional background concentrations of the pollutant helps explain the correlation between control implementation and ambient changes in the pollutant. In the two tables, note that ozone formation is complex, but seasonal and diurnal variability is generally well-understood. PM_{2.5} mass also has complex formation processes, but in contrast to ozone, it also exhibits complex seasonal and diurnal variability, which are not well-understood. Both pollutants have large spatial scales because of the importance of transport.

Table 2-4. Summary of information to consider for ozone.

Formation	A complex series of reactions including sunlight, NO _x , and VOCs. Photochemical modeling indicates ozone formation is sensitive to VOC/NO _x ratios.
Atmospheric residence time	Hours to days
Seasonal variability	Highest concentrations are expected during the warm season. Production of concentrations is increased with extended sunlight.
Diurnal variability	Photochemical production is highest during the day. In urban areas, titration by NO may occur in the early morning and overnight further suppressing ozone concentrations. Less diurnal variability is expected at rural sites.
Day-of-week variability	Weekday-weekend differences are possible for areas in which the VOC/NO _x ratio changes significantly.
Regional background concentrations	Thirty to 40 ppb remote background; regional background concentrations can be significantly higher.
Spatial scale	Local, subregional, and regional. Transport on a regional scale (hundreds of kilometers) can contribute significantly to urban ozone concentrations. Measurements of ozone above ground, but within an altitude that could be part of the mixed layer the next day, have contributed significantly to ground-level ozone.

Table 2-5. Summary of information to consider for PM_{2.5} mass.

Formation	Chemical reaction, nucleation, condensation, coagulation, and cloud/fog processing. Secondary components of PM _{2.5} mass include ammonium sulfate, ammonium nitrate, and secondary organics.
Atmospheric residence time	Days to weeks
Seasonal variability	PM _{2.5} exhibits a seasonal pattern. In areas with sulfate-dominated PM, warm season peaks may be expected (e.g., eastern United States). In areas with high nitrate concentrations, cool season peaks may be expected (e.g., California). High PM _{2.5} concentrations can occur during any season.
Diurnal variability	Diurnal peaks in PM _{2.5} mass concentrations vary by location (e.g., proximity to sources) and by season (e.g., meteorology, source type).
Day-of-week variability	Weekday-weekend differences are not usually observed in PM _{2.5} mass but have been observed for PM components such as EC.
Regional background concentrations	Visually protected area measurements (i.e., IMPROVE ^a) provide typical concentration ranges. Regional contributions of PM _{2.5} can be significant but vary by PM component and region. For example, local contributions of sulfate are very small compared to local contributions of OC.
Spatial scale	Local, subregional, regional, and global. Transport distances range from hundreds to thousands of kilometers. Visible dust and smoke plumes can be tracked by satellite.

^a IMPROVE = Interagency Monitoring of Protected Visual Environments monitoring network.

2.5 IDENTIFY AVAILABILITY OF AMBIENT DATA

Data sets available include routinely collected data such as national and state criteria pollutant monitors (available in the EPA's Air Quality System [AQS]), ozone precursor network (Photochemical Assessment Monitoring Stations [PAMS]), particulate matter networks (Speciation Trends Network [STN], IMPROVE monitoring network), other networks including the Clean Air Status Network (CASTNET) and Southeast Aerosol Research Characterization Study (SEARCH), special studies data (such as PM_{2.5} Supersites), and large-scale field studies, such as the California Regional PM₁₀/PM_{2.5} Air Quality Study (CRPAQS), Central California Ozone Study (CCOS), Texas Air Quality Study (TxAQS), and NARSTO-Northeast.

For trends analysis, **Table 2-6** provides a checklist of important issues about data availability. For an effective analysis of trends, the analyst needs to know the proximity of the monitor relative to emissions, have data of sufficient length of record (preferably several years before and after an emission change), have stable data (i.e., no analytical changes, or if a change

did occur, have knowledge of the effect on the data), and have data of known validity with few gaps in the record. For an emission control that likely affected a broad spatial area, it will be important to have data from nearby sites and urban/rural pairs to understand spatial representativeness and urban excess contributions. Meteorological data are needed for adjusting trends and to improve the understanding of source-receptor relationships. Finally, the availability of previous data analysis results for an area or a monitor can save time and provide additional insight into the investigation.

Table 2-6. Important issues about data availability.

Data Issue	Importance/Relevance
Proximity of emissions to monitor	For some analyses, the analyst may want data collected near a type of emission source (local impact of primary emissions, for example), while for other analyses (regional impact for secondary pollutant, for example), the analyst will want data far removed from the emission source.
Length of data record	<ul style="list-style-type: none"> • Data collected throughout the complete period of interest at a long-term site are vital. • A longer data record is preferable.
Stability, validity, and completeness of record	<ul style="list-style-type: none"> • Few analytical changes, valid data, and sufficient data above the detection limit are desirable. • Complete data for all periods of interest (i.e., hours, days, months, and seasons) are vital to properly represent each period and to enable valid comparisons between periods. • Consistent measurement methods are needed to enable comparisons across years.
Data from nearby sites	Several sites in an area may be needed to understand spatial variation and to document the local impact of a control.
Urban/rural pairs	Urban/rural pairs are desirable for analyses; however, the analyst needs to understand data collection and chemical analysis differences prior to data analysis.
Spatial representativeness of a site (and how it may have varied over time)	Understand site characteristics and possible changes over time (e.g., a suburban site is now considered urban due to growth of the city).
Meta data availability	More details about the monitoring location, sample collection, and analysis are preferable.
Collocated measurements including meteorological data	Additional supporting data are preferable.
Previous data analysis results	Previous characterization of data at a site, including source apportionment and trends analyses, is desirable.

3. PERFORM STATISTICAL AND GRAPHICAL ANALYSES

This section is intended to set up the types of analyses to perform and provide the relationship of the analysis to the conceptual model. A few examples are given in this section to illustrate some of the steps, issues, and pitfalls of performing the types of analyses that are identified in Section 2.

3.1 BASIC DATA ANALYSES

Prior to any analysis, data validation must be performed to ensure the integrity of the data. Data validation steps for ozone precursor data, ozone data, and PM_{2.5} data are provided elsewhere (e.g., Main and Roberts, 2000, 2001; Hafner, 2003). Once the data are validated, basic data analyses should be performed to select the appropriate next steps in the analysis. Basic data analyses include

- Reviewing summary statistics by site, month, and year for similarity among sites, such as local versus regional patterns, urban versus rural differences, and other spatial gradients.
- Inspecting the time series of all data and monthly and annual metrics to identify potential trends, breaks in the data, and diurnal, seasonal, or annual patterns. For example, ozone concentrations peak during the warm season (May through October) throughout the country. PM_{2.5}, in contrast, exhibits high concentrations in either winter or summer seasons (and in some places, both seasons) depending on a monitor's location relative to source types. NO_x is a precursor both to ozone and to PM_{2.5}; thus, the impact of warm-season reductions in regional NO_x versus reductions in cool-season regional NO_x needs to be considered in the technical approach.
- Inspecting day-of-week variations to identify potential influences from emissions sources.
- Investigating species relationships (e.g., scatter plots, correlation matrices) to identify which species co-vary.
- Performing monitor-to-monitor correlations to understand how measured concentrations at one monitor compare to concentrations at other monitors. Monitors with concentrations that correlate well (e.g., $r^2 > 0.75$) with concentrations from a different monitor indicate similar influences. Conversely, a monitor with concentrations that do not correlate with other nearby monitored concentrations may be affected by a local source.

One type of analysis that may arise is the need to identify the impact that a control of a local PM_{2.5} source has on ambient concentrations. A basic analysis should be performed to determine whether the available data prior to the control being implemented shows a spatial gradient—with higher concentrations closer to the source. For example, in Detroit, Michigan, a “bubble” of PM_{2.5} mass in the city, with gradients away from the industrial sites (i.e., the Dearborn site, **Figure 3-1**), indicates influences of local sources on PM_{2.5} mass (and source apportionment results of speciated PM_{2.5} indicate that local influences exist). A similar plot showing the PM_{2.5} composition by site (**Figure 3-2**) also indicates that a possible local influence

from sulfate, OC, EC, and metals (incorporated in the computation of “soil”) at the Dearborn site.

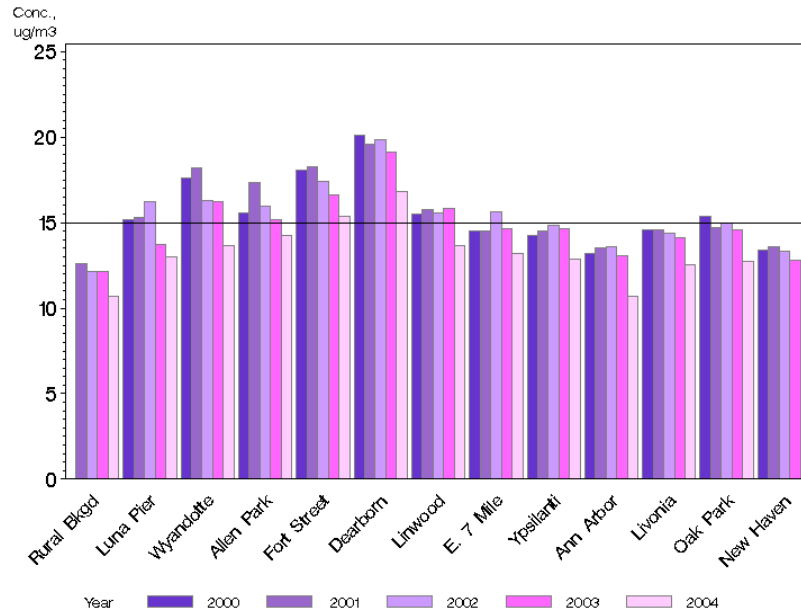


Figure 3-1. Annual average PM_{2.5} mass (2000-2004) concentrations (µg/m³) at sites in the Detroit area (Kenski, 2006). Sites are ordered roughly south to north. The line indicates the PM_{2.5} National Ambient Air Quality Standards (NAAQS).

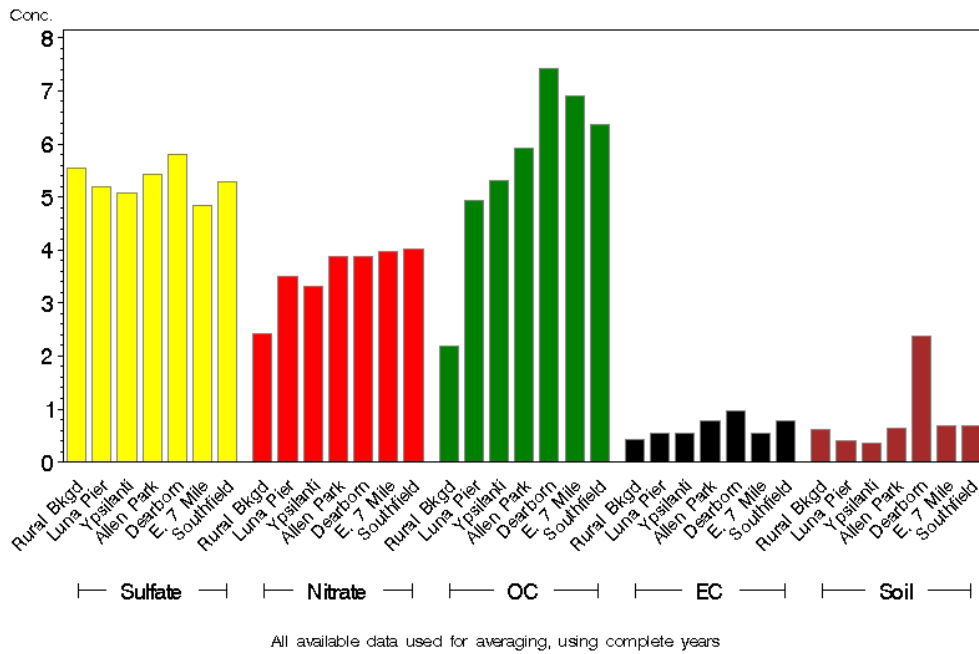


Figure 3-2. Annual average reconstructed PM_{2.5} mass (2000-2004) components (µg/m³) at sites in the Detroit area (Kenski, 2006). Sites are ordered roughly south to north.

A spatial gradient with high concentrations at a monitor close to a source and a decrease in concentrations at monitors further away gives the analyst confidence that the data set can be used to look for trends related to local or regional controls, provided the data record is sufficient and controls were implemented during the data record time frame. Similar analyses characterizing the spatial gradient of a selected pollutant will be necessary to identify locations and pollutants where the influence of local controls may be observed.

Another consideration in the basic analysis stage is the use of derived parameters. For example, ratios (e.g., EC/OC), differences (e.g., urban versus rural), and other derived values (e.g., non-soil iron [Fe]) may be vital to understand how (and how widespread) a control program affected a specific area. It can be useful to understand the regional-level concentrations, as observed at rural sites, compared to the concentrations at urban sites, where the difference is the urban excess. Further analysis of trends in the regional level and urban excess may be important to explain whether overall trends/changes over time are caused by changes in regional level and urban excess levels. An example of an urban excess computation is provided in a trends report in which urban and rural pairs were used to quantify the PM_{2.5} components and the urban excess (Frank, 2005).

Basic analyses help the analyst understand the data set characteristics and limitations and build a conceptual model and hypotheses.

3.2 ASSESSING TRENDS

In a trend analysis, the general approach is to begin with validated data; select a pollutant and/or derived parameter; select indicators (metrics), such as mean, median, maximum, minimum, selected percentiles, etc.; select appropriate time periods to investigate (e.g., season, episode, annual, etc.); apply statistical procedures for detecting trends; evaluate the trend for direction, rate of change, statistical significance, etc.; and compare trends among indicators. A consensus of indicators for trends analyses will reduce the uncertainty of any conclusion. This approach can be applied to raw indicators, or the indicators can be adjusted to remove meteorological influences (Section 3.3).

3.2.1 Metrics for Consideration: Ozone

Statistical indicators to consider for ozone include

- Design values for ozone by year at each site. The design value is the average of the fourth highest value recorded at a site for three consecutive years. A recent study showed that the EPA ozone design value method provides a reasonable estimate of the “true” air quality design value for ozone nonattainment areas and of peak ozone levels within those nonattainment areas for the initial three-year compliance period.
- The average maximum 1-hr and 8-hr ozone concentrations by year at each site. This statistic is another measure of the peak concentrations.

- The average of various percentile bin values (e.g., average of the 1st, 5th, and 10th percentile concentrations) by year at each site. This metric also helps illustrate the distribution of ozone and precursor concentrations.
- The average exceedance concentration for each year at each site. This statistic is used to indicate the average air quality on days when the air is considered poor according to the 1-hr ozone NAAQS.
- The number of exceedances above selected thresholds. This statistic can also be used to gain insight into the nature of the average exceedance concentration, and lends a different perspective than the ranking of the top three exceedance concentrations when a site experiences more than three exceedances within a single year. Similar to the ranking statistic, the identification of exceedance thresholds gives the analyst a sense of whether the highest exceedance concentrations are anomalies or are representative of commonly experienced elevated ozone concentrations.
- A ratio of early morning VOC-to-NO_x measurements. This ratio can be used to assess whether VOCs or NO_x (or both) in the ambient air limit the overall formation of ozone. This insight can be gained despite the complexity of the photochemistry because certain observed ratios of early morning VOC and NO_x concentrations show reductions or increases in the amount of ozone formed over the course of a day (National Research Council, 1991). However, attention to detail is needed in defining and consistently computing the VOC portion of the ratio. VOC definitions and concentrations vary by measurement and analytical method.
- Ozone formation potential using maximum incremental reactivity (MIR) applied to the VOC data. While VOC concentrations may not have changed significantly, the VOC composition and ozone formation potential may have changed over time in response to a control measure.
- Other species concentrations, such as morning CO or individual hydrocarbon species. CO trends may be an indicator of benzene trends (both pollutants are present in motor vehicle exhaust) at some sites, for example. However, this relationship is likely site- and year-specific and may be difficult to use in a trend analysis.

3.2.2 Metrics for Consideration: PM_{2.5} Mass

In addition to design values, average of various percentile bin values, and number of exceedances above selected thresholds, statistical indicators to consider for PM_{2.5} mass include

- Annual PM_{2.5} average concentrations by site and year.
- Composition metrics, such as mass and percent of pollutant contribution to total mass of the major PM components (ammonium sulfate, ammonium nitrate, OC, EC, and soil) by site and year. Overall mass may not have changed over time, but specific components of the PM_{2.5} may have changed.
- Days in a selected air quality index (AQI) range by site and year.

3.2.3 Trends Analysis

Multiple approaches to long-term trends should be considered because individual trend analyses are often limited by uncertainties in the measurements and/or the unavailability of critical measurements. Because the relationship between emissions controls, population growth, meteorology, and air quality is complex, true trends are difficult to establish using any single method. When measurement issues such as missing years of measurements or differences in reporting units are also considered, trends can be less conclusive or misleading. However, the use of multiple trend analyses involving meteorological adjustment techniques suggests when one method is not robust enough or applicable for a particular site.

Of the several approaches to trend analyses, the approaches differ in their measurement requirements and numerical intensity. Even simpler approaches require considerable amounts of air quality measurements (e.g., at least three continuous years of valid ozone exceedance concentrations). More complex approaches have greater measurement requirements (e.g., at least three continuous years of valid daily maximum ozone concentrations, concurrent precursor measurements, and/or concurrent meteorological measurements). To enable more complete analyses, gaps in meteorological measurements must be supplemented with nearby measurements.

The following are examples of trends analysis methods:

- Linear model. Simple linear regression applied to annual summary statistics can be used to quantify trends. The significance of the difference between two years could be confirmed using a t-test or a comparison of confidence intervals around the means of the metric under consideration. The years before and after a change can be grouped to partially remove meteorological changes among years.
- Nonparametric methods. These approaches test for and estimate a trend without making distributional assumptions using such techniques as the Mann-Whitney U test, Spearman's rho test of trend, and Kendall's tau test of trend. The Mann-Whitney U test is analogous to the t-test; however, the Mann-Whitney U test is based on the ranks of the values within each year, not the actual values. Thus, it essentially is a test based on the median values that does not assume any underlying distribution for the data. Nonparametric tests provide the advantage of not assuming a linear trend and are reasonably robust against outliers.
- Time series models. Statistical modeling of ozone concentrations using time series models takes into account the serial dependence of the concentrations. An example of time series model is the auto-regressive integrated moving average—ARIMA.
- Extreme-value theory. This method estimates the distributions of annual maximum hourly concentrations and estimates the distributions of the number of days exceeding the NAAQS. Examples include the Chi-square test and Poisson process approximation.

In order to guide the analyst in selecting the appropriate trends analyses methods, data completeness and the metrics under consideration need to be assessed. It is useful to initially, and more qualitatively, inspect trends in the data by generating maps that show the trends of selected metrics at all sites (or combinations of sites), such as **Figure 3-3**.

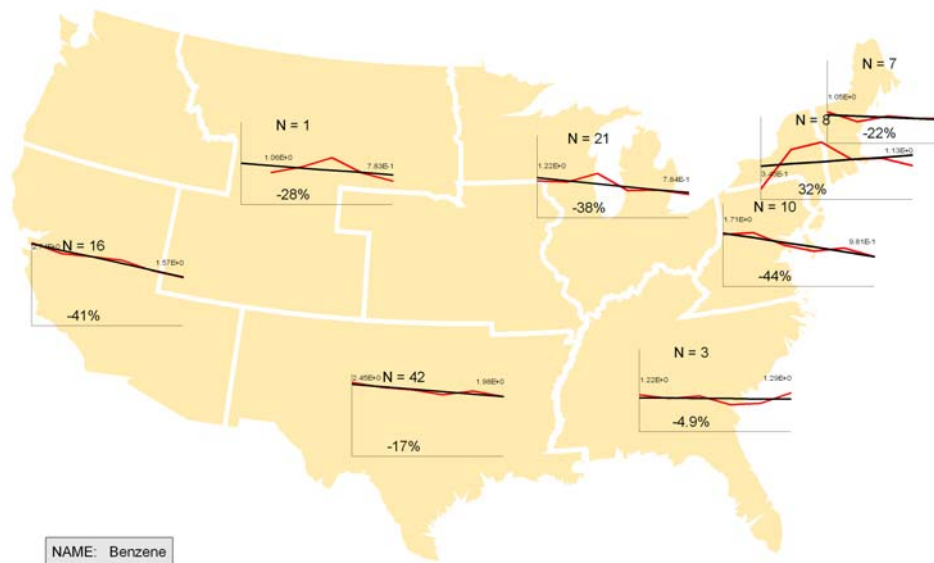


Figure 3-3. Preliminary map of annual average benzene concentration ($\mu\text{g}/\text{m}^3$) trends from 1990-2005 by EPA region. N is the number of sites included in the trend; the percent change shown is based on the linear regression line. Site data in 75% of the trend period years needed to be included in the trend.

Figure 3-3 also helps illustrate another consideration when investigating trends. The trend for Region 2 shows the only upward trend among the regions over the entire trend period. However, if the first year or two of data were not included, a downward trend would be observed. This example points out that trend assessments can be misleading because different groupings of years can lead to different trends. Consistent sets of sites and years should be used in trend assessments.

3.3 ADJUSTING FOR METEOROLOGICAL IMPACTS

Day-to-day variations in pollutant concentrations may be caused by meteorology alone. When changes in emissions over decades are considered, the influence of emissions on pollutant concentrations becomes more significant. The variations in pollutant concentrations caused by meteorological influences can conceal or mask the influence of emissions reductions. Therefore, when looking at trends in ambient pollutant concentrations, it is desirable to remove the meteorological influences from the trends.

It has long been known that weather conditions have a strong influence on ozone concentrations, for example. Because of this, multi-year cycles in weather can influence multi-year ozone and ozone precursor trends. Therefore, when trying to explain ozone and ozone

precursor (or other pollutant) trends that are caused by changes in emissions, the analyst needs to account for the influence of weather on the trends. To properly perform this “meteorological adjustment” to the trend requires (1) the use of mathematical techniques that account for the influence of weather on long-term air quality trends and (2) the use of data that capture the full range of meteorological processes that influence the pollutant.

3.3.1 Mathematical Techniques for the Meteorological Adjustment of Air Quality Trends

Mathematical techniques to statistically adjust ozone concentrations to account for meteorological variables have been developed (Thompson et al., 2001; Porter et al., 2001). Different approaches have various advantages and disadvantages depending on the type of ozone metric and monitoring site being investigated. Therefore, it is best to investigate various techniques to achieve the most accurate meteorologically adjusted trends. These techniques have been less broadly applied to PM_{2.5} data and even less work has been done with air toxics. An overview of available approaches follows:

Regression-based Models. Linear regression-based models are the simplest method for the meteorological adjustment of trends. These models use linear and additive relationships between the pollutant and meteorological variables to adjust pollutant trends. While relatively simple, these models are open to criticism because ozone and PM_{2.5} formation and some meteorological variables do not have a linear and/or additive relationship.

Tree-based and Stratified Models. Tree-based and stratified models adjust ozone trends in two steps based on differences in the association between ozone and meteorology under different meteorological regimes. Typically, these models use Classification and Regression Trees (CART) or cluster analyses (Larsen, 1999) to identify the different meteorological regimes under which ozone formation occurs. Regression analyses are then applied separately for each regime to quantify the trends. This two-part analysis provides the added benefits of better identifying meteorological regimes under each cluster (which may lead to more sensitive models of extreme values) and accommodating seasonal effects.

Extreme-value Models. The inherent averaging in regression models makes them poor predictors of extreme values that can be important for attainment designations. Extreme-value models avoid this problem. One example is the approach used by Cox and Chu (1993; 1996). In this technique, concentrations are adjusted by fitting ozone measurements to a Weibull probability distribution as a function of multiple meteorological parameters. This process is somewhat labor-intensive and requires specialized software, but is preferable when modeling the highest ranges of concentrations rather than using regression-based models, which can be unduly influenced by extreme values.

Filter-based Models. Filter-based models adjust ozone and its precursor concentrations by smoothing out the non-trend variation in the data (Rao et al., 1995; Flaum et al., 1996; Milanchus et al., 1997; Porter et al., 2001). For example, ozone varies on diurnal, synoptic, seasonal, annual, and interannual frequencies. Moving averages or band-pass filters can be used to remove the higher-frequency variations in ozone concentrations to identify underlying trends. These models assume that meteorology affects ozone concentrations at short- and long-term

scales and that these components are separable. Applying a filter to the data removes shorter-frequency variability while leaving the long-term underlying trend. A disadvantage of this type of model is that it requires a data set that is at least three to four times longer than the longest-frequency meteorological variation in order to identify and remove that variation. This technique requires at least three to four years of historical data for development.

3.4 CORROBORATIVE ANALYSES

Once the conceptual model has been prepared, trends in ambient data identified and quantified (with and without meteorological adjustments), and possible control effects identified, the analyst can improve the confidence in the results by finding corroborative evidence for correlation between the trends and the controls. In addition, the null hypothesis must be tested. For example, Main et al., (1998) verified that ambient benzene did not decline at a site without RFG. Multiple methods for detecting and quantifying trends in air quality caused by changes in emissions (i.e., consensus) are needed to provide more confidence in the conclusions.

Corroborative analyses include

- Trajectory analyses to separate local and transport days or identify source areas. These results can then be used to further stratify and investigate the ambient data and verify the source areas versus the spatial coverage of the control implementation. Other investigations of transport include flux calculations to show the relative magnitude of a pollutant or precursor concentration across a boundary or ventilation/recirculation measures to show the relative importance of transport versus stagnation.
- Source apportionment (positive matrix factorization [PMF], chemical mass balance [CMB], principal component analysis [PCA], and others) applied to data collected before and after a change, for example, to investigate changes in source profiles and source categories.
- Modeling or application of methods for estimating relative emission contributions (Transported Emissions Assessment Kit [TEAK], Section 3.4.3) to identify source areas or quantify expected changes.
- Other measures of emissions, such as population and vehicle miles traveled (VMT), to illustrate, in the absence of a decline in ambient concentrations, what changes may have occurred but were obscured by increases in emissions caused by more emitters.

3.4.1 Transport and Trajectories

The following issues should be considered when assessing pollutant transport:

- Definition of boundaries (and scale). Over which boundary is transport being assessed? Where do pollutants and precursors transported into an area originate? What route(s) do they take?

- Pollutants and continuing reactions. The pollutant and its precursors need to be considered. How much of the high pollutant concentrations can be attributed to transport?
- Transport versus carryover (recirculation). Are pollutants transported into the area of interest; are the pollutant and precursors from the area carried over from day to day; or is the area experiencing a combination of transport and recirculation? How frequently does transport occur?
- Aloft and surface air quality and winds. What data are available and where are the sites located? Are special studies data available?
- Transport time. How long does it take for air parcels from upwind sources to reach the receptor? When do pollutants and precursors arrive in the area?
- Characteristics along the transport path. Are important pollutant or precursor emissions sources situated along the transport path?

Methods for quantifying transport include computing the relative emissions in the upwind and downwind areas using ratios of precursor emissions in the upwind airshed to those in the downwind airshed; comparing the ratio of upwind to downwind emissions with emissions accumulated along a typical trajectory path; and computing the ratio of upwind to downwind emissions using meteorological and photochemical models.

Many tools are available for preparing trajectories and exploring the differences between dates. A common approach is to prepare 72-hr backward trajectories for all sample dates using the National Oceanic and Atmospheric Administration (NOAA) Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler and Hess, 1997). For 24-hr sampling periods, trajectories are prepared for every three, four, or six hours at two to three heights. These trajectories can then be mapped as a spatial probability density (SPD⁰):

$$SPD = \frac{\text{Count of hourly trajectory endpoints within search radius}}{\text{Count of trajectories run}} \quad (3-1)$$

The largest SPD values are in areas where the backward trajectories have spent the most time.

A conditional probability function (CPF) can then be applied to help interpret the SPD results (Kim and Hopke, 2004; Kim et al., 2003, 2004; Ashbaugh et al., 1985). In CPF, the transport patterns of the 20%-highest concentration days for a source contribution are compared to the climatological transport patterns. After finding SPD⁰, backward trajectories for the 20%-highest concentration days are run and mapped (SPD'). This density is then compared to the SPD for all days (i.e., the climatology), so that the differences in transport and source areas on high concentration days of a given pollutant or source contribution are highlighted:

$$CoPIA' = SPD' - SPD^0 \quad (3-2)$$

This Conditional Probability Integrative Analysis (CoPIA) is similar to the CPF analyses employed in other studies (Kim and Hopke, 2004; Kim et al., 2003, 2004; Ashbaugh et al.,

1985); however, CoPIA is adapted to take advantage of tools available in a geographic information system (GIS) framework. An example is shown in **Figure 3-4**.

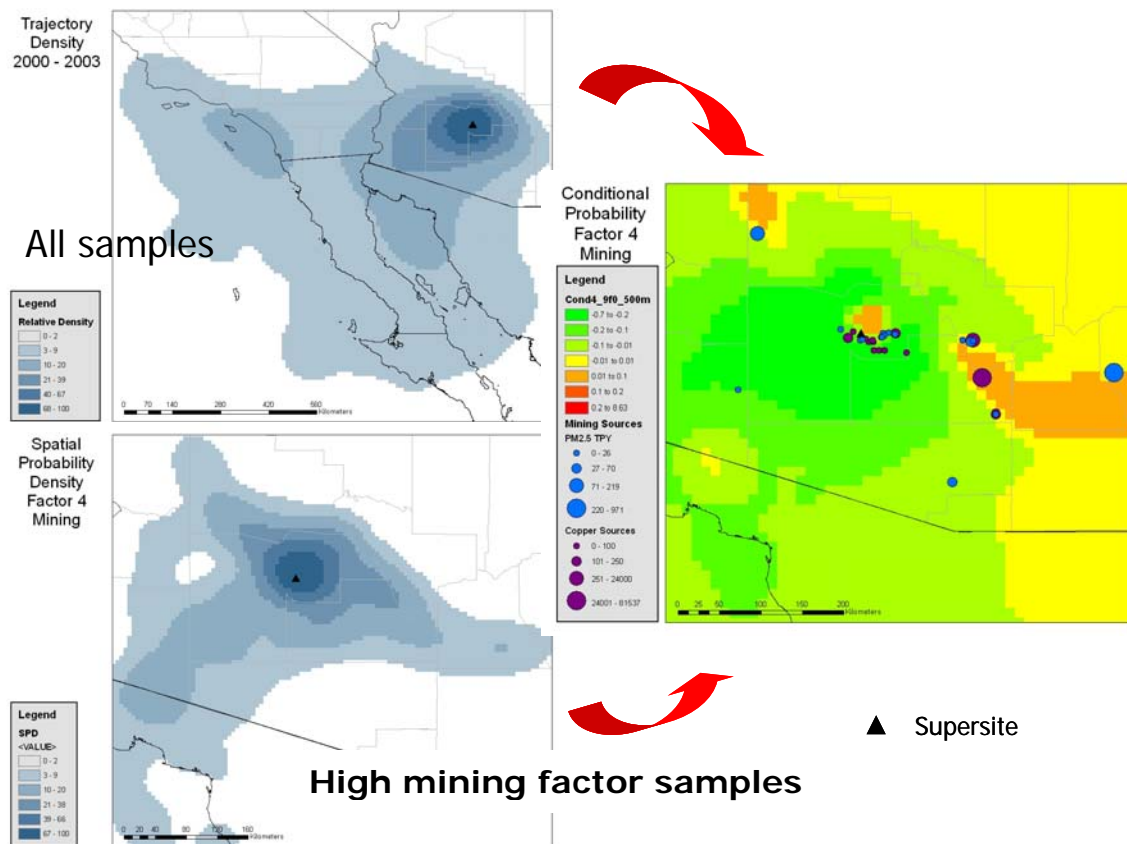


Figure 3-4. Spatial probability density plots for all samples and for samples with high contributions from a mining factor identified in source apportionment of $PM_{2.5}$ data collected in Phoenix, Arizona, during 2001-2003 (Brown et al., 2005). Also shown is the conditional probably plot overlaid with mining emissions.

In the example (Figure 3-4), source apportionment results for Phoenix, Arizona, $PM_{2.5}$ data were investigated (Brown et al., 2004). Composition of one factor identified in the analysis was consistent with copper mining emissions. On days with higher contributions from this factor, CoPIA and the emission inventory indicated a higher probability that air parcels were transported from an area with significant copper mining operations.

A similar approach was used by Grego et al., (2006) to investigate the impact of NO_x emissions reductions on ozone in the eastern United States. One of the approaches used in this work was to compare pollutant concentrations on days when air parcels were transported from the Ohio River Valley to days on which no transport occurred.

Limitations to trajectory analyses exist, such as spatial scale, that need to be considered, especially in investigating local influences. The commonly used HYSPLIT model grid-spacing is too large (40 km) to differentiate between “local” and transport influences. Local-scale

meteorology may not be modeled well—such as onshore/offshore flow near large bodies of water. It is clearly important to ensure that a trajectory analysis is appropriate and representative of the scale of interest.

3.4.2 Source Apportionment

Source apportionment analyses using receptor models are useful in quantifying the impact of specific sources and source types on ambient concentrations and identifying how these sources affect specific pollutants over time. The two receptor models currently in wide use are CMB (Watson et al., 1990) and PMF (Paatero, 1997; Paatero and Tapper, 1994). CMB can be applied to a single sample and quantifies sources by correlating known source profiles to ambient data; thus, correct and area-appropriate source profiles are vital. PMF quantifies sources by extracting the source profiles based on the internal variability and uncertainty of the ambient data and, therefore, requires larger data sets with well-characterized uncertainties. Both models can provide a wealth of information, but results need to be put in context; thus, development of a conceptual model and use of trajectory analysis are needed to fully understand source apportionment results.

CMB and PMF have been applied to routine and special study speciated $PM_{2.5}$ data at many rural and urban locations across the United States. Routine speciated $PM_{2.5}$ data are available at a number of rural locations as part of the IMPROVE monitoring network since the early to mid-1990s, while data collection started in urban areas as part of the Speciation Trends Network (STN) in 2001. Special study data, including molecular analysis of OC, have also been collected and analyzed. Thus, a fairly large amount of data are available for source apportionment of $PM_{2.5}$, especially after 2001. VOC data, mainly collected as part of Photochemical Assessment Monitoring Stations (PAMS) and special studies, have been used in source apportionment applications, with most analyses in the 1990s using CMB; more recent applications of PMF to VOC data have been made. Long-term speciated VOC data exist only at a handful of sites. Lastly, receptor models have rarely been applied to air toxics, continuous PM, and criteria pollutant data. Limitations to source apportionment include lack of site-specific source profiles (CMB), lack of detection limits low enough so that sufficient data are available (air toxics, trace metals), and lack of continuous data from source-specific markers (both CMB and PMF).

Source apportionment analyses can be applied to ambient data to understand changes due to known regulations by quantifying changes in

1. Concentrations of total mass (i.e., $PM_{2.5}$ or total VOC) or of specific components (i.e., benzene or lead) attributed to a specific source or source type using an “average” profile over the time period. Either model can be applied to a data set that spans the time frame of the regulatory change, while either using (CMB) or determining (PMF) an “average” profile over the time period. This method may have limitations because of the use/determination of an “average” profile.
2. Profiles (i.e., fingerprints) of specific sources or source types (PMF only). CMB requires a known profile; this analysis using PMF could then be used to provide “before” and “after” profiles for CMB.

- Both profiles and contributions to ambient concentrations. For CMB application, the changes in mass attributed to a specific source before and after a regulatory change will rely solely on accurate source profiles of all sources before and after the change—this is a challenge. For PMF application, sufficient data before and after the change are needed; typically, at least 100 samples are desirable, although more than 300 samples before and after the change would yield more certain results. Thus, if accurate profiles are available before and after a known regulatory change but data are limited, CMB application is likely the best method. PMF should be applied by itself, or concurrent with CMB if sufficient data are available.

To ensure quality results, analytical and collection methods need to be the same between source profiles and ambient data, or the differences in data between methods need to be known and quantified.

Source apportionment analyses have been applied to data sets over a long period of time in few locations. In Los Angeles, California, CMB and PMF analyses have been performed on VOC data; an example summarizing the average results in 1987 (CMB) and 2001-2003 (PMF) is shown in **Figure 3-5**. It shows that total VOC concentrations have decreased over time, mostly because of a decrease in mobile source influence. While mobile sources have decreased, non-mobile sources have not changed.

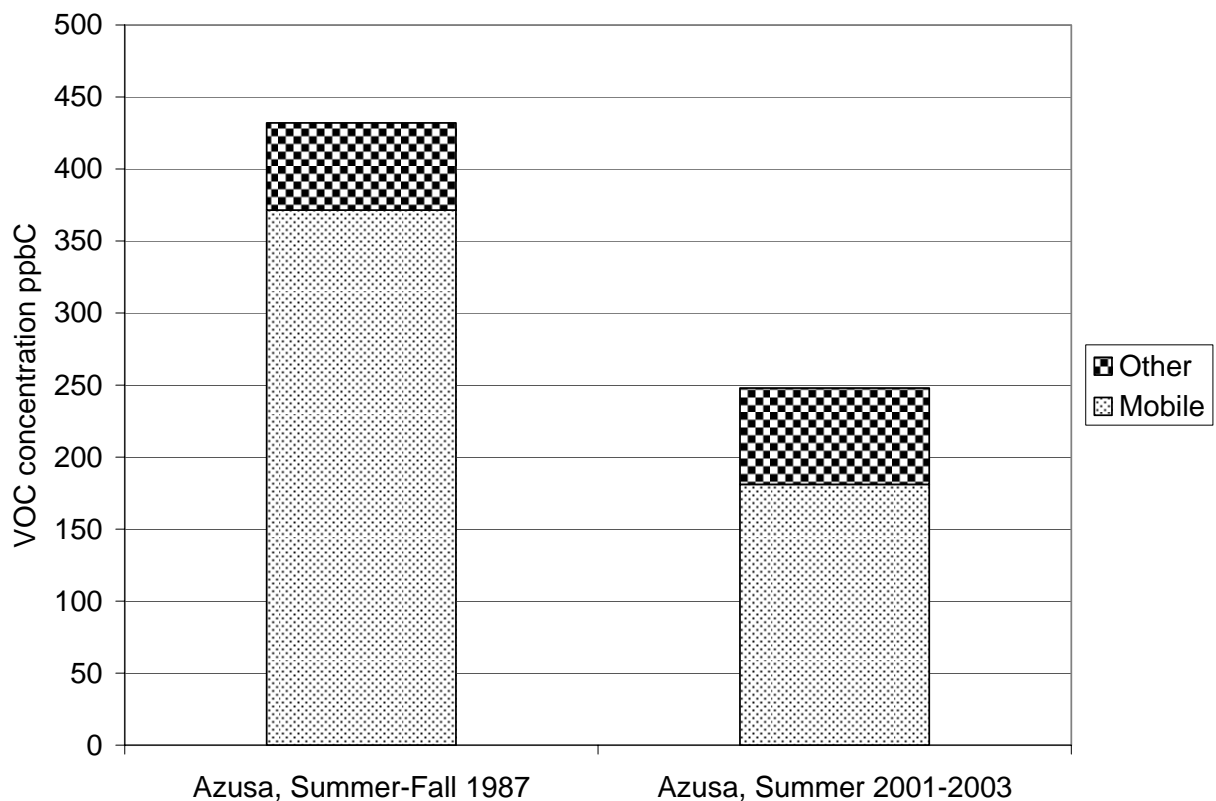


Figure 3-5. Average VOC mass (ppbC) apportioned to mobile and non-mobile sources at Azusa in Los Angeles, California, in 1987 (Harley et al., 1992) using CMB and in 2001-2003 (Brown et al., 2005) using PMF.

3.4.3 Estimating Relative Emission Contributions

Performing comparisons between trends in emission inventories and trends in ambient monitoring data requires an understanding of which emissions sources are likely impacting a given monitor during a specified time period. A suite of desktop tools has been developed called the Transported Emissions Assessment Kit (TEAK) for examining the probability of transport, source locations, and emissions with respect to air quality issues. TEAK employs SPD to explore where air masses originate and travel under certain conditions (e.g., on days with poor visibility or days with high ozone concentrations). SPD uses GIS technology to convert a large ensemble of modeled trajectories into a quantitative surface density plot that is easy to visualize. In addition, SPD output can be combined with emission inventory information to assess the emission impact potential (EIP) of a given source or geographic region. The result is an estimate of the relative potential for emissions from different facilities, counties, or grid cells to impact the receptor site.

The SPD is used to weight the emissions from individual counties and estimate the potential for specific upwind areas to impact the receptor. The EIP of any county is calculated as

$$EIP = \frac{E_p * D_0}{f(\text{distance})} \quad (3-3)$$

where:

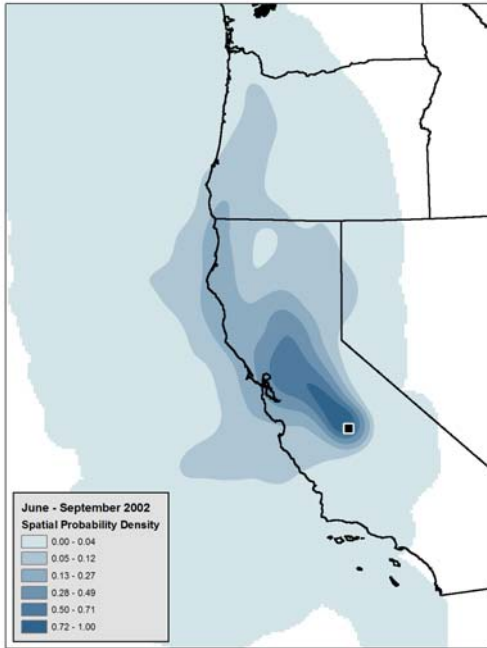
E_p = county total emissions of pollutant p

D_0 = spatial probability density at the county centroid

f = function of distance between county and receptor

Using a tool like TEAK can help ensure that the emission trends analyses are comparing emissions most likely impacting a given site—something that can change over time. For example, overall transport patterns for the Fresno, California, summer ozone seasons of 2002 and 2004 were analyzed and SPD plots produced for each ozone season. **Figure 3-6** shows that the SPD plots are very similar for these two years. However, **Figure 3-7** shows the difference between the two plots and reveals that in 2002, transport from the north of the San Joaquin Valley (SJV) was more likely (up to 10%) than in 2004. In summer of 2004, Fresno experienced about 25% more transport from the Pacific Coast. In terms of ozone exceedances, 2004 was a cleaner year than 2002.

(a)



(b)

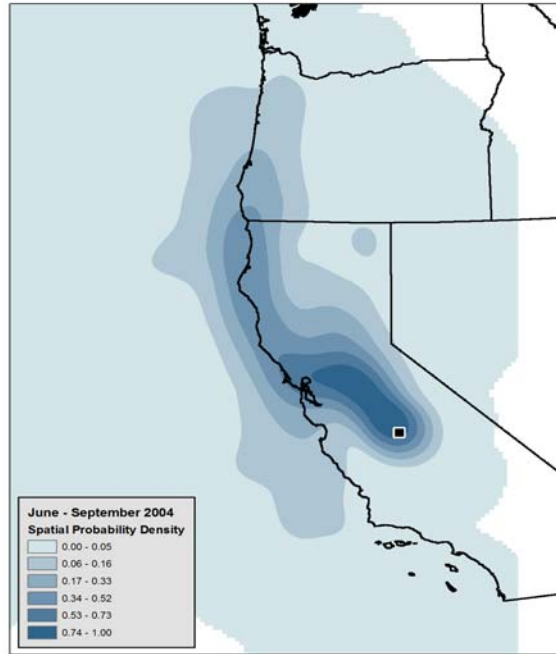


Figure 3-6. Spatial probability density plots for Fresno, California, for the summers of (a) 2002 and (b) 2004.

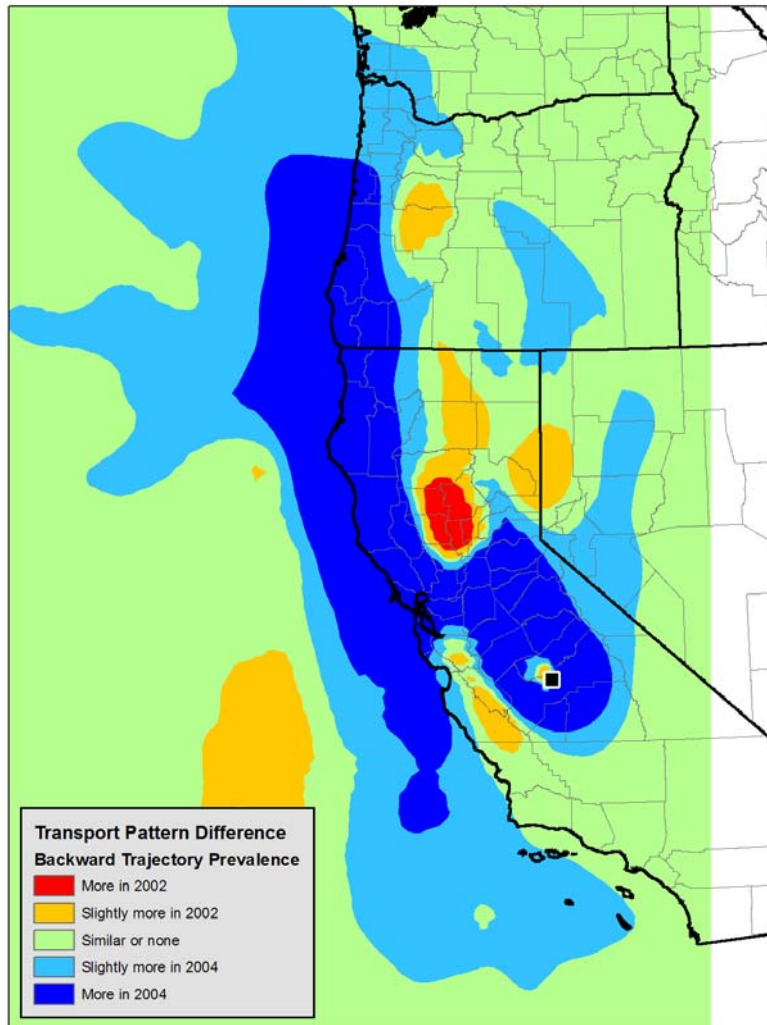


Figure 3-7. Transport pattern differences between the summers of 2002 and 2004 for Fresno, California.

The EIP may be divided by a distance function to roughly account for dilution and increased uncertainty in model outputs far from the receptor site. However, for the example shown in **Figure 3-8**, f equals 1. Figure 3-8 shows the SO_x and NO_x EIP values by county for the 20%-worst and 20%-best visibility days for measurements made at Hercules-Glades, Missouri. These analyses were used to help the central states assess important clean-air corridors¹ and the importance of emissions sources within and outside the central states that contribute to poor visibility.

¹ In this example, clean-air corridor is defined as the transport pathway predominantly associated with 20%-best visibility days.

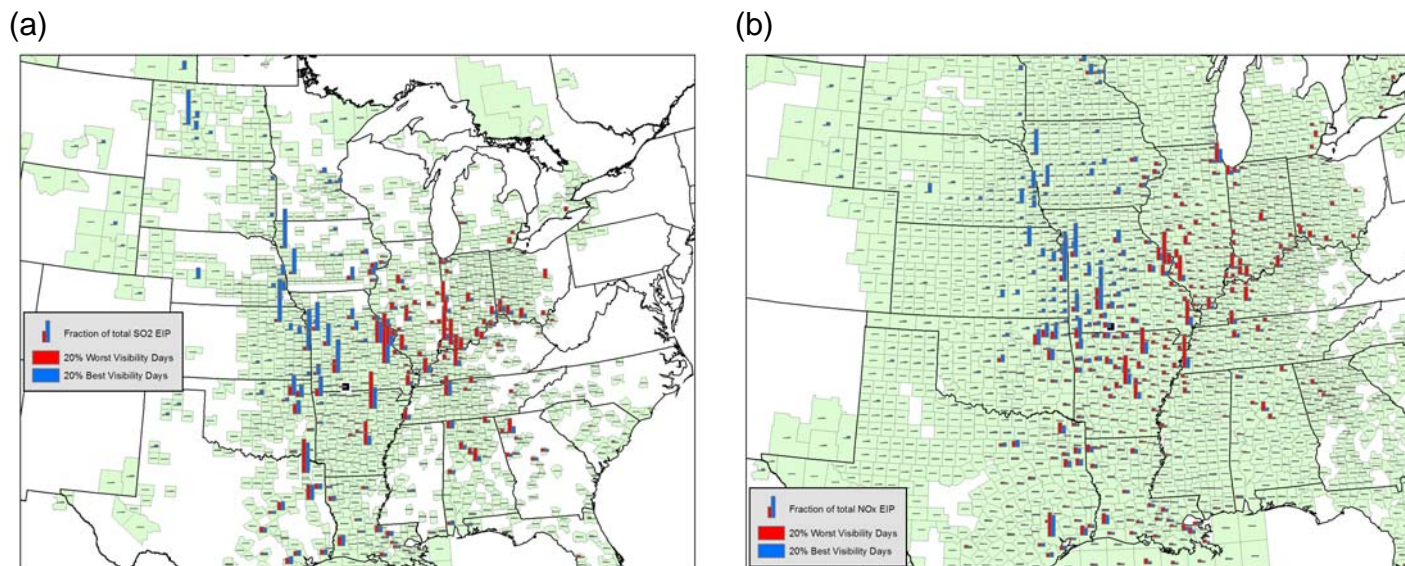
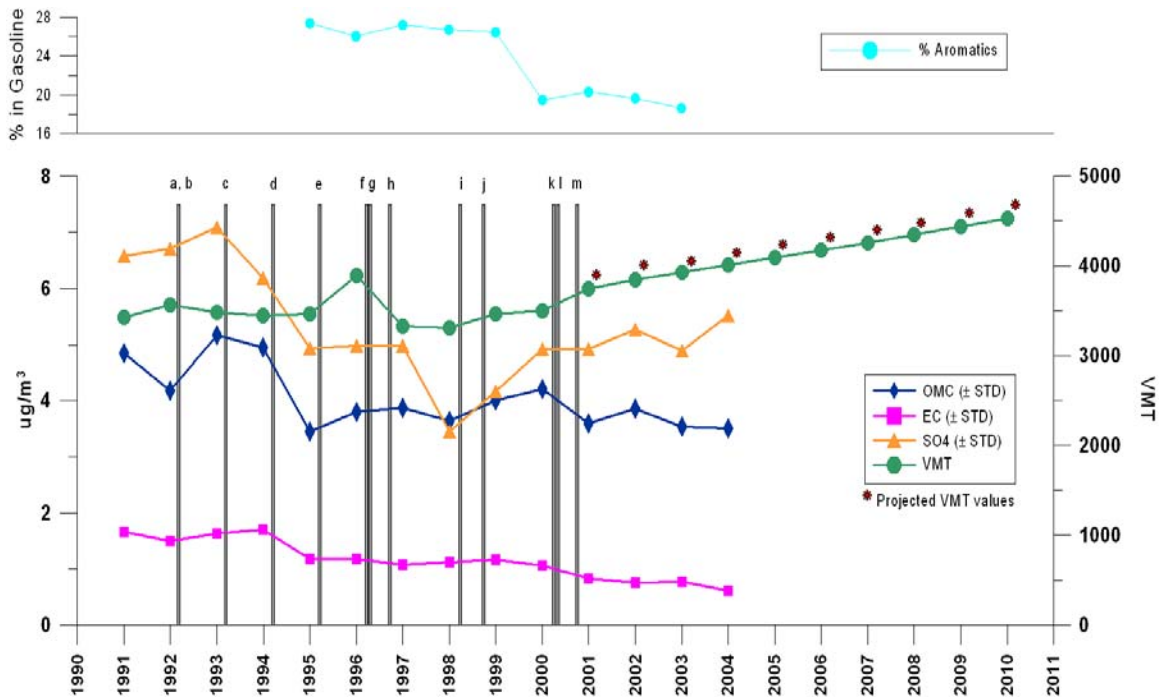


Figure 3-8. Geographic distributions of (a) SO₂ and (b) NO_x EIP for the 20%-worst visibility days (red bars) and 20%-best visibility days (blue bars) observed at Hercules-Glades, Missouri.

3.4.4 Other Corroborative Evidence

Other corroborative evidence or hypothesis testing is needed to add confidence to the results. **Figure 3-9** shows an example exploration of the ambient changes in PM_{2.5} organic carbon mass (OM = 1.8*OC) concentrations potentially tied to gasoline regulations, including the introduction of RFG in Washington, DC. This example combines several of the considerations discussed in the technical approach.

The premise behind the example was that RFG targeted reductions in the aromatic content of gasoline, and aromatic hydrocarbons have the potential to form secondary organic aerosol (SOA), which is a component of OM in PM_{2.5}. The question was asked whether future changes in gasoline formulation might result in OM reductions, and the historical data record was explored to identify evidence of such a link. Figure 3-9 shows the gasoline aromatics content in the top portion; aromatics content of pre-1995 gasoline was about 32%. The lower half of the figure shows annual average concentrations of OM, other PM_{2.5} components not expected to be affected by the change in gasoline formulation (nitrate, metal oxides [soil]), and VMT estimates. The controls that may affect OM concentrations are summarized in the accompanying table.



Graph Key	Regulation	Month	Year
a	Winter-Oxygenated, Season 1	11	1992
b	Vehicle Inspection/Maintenance Program	11	1992
c	Winter-Oxygenated, Season 2	11	1993
d	Winter-Oxygenated, Season 3	11	1994
e	Reformulated Gasoline (RFG) – Stage I	1	1995
f	Winter-Oxygenated, Season 4	11	1995
g	Prohibition of Leaded Gasoline for Motor Vehicles	1	1996
h	Final Phase-In of Tier 1 Standards	8	1996
i	Gasoline Distribution Stage I	12	1997
j	National Low Emissions Vehicle Program – Stage I	8	1998
k	Stage II Vapor Recovery	12	1999
l	Reformulated Gasoline (RFG) – Stage II	1	2000
m	National Low Emissions Vehicle Program – Stage II	8	2000

Figure 3-9. Corroborative information including vehicle miles traveled, gasoline composition, and controls to investigate changes in PM_{2.5} components in Washington, DC (Hafner et al., 2006).

Concentrations of OM and EC were significantly higher from 1991 through 1994 than from 1995 through 1998. This is also true for sulfate and nitrate but is not true for soil. An

analysis of the annual average concentrations does indicate that OM and EC concentrations decreased coincidentally with the introduction of RFG in 1995. However, the decrease in concentrations of other PM mass components like nitrate and sulfate makes attribution of this decrease problematic because it could simply be a systematic analytical or network difference. An additional analysis of major components of PM mass at the nearby rural Shenandoah monitoring site indicated that OM and EC concentrations were unchanged from 1994-1995. Personnel at the University of California, Davis, Crocker Nuclear Laboratory, which operates the IMPROVE monitoring network, confirmed no network changes during this time (White, 2005). Thus, it is unlikely that the decrease in OM or EC concentrations in Washington, DC, was caused by a systematic analytical or sampling change.

Other considerations in this example were not fully explored during this analysis but would be useful to improve the understanding of the relationship between the introduction of RFG and ambient OC changes. For example, meteorological adjustments were not made to the data to account for differences from year to year. However, statistically significant differences between three-year averages before and after the 1995 RFG introduction indicate that meteorology may not be the sole source of the changes. Other indicators to test include trends in the sulfate, nitrate, OM, and EC fractions in addition to the concentrations. Other hypotheses might be tested:

- Did SO₂ emissions decline over the same time period?
- Did ozone concentrations decline during this period, and, thus, change the photochemical production of OC (and possibly sulfate and nitrate)?
- Were similar changes observed in other cities with (or without) RFG introduction during this time frame?

This example illustrates the need to critically think through the analysis and identify the issues and considerations. Once the questions have been asked, the analyst can develop an approach to answer them.

4. SELECTING EXAMPLES FOR CASE STUDIES

4.1 OVERVIEW

We begin the process of selecting application examples for the technical approach discussed in this document. A spectrum of cause-and-effect types needs to be considered. For example, a local control/local effect case study could be one of the simpler analyses in which a regional effect is not expected, the emission control influenced a primary pollutant, and sufficient data existed before and after the change (e.g., Philadelphia, Pennsylvania, or St. Louis, Missouri, lead concentrations). In contrast, a combination of applied regional and local emissions controls affect a range of primary and secondary pollutants over varying spatial scales, thus complicating an individual analysis of one control and the resulting air quality.

Candidates for potential controls investigation include NO_x reductions (e.g., NO_x SIP call), SO₂ reductions (Acid Rain program), RFG, diesel retrofits, and maximum achievable control technology (MACT) (for air toxics). Earlier discussions with the EPA indicate that several cities are of interest for this analysis as well. The following sections provide ideas for consideration.

4.2 NO_x AND SO₂ REDUCTIONS

The objective of this control scenario is to assess the impact of NO_x and SO₂ emissions reductions on ambient air quality in a selected city. A likely candidate city is Detroit, Michigan, because of its wealth of data and mix of local and transported emissions. Using the thought process described in Sections 2 and 3, the following items will need to be addressed:

- *Air pollutants expected to be affected.* NO_x, SO₂, ozone, PM_{2.5} mass, PM sulfate, and PM nitrate are pollutants expected to be affected. As shown in Figures 3-1 and 3-2, speciated PM_{2.5} data are available for 2000-2004. Because ozone is a consideration due to the NO_x reductions, VOCs are also important.
- *Spatial scale of pollutants.* This assessment would likely focus on the secondary components (sulfate, nitrate, ozone) and the PM_{2.5} mass, which is a mix of primary and secondary components. A mix of regional (sulfate, nitrate, ozone) and urban-level (nitrate) scales needs to be addressed. Regional-level concentrations are important, so a mix of urban and rural monitoring data is needed.
- *Spatial scale of the control measure.* The NO_x SIP call and Acid Rain programs affected power generation and large industrial facilities. One area experiencing the most reductions was the Ohio River Valley. Other controls of NO_x may be important. As with the pollutants, a mix of regional- and urban-level scales needs to be addressed. Knowledge of local controls in Detroit is needed, such as whether other controls were implemented for PM_{2.5} mass.
- *Timing and magnitude of changes.* A summary of NO_x and VOC emissions controls is summarized in the EPA (2005) evaluation of the NO_x SIP call. A summary of the SO₂

reductions needs to be prepared, including information about the potential magnitude of changes in air quality.

- *Magnitude of expected ambient changes.* The EPA prepared model predictions at a regional level, at least for ozone. A summary of model results needs to be compiled.
- *Statistical metrics to consider.* Focusing on the PM_{2.5} mass in Detroit, seasonal and annual averages of PM_{2.5} mass and composition need to be investigated. Meteorological adjustments are likely important to remove meteorological influences.
- *Basic analyses.* Section 3.1 analyses need to be performed (or previous analysis results reviewed). The spatial patterns in PM_{2.5} and its components by season and year, including urban-rural comparisons, are of particular interest for analysis. The spatial pattern in the PM_{2.5} mass data indicates that if a control on industrial PM or precursor emissions were implemented, the effects would likely be observed at the Dearborn site, for example.
- *Corroborative analyses.* Separating the days dominated by transport from more local days is important for this assessment, but hampered by the model resolution. High sulfate or nitrate concentration days should be examined with SPD plots, for example. Extensive source apportionment analyses have been performed with data from Detroit; these analyses can be revisited to look for trends over time.
- *Other considerations.* Less emission inventory information is available from the Canadian sides of the Detroit area, which complicates an assessment of trajectories coupled with the magnitude of emissions.

4.3 MULTIPLE POLLUTANT REDUCTIONS

The objective of this control scenario is to assess the impact of RFG introduction on multiple pollutants in a selected city. A likely candidate city is Houston, Texas, with other choices including Phoenix, Arizona, and Washington, DC (initial investigation is shown in Figure 3-8); previous analysis of VOCs includes Main and O'Brien (2001) and Main et al. (1998). Using the thought process described in Sections 2 and 3, the following items need to be addressed:

- *Air pollutants expected to be affected.* For RFG, NO_x, ozone, total VOC, benzene, total aromatic hydrocarbons, 1,3-butadiene, formaldehyde, and possibly PM_{2.5} mass, nitrate, OC, and EC could be affected. Key to this analysis is the availability of speciated VOC data before and after the change in gasoline formulation. Highly time-resolved data (i.e., a 1-hr to 3-hr duration) are needed because of complicated meteorological and monitoring site issues. A focus on sampling during the morning rush hour should be considered because motor vehicle emissions are highest, mixing heights are low (thus, concentrations are high), and reactivity is minimized.
- *Spatial scale of pollutant.* The focus of this assessment is primary and secondary pollutants, with an emphasis on the urban scale. Local influences near the monitors need to be considered as well. Transport at this scale is important; thus, meteorological data collocated with the air quality data will be required.

- *Spatial scale of the control measure.* RFG was applied to a selected urban area. If Houston, Texas, was selected for study, the production of gasoline in the area would mean that the industrial emissions might be affected by RFG introduction as well. Other VOC or NO_x controls in the area need to be identified. For Houston, offshore VOC and NO_x emissions should also be considered.
- *Timing and magnitude of changes.* RFG changes to benzene, aromatics, VOC, and NO_x emissions have been documented and should be available for the Houston area.
- *Magnitude of expected ambient changes.* Model predictions have been made for ozone changes caused by the introduction of RFG. Early model work indicated formaldehyde increases (if MTBE was used as the oxygenate) and 1,3-butadiene decreases could be expected with RFG. A summary of model results needs to be compiled. Less is known about the potential impacts on PM_{2.5} mass and components from RFG.
- *Statistical metrics to consider.* Focusing on the VOCs, statistical summaries of concentrations, weight percents, and ratios would be needed. For ozone, the typical suite of metrics should be explored. Meteorological adjustments are likely important for ozone; less work has been performed on the VOCs. The VOCs of interest are influenced by temperature and photochemical production and destruction, so it is likely that meteorological adjustments need to be explored.
- *Basic analyses.* Section 3.1 analyses need to be performed (or previous analysis results reviewed). Of particular interest in this example is wind direction relative to mobile and industrial sources. **Figure 4-1** shows a satellite photo of the Clinton Drive site in the Houston Ship Channel. The U.S. I-610 freeway is located west of the site, a residential area located to the north, and industrial sources located to the south and east. To isolate the mobile source emissions, samples when winds are from the west are needed. Data from other sites outside the industrial section of town are also available for corroborative analyses.
- *Corroborative analyses.* Analysts need to look for changes in pollutants not expected with the introduction of RFG. For example, in the PM_{2.5} data, changes in the soil component would not be expected with the introduction of RFG. For the VOCs, acetylene or the biogenic isoprene should not have been affected the introduction of by RFG. Extensive source apportionment analyses have been performed with data from Houston; these analyses can be revisited to investigate trends over time. Some emission factor analyses using the tunnel in the Houston Ship Channel have been performed.
- *Other considerations.* The onshore-offshore flow reversals in Houston and the complexity and abundance of industrial emissions are factors that make analysis of data in Houston particularly challenging.

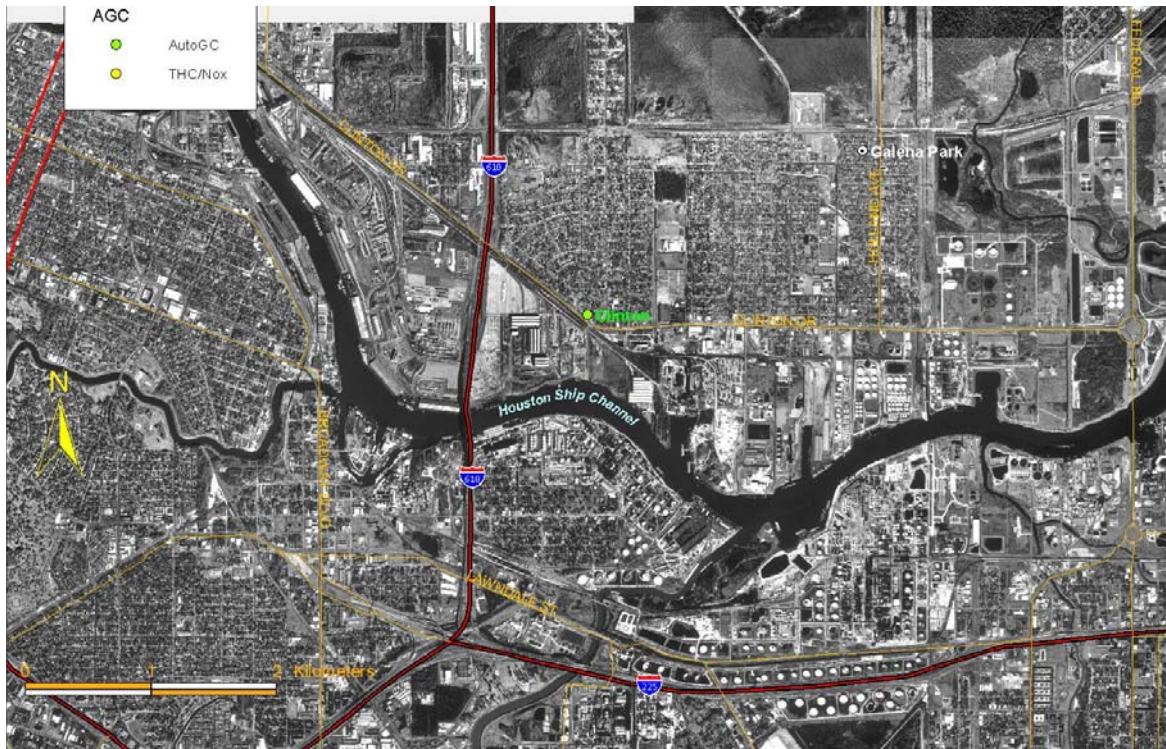


Figure 4-1. Clinton Drive site near the Houston Ship Channel (green dot in the center of the photo).

4.4 DIESEL EMISSIONS REDUCTIONS

The objective of this control scenario is to assess the impact of diesel vehicle retrofits to reduce PM emissions in a selected city. A candidate city is Los Angeles, California, but other cities may have cleaned up their bus fleets. Using the thought process described in Sections 2 and 3, the following items will need to be addressed:

- *Air pollutants expected to be affected.* The focus is likely EC, but other pollutants that could be affected include NO_x, SO₂, PM_{2.5} mass, OC, PM sulfate, and PM nitrate. Highly time-resolved data, such as hourly (or sub-hourly) Aethalometer™ BC data, are likely required to isolate the time periods and days of week with the most activity.
- *Spatial scale of pollutants.* The focus of this assessment is on primary EC emissions at an urban scale or smaller.
- *Spatial scale of the control measure.* Diesel particulate matter (DPM) emissions are typically a relatively small portion of the PM_{2.5} mass in most urban areas (using EC as a marker, although not a unique one, for DPM). The effect of a diesel retrofit program is likely measurable only near areas with a high density of retrofitted equipment. An ideal monitoring site would be located near a bus yard or transit center, for example. Local knowledge of program timing and number of buses, etc. is needed.

- *Timing and magnitude of changes.* A summary of the expected emissions reductions needs to be prepared.
- *Magnitude of expected ambient changes.* Based on the expected emissions reductions, an estimate of expected ambient changes needs to be made. Are model results available for these types of scenarios?
- *Statistical metrics to consider.* Summary statistics of concentrations, ratios of EC-to-PM components, and the fraction of EC in the PM_{2.5} will be needed. If dual channel Aethalometer™ data are available, an investigation of the channels may provide additional information about DPM. The signal produced by the near-ultraviolet (UV) channel (BC₃₇₀) may be useful for detecting UV-absorbing organic compounds. Jeong et al. (2004) reported an increase in BC₃₇₀ relative to BC₈₈₀ when a Pennsylvania site was impacted by smoke from a large forest fire in Quebec (Jeong et al., 2004). It has also been suggested that BC₃₇₀ is an indicator of fresh diesel emissions (Hansen, 1998). Meteorological adjustments are not likely very important for primary pollutants measured so close to a source.
- *Basic analyses.* Section 3.1 analyses need to be performed (or previous analysis results reviewed). Assessment of weekday-weekend differences in PM and its components is of particular interest when investigating DPM from mobile sources.
- *Corroborative analyses.* Source apportionment may be useful in this analysis. Maps and spatial distribution of sources of DPM are key supporting information.
- *Other considerations.* The relative importance of bus emissions to the total PM (or EC) mass and the influence from other diesel emissions (shipping, construction equipment, rail) need to be considered.

4.5 AIR TOXICS REDUCTIONS

The objective of this control scenario is to assess the reduction of air toxics as a result of the introduction of MACT regulations. A candidate city is Los Angeles, California, but other cities may have applied a range of MACT regulations focusing on specific pollutants or source types. Using the thought process described in Sections 2 and 3, the following items will need to be addressed:

- *Air pollutants expected to be affected.* The air toxics affected by control programs are MACT-specific. As an example, consider tetrachloroethylene (i.e., perchloroethylene, or perc) reductions in Los Angeles. Perc is primarily emitted by dry cleaners.
- *Spatial scale of pollutants.* The focus of this assessment is on primary emissions at an urban scale or smaller.
- *Spatial scale of the control measure.* The spatial scale of the control is also MACT-specific. This control will depend on the size and number of facilities affected and their proximity to the monitors. As with other examples, local knowledge of program timing and affected facilities is needed.

- *Timing and magnitude of changes.* A summary of the expected emissions reductions needs to be prepared.
- *Magnitude of expected ambient changes.* Based on the expected emissions reductions, an estimate of expected ambient changes needs to be made.
- *Statistical metrics to consider.* Summary statistics of concentrations and possibly ratios of the affected pollutant to other air toxics will be needed.
- *Basic analyses.* Section 3.1 analyses need to be performed (or previous analysis results reviewed).
- *Corroborative analyses.* Emissions estimates, promulgation and compliance dates, and maps and spatial distribution of pollutant sources are key supporting information. **Figure 4-2** shows an example from the Los Angeles basin for perc emissions reductions from dry cleaners. The figure shows MACT implementation and compliance dates, National Emission Inventory (NEI) emissions data for dry cleaners in Los Angeles County, and annual average concentrations of perc at three sites in the Los Angeles air basin. Note that two of the sites indicate a clear decline that seems to correspond to the MACT, but one site does not.

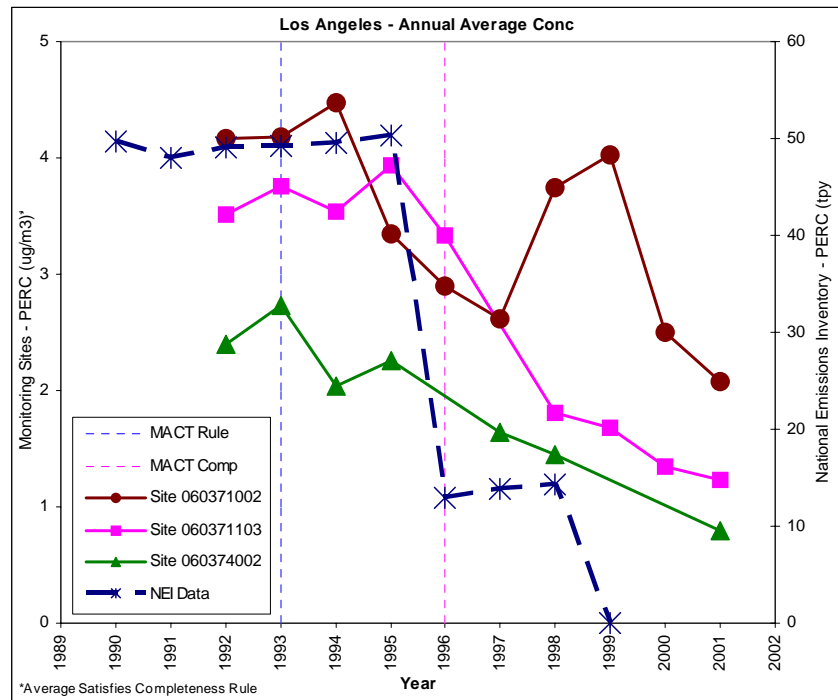


Figure 4-2. Annual average perc concentrations at three sites in Los Angeles, California, and the NEI estimates of perc emissions.

- *Other considerations.* One consideration is to understand how the air toxic pollutant was reduced—through controls or through substitution of a different chemical. If monitoring is performed for that new chemical, the analyst could look for increases. For area source

emissions, more may need to be known about where the emissions sources are actually located rather than relying on typical surrogates, such as population density or land use.

4.6 SUMMARY

The examples in this section are meant to provide indications of the thought process necessary to select examples for investigation. Other cities and controls of interest are available and a similar approach can be taken to aid in the selection process. Once the selection of the control measures and cities for investigation is complete, the technical approach discussed in this report will be applied to the case studies.

5. REFERENCES

- Ashbaugh L.L., Malm W.C., and Sader W.Z. (1985) A residence time probability analysis of sulfur concentrations at Grand Canyon National Park. *Atmos. Environ.* **19** (8), 1263-1270.
- Brown S.G., Frankel A., Raffuse S.M., Hafner H.R., and Anderson D.J. (2004) Source apportionment of PM_{2.5} in Phoenix, Arizona, using positive matrix factorization. *J. Air & Waste Manage. Assoc.* (submitted for publication).
- Brown S.G., Frankel A., and Hafner H.R. (2005) Source apportionment of VOCs in the Los Angeles area using positive matrix factorization. (submitted) (STI-2725).
- Cox W.M. and Chu S.H. (1993) Meteorologically adjusted ozone trends in urban areas: a probabilistic approach. *Atmos. Environ.* **27B**, 425-434.
- Cox W.M. and Chu S.H. (1996) Assessment of interannual ozone variation in urban areas from a climatological perspective. *Atmos. Environ.* **30**, 2615-2625.
- Draxler R.R. and Hess G.D. (1997) Description of the HYSPLIT 4 modeling system. Technical memorandum by NOAA, ERL ARL-224, December 24.
- Flaum J.B., Rao S.T., and Zurbenko I.G. (1996) Moderating the influence of meteorological conditions on ambient ozone concentrations. *J. Air & Waste Manag. Assoc.* **46**, 35-46.
- Frank N.H. (2005) Retained nitrate, hydrated sulfates, and carbonaceous mass in federal reference method PM_{2.5} for six eastern U.S. cities. *J. Air Waste Manag. Assoc.* (accepted).
- Grego E., Porter P.S., Gilliland A., and Rao S.T. (2006) An examination of the impact of nitrogen oxides emission reductions on ozone air quality over the eastern United States. *J. Appl. Meteorol.* (submitted).
- Hafner H.R. (2003) Data validation workshop. Prepared for the Lake Michigan Air Directors Consortium and U.S. Environmental Protection Agency, Region V, Chicago, IL, by Sonoma Technology, Inc., Petaluma, CA, STI-903510-2424, October.
- Hafner H.R., McCarthy M.C., Brown S.G., Raffuse S.M., Shields E., Stiefer P.S., and Wheeler N.J. (2004) Workshop on air toxics data analysis. *Prepared for Lake Michigan Air Directors Consortium; presented in Rosemont, IL, June 2-3* (STI-903555).
- Hafner H.R., Brown S.G., McCarthy M.C., Frankel A.P., Miller D.S., Raffuse S.M., and Roberts P.T. (2006) Midwest Urban Organics Study. Presented at the *Lake Michigan Air Directors Consortium Data Analysis Workgroup Meeting, Des Plaines, IL, January 26-27* by Sonoma Technology, Inc., Petaluma, CA.
- Hansen A.D.A. (1998) The AethalometerTM. Instrument guide prepared by the Magee Scientific Company, Berkeley, CA.

- Harley R.A., Hannigan M.P., and Cass G.R. (1992) Respeciation of organic gas emissions and the detection of excess unburned gasoline in the atmosphere. *Environ. Sci. Technol.* **26**, 2395-2408.
- Jeong C.H., Hopke P.K., Kim E., and Lee D.W. (2004) The comparison between thermal-optical transmittance elemental carbon and Aethalometer black carbon measured at multiple monitoring sites. *Atmos. Environ.* **38** (31), 5193-5204.
- Kenski D. (2006) Lake Michigan Air Directors Air Consortium, Des Plaines, IL. Personal communication to Hilary Hafner. Personal communication, February.
- Kim E., Hopke P.K., and Edgerton E.S. (2003) Source identification of Atlanta aerosol by positive matrix factorization. *J. Air & Waste Manage. Assoc.* **53**, 731-739.
- Kim E. and Hopke P.K. (2004) Improving source identification of fine particles in a rural northeastern U.S. area utilizing temperature-resolved carbon fractions. *J. Geophys. Res.* **109** (D9).
- Kim E., Hopke P.K., and Edgerton E.S. (2004) Improving source identification of Atlanta aerosol using temperature resolved carbon fractions in positive matrix factorization. *Atmos. Environ.* **38**, 3349-3362.
- Larsen L. (1999) A semi-empirical model relating meteorology and ozone in the San Francisco Bay Area. *Air Pollution VII*, WIT Press, Southampton, UK.
- Main H.H., Roberts P.T., and Reiss R. (1998) Analysis of photochemical assessment monitoring station (PAMS) data to evaluate a reformulated gasoline (RFG) effect. Report prepared for the U.S. Environmental Protection Agency, Office of Mobile Sources, Fuels and Energy Division, Washington, DC, by Sonoma Technology, Inc., Santa Rosa, CA, STI-997350-1774-FR2, April. Available on the Internet at http://www.epa.gov/oar/oaqps/pams/rfg_oms.pdf.
- Main H.H. and Roberts P.T. (2000) PAMS data analysis workbook: illustrating the use of PAMS data to support ozone control programs. Prepared for the U.S. Environmental Protection Agency, Research Triangle Park, NC, by Sonoma Technology, Inc., Petaluma, CA, STI-900243-1987-FWB, September.
- Main H.H. and O'Brien T. (2001) Statistical summary of PAMS data collected in the Northeast and Mid-Atlantic states. Report prepared for NESCAUM, Boston, MA, by Sonoma Technology, Inc., Petaluma, CA, STI-900860-2045-FR2, June 2001; updated summaries added as part of STI-902071-2257-TM, September 2002.
- Main H.H. and Roberts P.T. (2001) PM_{2.5} data analysis workbook. Draft workbook prepared for the U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, Research Triangle Park, NC, by Sonoma Technology, Inc., Petaluma, CA, STI-900242-1988-DWB, February.

- Milanchus M.L., Rao S.T., and Zurbenko I.G. (1997) Discerning changes in anthropogenic ozone in the presence of meteorological fluctuations. *J. Environ. Econ. Manage.*, 17-21.
- National Research Council (1991) *Rethinking the Ozone Problem in Urban and Regional Air Pollution*, National Academies Press, Washington, DC.
- Paatero P. and Tapper U. (1994) Positive matrix factorization: a non-negative factor model with optimal utilization of error estimates of data values. *Environmetrics* **5**, 111-126.
- Paatero P. (1997) Least squares formulation of robust non-negative factor analysis. *Chemometrics and Intelligent Laboratory Systems* **37**, 23-35.
- Porter P.S., Rao S.T., Zurbenko I.G., Dunker A.M., and Wolff G.T. (2001) Ozone air quality over North America: part II—an analysis of trend detection and attribution techniques. *Atmos. Environ.* **51**, 283-306.
- Rao S.T., Zalewsky E., and Zurbenko I.G. (1995) Determining spatial and temporal variations in ozone air quality. *J. Air & Waste Manag. Assoc.* **45**, 57.
- Thompson M.L., Reynolds J., Lawrence H.C., Guttorp P., and Sampson P.D. (2001) A review of statistical methods for the meteorological adjustment of tropospheric ozone. *Atmos. Environ.* **35**, 617-630.
- U.S. Environmental Protection Agency (2005) Evaluating ozone control programs in the eastern United States: focus on the NO_x budget trading program, 2004. Report prepared by the U.S. Environmental Protection Agency, Office of Air and Radiation, Office of Air Quality Planning and Standards, Office of Atmospheric Programs, Washington, DC, August. Available on the Internet at <<http://www.epa.gov/airtrends/2005/ozonenbp.pdf>> last accessed March 9, 2006.
- Watson J.G., Robinson J.F., Chow J.C., Henry R.C., Kim B.M., Pace T.G., Meyer E.L., and Nguyen Q. (1990) The USEPA/DRI chemical mass balance receptor model, CMB 7.0. *Environ. Soft.* **5**, 38-49.
- White W. (2005) IMPROVE network at the Crocker Nuclear Laboratory at U.C. Davis, Davis, CA. Telephone conversation with Steven Brown, Sonoma Technology, Inc., Petaluma, CA. November 22.