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Potential Air Toxics Hot Spots in Truck Terminals and Cabs

Thomas J. Smith, Mary E. Davis, Jaime E. Hart,
Andrew Blicharz, Francine Laden, and Eric Garshick



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with a Critique by the HEI Health Review Committee

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ABOUT HEI

The Health Effects Institute is a nonprofit corporation chartered in 1980 as an independent research organization to provide high-quality, impartial, and relevant science on the effects of air pollution on health. To accomplish its mission, the institute

- Identifies the highest-priority areas for health effects research;
- Competitively funds and oversees research projects;
- Provides intensive independent review of HEI-supported studies and related research;
- Integrates HEI's research results with those of other institutions into broader evaluations; and
- Communicates the results of HEI's research and analyses to public and private decision makers.

HEI typically receives half of its core funds from the U.S. Environmental Protection Agency and half from the worldwide motor vehicle industry. Frequently, other public and private organizations in the United States and around the world also support major projects or research programs. HEI has funded more than 280 research projects in North America, Europe, Asia, and Latin America, the results of which have informed decisions regarding carbon monoxide, air toxics, nitrogen oxides, diesel exhaust, ozone, particulate matter, and other pollutants. These results have appeared in the peer-reviewed literature and in more than 200 comprehensive reports published by HEI.

HEI's independent Board of Directors consists of leaders in science and policy who are committed to fostering the public–private partnership that is central to the organization. The Health Research Committee solicits input from HEI sponsors and other stakeholders and works with scientific staff to develop a Five-Year Strategic Plan, select research projects for funding, and oversee their conduct. The Health Review Committee, which has no role in selecting or overseeing studies, works with staff to evaluate and interpret the results of funded studies and related research.

All project results and accompanying comments by the Health Review Committee are widely disseminated through HEI's Web site (www.healtheffects.org), printed reports, newsletters and other publications, annual conferences, and presentations to legislative bodies and public agencies.

ABOUT THIS REPORT

Research Report 172, *Potential Air Toxics Hot Spots in Truck Terminals and Cabs*, presents a research project funded by the Health Effects Institute and conducted by Dr. Thomas J. Smith of the Harvard School of Public Health, Boston, Massachusetts, and his colleagues. This report contains three main sections.

The HEI Statement, prepared by staff at HEI, is a brief, nontechnical summary of the study and its findings; it also briefly describes the Health Review Committee's comments on the study.

The Investigators' Report, prepared by Smith and colleagues, describes the scientific background, aims, methods, results, and conclusions of the study.

The Critique is prepared by members of the Health Review Committee with the assistance of HEI staff; it places the study in a broader scientific context, points out its strengths and limitations, and discusses remaining uncertainties and implications of the study's findings for public health and future research.

This report has gone through HEI's rigorous review process. When an HEI-funded study is completed, the investigators submit a draft final report presenting the background and results of the study. This draft report is first examined by outside technical reviewers and a biostatistician. The report and the reviewers' comments are then evaluated by members of the Health Review Committee, an independent panel of distinguished scientists who have no involvement in selecting or overseeing HEI studies. During the review process, the investigators have an opportunity to exchange comments with the Review Committee and, as necessary, to revise their report. The Critique reflects the information provided in the final version of the report.

PREFACE

HEI's Research Program on Air Toxics Hot Spots

INTRODUCTION

Air toxics comprise a large and diverse group of air pollutants that, with sufficient exposure, are known or suspected to cause adverse effects on human health, including cancer, effects on the development of organs and tissues, and damage to the respiratory, immune, neurologic, and reproductive systems. These compounds are emitted by a variety of indoor and outdoor sources, and large numbers of people are exposed to them. Therefore, the compounds are a cause for public health concern, even though the ambient levels are generally low. The low ambient levels are one reason that tools and techniques for assessing specific health effects of air toxics are very limited.

Air toxics are not regulated by the U.S. Environmental Protection Agency (EPA) under the National Ambient Air Quality Standards. However, the EPA is required under the Clean Air Act and its amendments to characterize, prioritize, and address the effects of air toxics on public health and the environment, and it has the statutory authority to control and reduce the release of air toxics. The EPA is also required to regulate or consider regulating air toxics derived, at least in part, from motor vehicles (referred to as mobile-source air toxics [MSATs]) by setting standards for fuels, vehicle emissions, or both. In 2001 the EPA designated 21 high-priority MSATs that needed to be reduced (U.S. EPA 2001a). However, the EPA did not take any specific regulatory action at that time because rules mandating the reduction of sulfur in both gasoline and diesel fuels as a way to decrease particulate matter (PM) in emissions were expected to result in the reduction of several MSATs as well (U.S. EPA 2000, 2001b). Subsequently, the EPA identified eight MSATs that, based on their emissions and reported

toxicity, pose the greatest risk to health — benzene, 1,3-butadiene, formaldehyde, acrolein, naphthalene, polycyclic organic matter, diesel PM, and diesel exhaust organic gases — and mandated the reduction of benzene in gasoline and of hydrocarbons (including MSATs) in exhaust (U.S. EPA 2007). In 2007, HEI published a critical review of the literature on exposure to and health effects associated with these highest-priority MSATs (HEI Air Toxics Review Panel 2007).

In trying to understand the potential health effects of exposure to toxic compounds, scientists often turn first to evaluating responses in highly exposed populations, such as occupationally exposed workers. However, workers and their on-the-job exposures are not representative of the general population, and therefore such studies may be somewhat limited in value.

Another strategy is to study populations living in "hot spots" — areas that have high concentrations of these pollutants owing to their proximity to one or more sources. Some hot spots may have sufficiently high pollutant concentrations to make them suitable locations for studies to determine whether there is a link between exposure to air toxics and an adverse health outcome. Such areas offer the potential to conduct health investigations in groups that are more representative of the general population. Before health effects studies can be initiated, however, actual exposures to pollutants — including their spatial and temporal distributions — in such hot-spot areas must be characterized.

DESCRIPTION OF THE PROGRAM

In January 2003, HEI issued a Request for Applications (RFA 03-1) entitled "Assessing Exposure to Air

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Toxics,” seeking studies aimed at identifying and characterizing exposure to air toxics from a variety of sources in areas or situations where concentrations were expected to be elevated. The rationale for the RFA was that understanding exposures in hot spots, as well as the sources of these exposures, would improve our ability to select the most appropriate sites, populations, and endpoints for subsequent health studies. HEI was particularly interested in studies that focused on the high-priority MSATs.

Five studies were funded under this RFA to represent a diversity of possible hot-spot locations and air toxics. The study by Smith and colleagues described in this report (Research Report 172) is the last of the five to be published. The five studies are summarized below.

“Air Toxics Hot Spots in Industrial Parks and Traffic,” Thomas J. Smith, Harvard School of Public Health, Boston, Massachusetts (Principal Investigator)

In the study presented in this report, Smith and colleagues measured levels of air toxics and PM at upwind and downwind locations around the perimeter of 15 truck terminals across the United States and in cabs of pickup and delivery trucks during a work shift. The HEI study was added to an ongoing study, funded by the National Cancer Institute, of the relationship between exposure to diesel exhaust and mortality from lung cancer among dockworkers and truck drivers at more than 200 truck terminals in the United States. The degree of variation at different locations and the influence of wind direction were also evaluated with the goal of identifying the potential impact of truck terminals on the surrounding areas.

“Measurement and Modeling of Exposure to Air Toxics and Verification by Biomarkers,” Roy M. Harrison, University of Birmingham, Birmingham, United Kingdom (Principal Investigator)

In the study described in HEI Research Report 143 (2009), Harrison and colleagues investigated personal exposure to a broad range of air toxics, with the goal of developing detailed personal-exposure models that would take various microenvironments into account. Repeated measurements of exposure to selected air

toxics were made for each of 100 healthy nonsmoking adults who resided in urban, suburban, or rural areas of the United Kingdom, among which exposures to traffic were expected to differ; repeated urine samples were also collected for analysis. Harrison and colleagues developed models to predict personal exposure on the basis of microenvironmental concentrations and data from time–activity diaries; they then compared measured personal exposure with modeled estimates of exposure.

“Assessing Exposure to Air Toxics,” Eric M. Fujita, Desert Research Institute, Reno, Nevada (Principal Investigator)

In the study presented in HEI Research Report 156 (2011), Fujita and colleagues measured the concentrations of PM and MSATs on major California freeways and compared them with corresponding measurements obtained at fixed monitoring stations. The diurnal and seasonal variations in concentrations of selected pollutants and the contribution of diesel-and gasoline-powered vehicles to selected air toxics and elemental carbon were also determined.

“Air Toxics Exposure from Vehicular Emissions at a U.S. Border Crossing,” John Spengler, Harvard School of Public Health, Boston, Massachusetts (Principal Investigator)

The study by Spengler and colleagues, presented in HEI Research Report 158 (2011), assessed concentrations of MSATs surrounding the plaza adjacent to the Peace Bridge, a major border crossing between the United States and Canada, located in Buffalo, New York. Three fixed monitoring sites were used to compare pollutant concentrations upwind and downwind of the plaza. Meteorologic measurements and hourly counts of trucks and cars crossing the bridge were used to examine the relationship between the concentrations of air toxics and traffic density. To study spatial distributions of pollutants, members of the investigative team used portable instruments and a Global Positioning System device to obtain location-specific, timestamped measurements as they walked along four routes in a residential neighborhood near the plaza.

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“Assessing Personal Exposure to Air Toxics in Camden, New Jersey,” Paul J. Lioy, Environmental and Occupational Health Sciences Institute, Piscataway, New Jersey (Principal Investigator)

In the study presented in HEI Research Report 160 (2011), Lioy and colleagues measured ambient and personal exposure concentrations of air toxics and fine PM for 107 nonsmoking participants in two neighborhoods of Camden, New Jersey. One, considered to be a hot spot, had a high density of industrial facilities serviced by truck traffic and nearby busy roads. The other, with no industrial sources but near several highways, was considered an urban reference site. The investigators collected four sets of 24-hour personal air samples for the study subjects and made simultaneous measurements of ambient pollutant concentrations at a fixed monitoring site in each neighborhood. To characterize finer spatial variability in pollutant levels, air toxics levels were also measured at multiple sampling sites in each neighborhood during three sampling periods. The investigators used modeling to estimate the contribution of ambient sources to personal exposure.

HEI is committed to continuing research on air toxics — for example, as part of studies to assess the health outcomes of air quality actions or studies to evaluate the effects of new technologies and fuels. Further information on these programs can be obtained at the HEI Web site (www.healtheffects.org).

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HEI STATEMENT

Synopsis of Research Report 172

Potential Air Toxics Hot Spots in Truck Terminals and Cabs

BACKGROUND

Motor vehicles and other combustion sources emit many air toxics whose ambient concentrations are not regulated by the U.S. Environmental Protection Agency (EPA) but that are known or suspected, with sufficient exposure, to cause adverse human health effects. Among these are mobile source air toxics (MSATs), compounds that the EPA has identified as being derived, at least in part, from motor vehicles and whose emissions need to be reduced. Although ambient concentrations of air toxics are generally low, so-called hot spots might exist where concentrations of one or more air toxics, and consequent exposures of area populations, could be elevated. Such areas may be in proximity to one or more pollution sources or may be affected by transient or sustained localized conditions that lead to elevated concentrations of some pollutants. In 2003, HEI targeted research to identify and characterize potential air toxics hot spots.

APPROACH

Dr. Thomas Smith of the Harvard School of Public Health and his colleagues measured concentrations of selected volatile organic compounds (VOCs) and particular matter (PM) in locations with potentially high levels of air pollution that could make them hot spots for human exposure, that is, around the perimeter of terminals for pick-up and delivery trucks and in truck cabs during daily runs. The premise underlying the selection of the sampling sites was that locations upwind of the terminals would have lower concentrations than downwind locations. The investigators hypothesized that the upwind locations' concentrations would be influenced by "industrial parks and other commercial zones" while the downwind locations' concentrations would reflect the added contribution from truck traffic inside the terminal and could

be representative of exposures in nearby downwind residential neighborhoods.

The investigators had access to the terminals as part of a then-ongoing study funded by the National Cancer Institute (NCI) that involved truck drivers, loading-dock workers, and mechanics at 36 truck terminals chosen randomly in major metropolitan areas across the United States. At the time of the authors' application to HEI, 15 of these terminals had not yet been visited for exposure assessment. For these 15 terminals, concurrent measurements of air toxics were added. This phase is referred as Phase 1. During Phase 2, Dr. Smith and colleagues went back to six of the 15 terminals to make additional measurements.

Dr. Smith and colleagues measured VOCs (hydrocarbons and carbonyls) and PM with an aerodynamic diameter $\leq 2.5 \mu\text{m}$ ($\text{PM}_{2.5}$) at the following locations:

- At the upwind fence line (also referred to as the "terminal background") and downwind fence line of the terminal perimeter. Sampling entailed consecutive 12-hour integrated sampling periods for five days in a row at each terminal. In Phase 2, sampling was repeated at six terminals, and continuous sampling for total VOCs and $\text{PM}_{2.5}$ was added at each of the four primary wind directions to allow more flexibility in classifying upwind or downwind locations during sampling. Downwind contributions were expressed as ratios of the mean downwind and upwind concentrations for various pollutants by terminal.
- In the docks and repair shops (at the six repeat-visit terminals). Sampling in these two indoor locations were added in Phase 2 of the study.
- In truck cabs during 8-hour daily pick-up and delivery trips (for a total of 36 trips). Continuous sampling for total VOCs and $\text{PM}_{2.5}$ was

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added in the cabs of two trucks equipped with a global-positioning-system unit to allow correlation of the exposure measurements with the route characteristics.

The compounds measured with the integrated monitors are listed below. The compounds in italics were those targeted in the original Request for Applications.

- Hydrocarbons: *1,3-butadiene*, aromatic compounds, (*benzene*, *toluene*, *xylanes*, *ethylbenzene*, and *styrene*), alkane compounds (*n-hexane*, trimethylpentane, dimethylpentane, 2-methylhexane, methylpentane, 3-methylhexane, and methylcyclohexane);
- *Methyl tert-butyl ether (MTBE)*; and
- Carbonyls: aldehydes (*formaldehyde* and *acet-aldehyde*) and acetone.

PM_{2.5} was characterized as part of the NCI study for mass by gravimetric analysis. In Phase 2 of their study, the investigators made continuous mass measurements using a PM_{2.5} aerosol monitor.

Structural equation modeling was used to identify the indirect effects of intermediate variables (including temperature, wind speed, distance of the terminal to a major road, and regional census variables) on primary dependent variables, which were the fence-line upwind concentrations of 1,3-butadiene, benzene, toluene, and formaldehyde.

RESULTS AND INTERPRETATION

The results of the sampling at terminals' fence lines indicated that overall there was little or no difference between the concentrations at the upwind and downwind sites. Concentrations at terminal upwind locations were generally lower than those at indoor locations.

Analyses of the downwind-upwind pollutant ratios showed wider ranges for VOCs than for aldehydes and PM_{2.5}. The investigators acknowledged that wind directions were not constant during the 12-hour sampling periods and that this probably contributed to reducing the differences between the upwind and downwind locations. The analyses of continuous total VOC measurements made in Phase 2 provided a more detailed pattern of concentration variations in relation to changes in wind directions. Here, unlike the results in Phase 1, analyses

combining data from all six terminals showed significant upwind-to-downwind differences for about 60% of the sessions. Although these data were not fully analyzed and were limited to total VOCs, they pointed to the importance of wind direction in determining the impact of pollutant sources.

Higher temperatures were associated with higher concentrations of formaldehyde and lower concentrations of 1,3-butadiene. Wind speed was inversely correlated with the concentrations of all four pollutants. Distance to an interstate highway was significantly and inversely associated only with toluene and benzene. An analysis by U.S. census regions (i.e., Midwest, Northeast, and West) showed much variability across the regions, with higher concentrations of benzene in the West and of formaldehyde in the Northeast; the reason for this regional pattern is not clear.

Analyses of the in-cab measurements showed that the concentrations of benzene, MTBE, styrene, and hexane measured in the cabs of the nonsmoking drivers were higher on average than those measured at the upwind locations and indoor work locations. Analysis of the effects of open or closed windows on in-cab concentrations showed that when the windows were "predicted to be open" there were significantly lower concentrations of aldehydes and higher concentrations of PM_{2.5} and 1,3-butadiene. The authors suggest that some of the pollutants (such as aldehydes) are generated within the truck's own cab, and that others originate from the surrounding traffic.

Hot Spot Determination

The authors used different criteria to determine whether the terminals were hot spots in different sections of the report, and their conclusions depended on the comparison being made. They compared the concentrations found in their study with those measured by the EPA air toxics monitoring network (which included urban, suburban, industrial, and rural locations throughout the United States), those reported in various exposure studies conducted in urban areas and inner-city neighborhoods in the United States, and the EPA's screening values for noncancer and cancer risk. The authors reported that the means and medians of upwind concentrations of the VOCs that were also measured at the EPA air toxics monitoring sites

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were very similar to the mean concentrations measured at the EPA sites. The measured concentrations of 1,3-butadiene and aldehyde concentrations were comparable to those measured in exposure studies in urban areas, and all the aromatics (such as benzene, toluene, and the xylenes) were lower in the current study.

In their comparison with the EPA screening values, the investigators found that 100%, 93%, 61%, and 6% of the upwind mean concentrations of formaldehyde, acetaldehyde, 1,3-butadiene, and benzene, respectively, exceeded the screening values for cancer risk. These values were calculated by the EPA using the cancer unit risk value as a starting point, with various corrections that resulted in more conservative (i.e., health protective) values.

Finally, as planned, the investigators compared the fence-line upwind measurements with the downwind measurements, primarily using time-integrated measures, and found that they were similar.

The HEI Review Committee, which conducted an independent review of the study, noted that a limitation of the study as a hot spot study was the lack of parallel measurements at suitable background sites (i.e., sites at an appropriate distance from the terminals and not impacted by local sources) and of discussion of the local context of the terminals (such as the quality and quantity of the sources around and within the terminals).

CONCLUSIONS

The Review Committee thought that a major strength of the study was to document concentrations of air toxics in various environments in and around truck terminals and inside truck cabs. The Committee noted that the terminals were not selected to meet the initial hypothesis of there being industrial areas upwind of the terminals and neighborhoods downwind. In addition, the upwind location was defined operationally as being upwind

with respect to the prevailing wind direction, whereas in fact wind direction proved to be variable over the course of a day and from day to day.

The investigators made several comparisons for hot spot determination and concluded that the terminals were hot spots when compared with EPA screening values. This comparison is problematic, however, because the screening values are often exceeded in many urban areas, as can be observed by comparing them with the concentrations measured at the EPA air toxics monitoring sites. Comparisons with measures from other studies did not support defining the terminals as hot spots; comparison of upwind and downwind measurements also showed little or no difference because of shifting wind patterns. Measurements at appropriately selected background sites would be needed to establish exactly how "hot" the terminal fence-line locations were at any given time. Overall, the Committee noted that the study does not provide conclusive evidence as to whether the truck terminals were pollution hot spots, but pointed out the existence and variability of localized elevated pollutant levels that could affect human health. The measurements represent potential exposures of workers who work at the terminals frequently and for prolonged periods of time.

With regard to the in-cab measurements, the continuous and time-weighted-average measurements in the truck cabs did document elevated concentrations of a range of components compared with the fence-line measurements. The Committee thought that these should be considered occupational exposures.

Overall, this study provides useful information on measurements of a series of air toxics at truck terminals. It also illustrates the challenges encountered in defining and documenting air pollution hot spots without accounting for the role of meteorologic conditions or establishing adequate background sites for comparison.

INVESTIGATORS' REPORT

Potential Air Toxics Hot Spots in Truck Terminals and Cabs

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Exposure, Epidemiology, and Risk Program, Department of Environmental Health (T.J.S., M.E.D., A.B.), and Department of Epidemiology (J.E.H., F.L.), Harvard School of Public Health, Boston, Massachusetts; Department of Urban and Environmental Policy and Planning, Tufts University, Medford, Massachusetts (M.E.D.); Channing Laboratory, Department of Medicine, Brigham and Women's Hospital, Harvard Medical School, Boston, Massachusetts (J.E.H., F.L., E.G.); Pulmonary and Critical Care Medicine Section, Medical Service, VA Boston Healthcare System, Boston, Massachusetts (E.G.)

ABSTRACT

INTRODUCTION

Hot spots are areas where concentrations of one or more air toxics — organic vapors or particulate matter (PM) — are expected to be elevated. The U.S. Environmental Protection Agency's (EPA*) screening values for air toxics were used in our definition of hot spots. According to the EPA, a screening value “is used to indicate a concentration of a chemical in the air to which a person could be continually exposed for a lifetime ... and which would be unlikely to result in a deleterious effect (either cancer or noncancer health effects)” (U.S. EPA 2006). Our characterization of volatile organic compounds (VOCs; namely 18 hydrocarbons, methyl *tert*-butyl ether [MTBE], acetone, and aldehydes) was added onto our ongoing National Cancer Institute–funded study of lung cancer and particulate pollutant concentrations (PM with an aerodynamic diameter $\leq 2.5 \mu\text{m}$ [$\text{PM}_{2.5}$]), elemental carbon [EC], and organic carbon [OC]) and source apportionment of the U.S. trucking industry. We focused on three possible hot spots

This Investigators' Report is one part of Health Effects Institute Research Report 172, which also includes a Critique by the Health Review Committee and an HEI Statement about the research project. Correspondence concerning the Investigators' Report may be addressed to Dr. Mary E. Davis, Department of Urban and Environmental Policy and Planning, Tufts University, 97 Talbot Avenue, Medford, MA 02155; mary.davis@tufts.edu.

Although this document was produced with partial funding by the United States Environmental Protection Agency under Assistance Award CR-83234701 to the Health Effects Institute, it has not been subjected to the Agency's peer and administrative review and therefore may not necessarily reflect the views of the Agency, and no official endorsement by it should be inferred. The contents of this document also have not been reviewed by private party institutions, including those that support the Health Effects Institute; therefore, it may not reflect the views or policies of these parties, and no endorsement by them should be inferred.

* A list of abbreviations and other terms appears at the end of the Investigators' Report.

within the trucking terminals: upwind background areas affected by nearby industrial parks; downwind areas affected by upwind and terminal sources; and the loading docks and mechanic shops within terminal as well as the interior of cabs of trucks being driven on city, suburban, and rural streets and on highways.

METHODS

In Phase 1 of our study, 15 truck terminals across the United States were each visited for five consecutive days. During these site visits, sorbent tubes were used to collect 12-hour integrated samples of hydrocarbons and aldehydes from upwind and downwind fence-line locations as well as inside truck cabs. Meteorologic data and extensive site information were collected with each sample. In Phase 2, repeat visits to six terminals were conducted to test the stability of concentrations across time and judge the representativeness of our previous measurements. During the repeat site visits, the sampling procedure was expanded to include real-time sampling for total hydrocarbon (HC) and $\text{PM}_{2.5}$ at the terminal upwind and downwind sites and inside the truck cabs, two additional monitors in the yard for four-quadrant sampling to better characterize the influence of wind, and indoor sampling in the loading dock and mechanic shop work areas.

RESULTS

Mean and median concentrations of VOCs across the sampling locations in and around the truck terminals showed significant variability in the upwind concentrations as well as in the intensity of exposures for drivers, loading-dock workers, and mechanics. The area of highest concentrations varied, although the lowest concentrations were always found in the upwind background samples.

However, the downwind samples, which included the terminal's contribution, were on average only modestly higher than the upwind samples. In the truck terminal, the mechanic-shop-area concentrations were consistently elevated for many of the VOCs (including the xylenes, alkanes, and acetone) and particulates; the loading-dock concentrations had relatively high concentrations of 1,3-butadiene, formaldehyde, and acetaldehyde; and non-smoking driver exposures were elevated for benzene, MTBE, styrene, and hexane. Also, the loading dock and yard background concentrations for EC and PM_{2.5} were highly correlated with many of the VOCs (50% of pairs tested with Spearman $r > 0.5$ and 75% with $r > 0.4$); in the mechanic shop VOCs were correlated with EC but not PM_{2.5} ($r = 0.4\text{--}0.9$ where significant); and for driver exposures VOC correlations with EC and PM_{2.5} were relatively low, with the exception of a few aromatics, primarily benzene ($r = 0.4\text{--}0.5$).

A principal component analysis of background source characteristics across the terminal locations that had repeat site visits identified three different groupings of variables (the "components"). This analysis suggested that a strong primary factor for hydrocarbons (alkanes and aromatics) was the major contributor to VOC variability in the yard upwind measurement. Aldehydes and acetone, which loaded onto the second and third components, were responsible for a smaller contribution to VOC variability. A multi-layer exposure model was constructed using structural equation modeling techniques that significantly predicted the yard upwind concentrations of individual VOCs as a function of wind speed, road proximity, and regional location ($R^2 = 0.5\text{--}0.9$). This predicted value for the yard background concentration was then used to calculate concentrations for the loading dock and mechanic shop. Finally, we conducted a detailed descriptive analysis of the real-time data collected in the yard and in truck cabs during the six repeat site visits, which included more than 50 12-hour sessions at each sampling location. The real-time yard monitoring results suggested that under some conditions there was a clear upwind-to-downwind trend indicating a terminal contribution, which was not apparent in the integrated sampling data alone. They also suggested a nonlinear relationship with wind speed: calm conditions (wind speed < 2 mph) were associated with erratic upwind–downwind differences, lower wind speeds (2 to 10 mph) favored transport with little dilution, and higher wind speeds (> 10 mph) favored dilution and dispersal (more so for VOCs than for PM). Finally, an analysis of the real-time data for driver exposures in trucks with a global positioning system (GPS) matched with geographic

information system (GIS) data suggested a clear influence of traffic and industrial sources along a given route with peaks in driver exposures. These peaks were largely associated with traffic, major intersections, idling at the terminals, and pickup and delivery (P&D) periods. However, VOCs and PM_{2.5} had different exposure patterns: VOCs exposures increased when the vehicle was stopped, and PM_{2.5} exposures increased during travel in traffic.

CONCLUSIONS

All three types of testing sites — upwind and downwind fence-line locations and inside truck cabs while in heavy traffic — met the established definition for a hot spot by having periods with concentrations of pollutants that exceeded the EPA's screening values. Most frequently, the pollutants with concentrations exceeding the screening values were formaldehyde, acetaldehyde, and EC (which serves as a marker for diesel particulate); less frequently they were 1,3-butadiene and benzene. In the case of the downwind location of a single truck terminal without an aggregation of other sources, high concentrations of VOCs and PM were infrequent. Using structural equation modeling, a model was developed that could identify combinations of conditions and factors likely to produce hot spots. Source apportionment analyses showed that EC came predominantly from diesel emissions. As expected from the sites studied, organic vapors associated with vehicle emissions (C₆–C₈ alkanes and aromatics) were the predominant components of VOCs, followed by formaldehyde and acetaldehyde. For driver exposures, high VOC values were associated with stopped vehicles, and high PM_{2.5} values were associated with conditions during driving.

INTRODUCTION

HOT-SPOT STUDY

Our study was an add-on to the Trucking Industry Particle Study, a national study of lung cancer and particulate exposures in the U.S. trucking industry (Smith et al. 2006). For more detail on this study, which was funded by the National Cancer Institute (NCI), see Appendix A. For our study, we defined potential hot spots as areas where pollutant emissions from common sources of concern (in this case, diesel vehicles or heavy traffic) were likely to be present at significantly higher levels than in areas without these pollutant sources. Specifically, because our study was tied to the trucking industry, we proposed three types of hot spots as defined by their location: (1) downwind of

industrial parks or other commercial zones (the upwind edge of truck terminals), (2) downwind of truck terminals, and (3) inside the cab of a truck being driven in urban traffic.

Truck terminals are often located in industrial parks or commercial zones just outside the center of large cities. These areas have high levels of truck traffic not only because of the truck terminal itself, but also because there are other terminals, large retail stores, and distribution warehouses with frequent truck deliveries nearby. Areas near terminals themselves might also be hot spots because of the concentrated truck traffic at the terminals. Driving in heavy urban traffic with substantial truck traffic is also a high exposure setting. Because residential areas are commonly located near truck terminals or commercial areas, although not always, PM_{2.5} and VOC concentrations at the upwind fence line of terminals are intended as the surrogate for residential levels without any contribution from the terminal. Between-city and temporal within-terminal variations would represent the variation in residential neighborhood concentrations for those neighborhoods located in comparable settings. Similarly, samples collected at the downwind terminal fence line represent the conditions where truck traffic in a terminal adds emissions to those of upwind sources and contributes to downwind exposures in nearby residential neighborhoods. Thus our samples were representative of VOC concentrations in neighborhoods close to areas with high truck traffic across the United States. By measuring the upwind and downwind concentrations near truck terminals we could determine the frequency with which commercial areas and truck terminals were hot spots for diesel emissions. No attempt was made to characterize the populations that were actually living in communities near the terminals we sampled. Some terminals were not near (< 1 kilometer) any residential areas. Our measurements represented “worst case” scenarios and thus might set an upper boundary on the distribution of likely community exposures at terminal fence lines. The VOC sampling strategy was matched to the particle measurements being made concurrently as part of the NCI study.

Finally, truck drivers from these terminals were monitored with samplers in the truck cabs to characterize VOC and PM_{2.5} concentrations in the microenvironment of a truck cab, and therefore the personal exposures of the truck drivers, while driving under a wide range of conditions across the United States. The in-cab exposures are anticipated to be similar to the general public's exposures while driving.

Definitions of Hot Spots

When it came to defining hot spots, the issue was: hot relative to what? We chose to use two definitions — one was that the site mean for a location was likely to be higher than the regional background and that the location was away from major sources in the urban metropolitan area. Although no regional background was measured, we did make comparisons to EPA data from nearby monitors when available. The other definition was how often the concentrations at a site exceeded a relevant EPA limit, such as the screening value (U.S. EPA 2006). These definitions reflected different issues. By the first, a hot spot is relative. In highly polluted areas our chosen hot spots might not have differentially higher concentrations; in clean areas trucking operations might produce substantially higher concentrations. For the second definition, we assumed that the EPA screening-value limits represented a meaningful definition of potentially increased risk.

The EPA screening-value approach was developed to define when an exposure situation for a chemical might pose a “potential public health concern” and was only meant to imply that the chemicals exceeded the screening level. As stated by the EPA, “To clarify the actual level of concern posed by any given chemical that fails the screen will necessarily require a more in-depth risk analysis and may even require the collection of additional data” (U.S. EPA 2006). The screening values are based on a combination of risk assessment and the adoption of an agency's limits, such as those of the International Agency for Research on Cancer. The screening values have been developed for a set of acute and chronic health effects. The EPA's goal was to set screening values at levels at which effects are unlikely, using appropriate adjustments for possible exposure to multiple contaminants if there is an animal-to-human extrapolation, which produces a bias against underestimating risks and yet might also overestimate risk. By definition, values that exceed the screening values are a potential concern because they represent situations where there might be increased risk. The actual level of risk, if any, represented by these exposures is highly uncertain, and considerable further investigation will be needed to define the risk. Thus, this is a very conservative approach to defining the hazard.

STUDY OBJECTIVES

Our primary objective was to characterize selected VOC, hydrocarbon, and aldehyde exposures in three settings where we hypothesized there would be potentially high levels of exposure, which could make these settings hot spots for exposure, that is, locations with much higher exposures than general background conditions in

residential areas. The VOCs included in our measurements are listed in Table 1. Although acrolein and naphthalene were of interest to the sponsors of the study, they were not included, because a suitable measurement method was not available for acrolein at the time of the study (discussed in more detail in the Methods section) and the VOC sampling method we were using could not measure naphthalene. A separate system would have been needed, and this was beyond the resources of the project.

The contract for the study began on January 1, 2004, but the initiation of work was delayed by the completion of the formal agreement and the purchasing of parts and equipment to revise our sampling system for VOC collection. Field work was done in two phases.

Phase 1

Visits, each consisting of 5 days of sampling, were made to 15 large terminals (> 100 employees) chosen at random to measure upwind and downwind conditions using two perimeter sampling sites with full-shift, time-weighted-average (TWA) samples (~12 hours). P&D drivers' exposures were measured with a microenvironment sampling box attached to the truck dashboard. Both VOCs and PM were measured. Concurrently with the field work, a data analysis strategy was developed using our existing particulate data. The NCI project finished at the end of this phase.

Interim Data Analysis At HEI's request, a preliminary data analysis was done to assess the findings and identify gaps or problems. This identified a need for repeat visits to previously visited sites to determine the stability of exposure conditions over 1 to 2 years. This analysis also showed the limitations of integrated 12-hour TWA filter samples for evaluating variable wind transport of emissions around the terminals. TWA driver exposures were also very difficult to link to variable conditions during

driving. Real-time measurements of VOCs and PM at terminal sites and in truck cabs were added to solve these problems. The use of GPS tracking devices was implemented to monitor P&D truck movements during real-time sampling. A GIS was used to integrate all of the sampling data with location data, weather data, and EPA data.

Phase 2

Follow-up tests were made at six previously visited terminal sites. At these six terminals, four-quadrant sampling was performed at the terminals' perimeter by direct-reading measurements to capture upwind–downwind differences in real-time measurements of total VOCs and PM_{2.5}. In addition, the effects of temporal variation in on-road emissions, local industries, and vehicle factors were assessed for drivers. The same TWA integrated VOC and PM variables were measured during these visits to enable comparison with the earlier measurements.

SPECIFIC AIMS

The study had five specific aims:

1. To modify our existing sampling system and add integrated VOC collection capabilities for selected hydrocarbons and aldehydes.
2. To measure TWA exposure intensity and variation of VOC components by location characteristics at truck terminal sites across the United States, focusing on three potential hot spots: (a) areas nominally upwind of terminals, (b) areas downwind of terminals, and (c) truck cabs.
3. To examine the relationships between VOC exposures and the concentrations and composition of particulates upwind of trucking activities, downwind of trucking activities, and within vehicles.

Table 1. Volatile Organic Compounds (VOCs) Specified in RFA^a and Found in Diesel Exhaust

Aldehydes, Ketones, and Ethers	Alkanes and Alkenes	Aromatics	Polyaromatics
Formaldehyde	n-Hexane	Benzene	Naphthalenes
Acetaldehyde	1,3-Butadiene	Toluene	
Acrolein		Xylene	
Acetone		Ethylbenzenes	
MTBE		Styrene	

^a See Table 1 of HEI RFA 03-1 (HEI 2003).

4. To determine the variation in VOC composition and exposure intensity associated with a mix of sources in industrial parks, downwind neighborhoods, and in vehicles observed in our source-apportionment measurements.
5. To develop a GIS-based statistical modeling method that could deal with both the spatial and temporal dimensions of the data.

Implicit in Specific Aims 3 and 4 was the development of a data analysis protocol and methods for this complex and diverse data set. The addition of Specific Aim 5 reflected the importance of the need for new or expanded data analysis methods for the study.

STUDY DESIGN

OVERALL SAMPLING STRATEGY

The transport of vehicle emissions by local winds was the defining feature of our downwind hot spots. As is well known, wind is highly variable across time in both direction and speed. Weather systems can rapidly change wind conditions. Diurnal variation is associated with daytime solar heating and nighttime cooling. The wind speed during nighttime hours was frequently insufficient for transport; it was “calm” ($< \sim 2$ mph) or “light and variable.” Under these conditions, emissions accumulate locally near sources. When ground-level winds exceed some minimum, ~ 1 mph (0.5 m/sec), then transport occurs. Thus, we could identify two important sets of conditions: periods of calm (accumulation) and periods of transport. During transport we could measure the concentrations of materials that had moved from upwind sources into the terminals and of materials leaving the terminals that could have moved into local residential neighborhoods. We could only distinguish the terminals’ contributions to downwind areas when there was transport and when the upwind contributions could be subtracted out.

To evaluate in-cab exposures of P&D truck drivers, cab air was sampled during work shifts. Because most large terminals are located in suburban areas near major highways, P&D drivers spend a portion of their time driving on major metropolitan highways and secondary roads. They are primarily exposed to the exhaust from the traffic in front of them, which is a mixture of cars and trucks.

Our VOC sampling strategy was matched to that of the particle (EC, OC, and PM_{2.5}) measurements made in the NCI-funded study (Appendix A). In that study, 36 large terminals (each with more than 100 employees) and one or two smaller terminals located nearby were visited. One

large terminal was visited each month for 5 days of round-the-clock sampling, with 12-hour sessions (approximately 7:00 AM to 7:00 PM or vice versa) to measure air quality at fixed locations in the terminal: the upwind side of the yard’s perimeter, the dock, the repair shop, and the office. Samples representing personal exposures were concurrently collected for dock workers, mechanics, and office workers. Additionally, samples were collected in the truck cabs of P&D drivers, who pick up and deliver freight to local customers within ~ 50 kilometers of their base terminal.

Figure 1 shows a diagram of a hypothetical truck terminal. There is a flow of trucks in and out every day; on the weekend traffic is somewhat lighter. The P&D drivers take trailers loaded with freight out in the morning and return at 4:00–7:00 PM with freight to be shipped. Thus, there are two periods of heavy traffic: in the morning (roughly 6:00–10:00 AM) and in the late afternoon and evening (roughly 4:00–10:00 PM). At all hours there is yard traffic as trailers are moved to and from the dock and tractors go to and from the refueling and inspection areas. When a trailer is loaded, a hostler connects it to a tractor and moves it to the ready line, where a P&D driver will take it out. Our sampling sessions began at approximately 6:00 AM with the placement of samplers in P&D truck cabs, followed by the change-out of the fixed-location samplers at the terminal perimeter.

The HEI’s Request for Applications (HEI 2003) identified a set of VOCs to be measured. These are listed in Table 1. Established methods existed for all of these materials except acrolein (discussed below). All of the hydrocarbons, except for naphthalene, could be measured with our triple sorbent tube. Naphthalene measurements required a separate thermal desorption tube and analysis; because of the substantial added sampling and analysis costs, naphthalene was dropped from our study. Our first task was to add a sorbent collection system to our existing particle collection box (developed by the Harvard research team to meet the special requirements of Phase 1 of the project; referred to as the Harvard field monitor), which had worked very well in the field. Figure 2 shows the inside and front of the Harvard field monitor and identifies its components. The sampling box made sampling simple and easy: all collection media are external and readily exchanged; power is provided by a single, replaceable external battery; three internal pumps are field-calibrated with an external precision rotometer; calibration settings are tamper-proof; the box has fittings to mount on a tripod; and there is an internal monitor for real-time data on temperature and relative humidity.

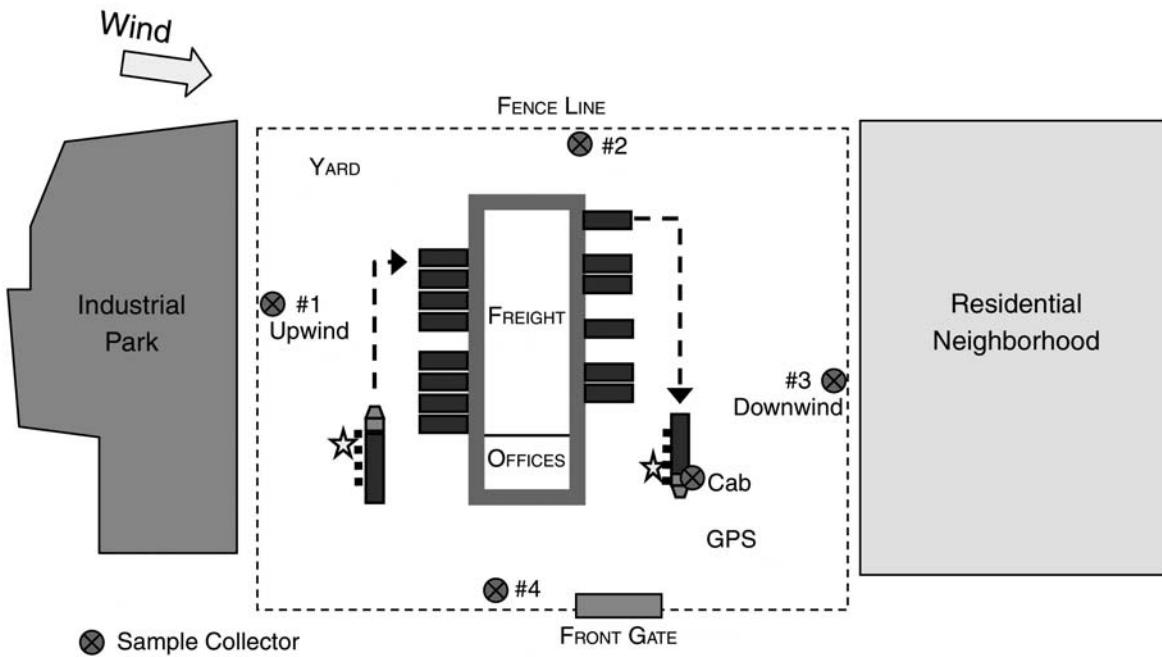


Figure 1. Diagram of hypothetical truck terminal and surrounding area, showing real-time monitors in four quadrants to resolve upwind and downwind pollutant concentrations and contributions.

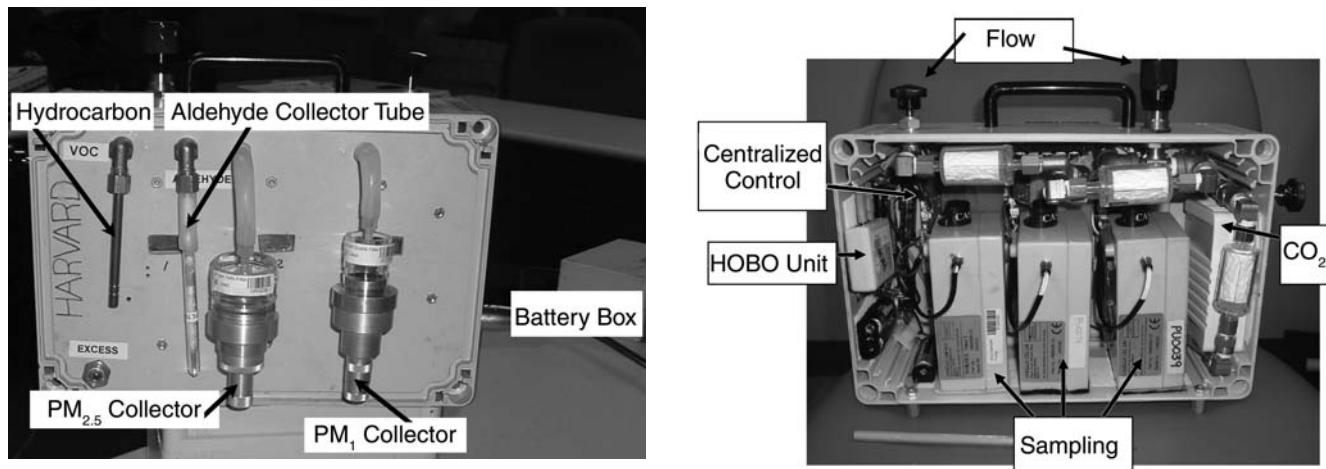


Figure 2. Sampling box used in study. (Left) Front of the sampling box, showing the collection devices for VOCs and particles. (Right) Inside of the sampling box, showing principal components.

PHASE 1 — INTEGRATED SAMPLING STRATEGY

Terminals

The terminals included in this study are listed in Table 2 along with information about their location and workforce. The terminals were highly heterogeneous in terms

of their settings. In some cases, a terminal was located in a suburban town within the metropolitan area of a large city (see Table 2). We developed models for estimating exposures using the more common factors affecting exposures measured in the terminals (Davis et al. 2006). We also collected considerable detail through aerial photos and maps

Table 2. Characterization of the 15 Large Truck Terminals

Sampling Period	Location	Employees (n)	Dock Area (acre)	Dock Doors (n)	P&D Trucks (n)	Proximity to Interstate (m)	ICT Land Use Area (%) ^a	Urban	AIRS Data ^c		
									Metro Area Population ^b	PM ₁₀ (µg/m ³)	NO ₂ (ppm)
Jan. 2004	Elizabeth, NJ (NYC area)	122	1.39	122	54	659	46	Y	21,199,865 (NYC area)	33.00	0.034
Feb. 2004	Oklahoma City, OK	201	2.27	154	23	1166	19	Y	506,132	22.50	0.012
Mar. 2004	Columbus, OH	413	1.79	152	40	1129	23	Y	711,470	25.56	0.016
Apr. 2004	Milwaukee ^d , WI	313	1.86	157	57	2046	26	Y	1,689,572	22.77	0.017
May 2004	Memphis, TN	769	3.40	196	50	2238	30	Y	650,100	23.00	0.022
June 2004	Phoenix, AZ	254	1.96	112	57	3506	13	Y	1,321,045	51.03	0.031
July 2004	Portland, OR	381	1.79	192	45	2115	38	Y	529,121	16.64	0.012
Aug. 2004	Denver ^d , CO	351	1.09	108	57	875	27	Y	2,581,506	35.06	0.020
Sept. 2004	Miami, FL	124	0.74	60	40	7395	92	Y	362,470	20.75	0.010
Oct. 2004	Hagerstown, MD	324	1.42	146	37	765	6	N	36,687	24.00	0.015
Nov. 2004	Nashville ^d , TN	528	1.79	180	34	1158	17	Y	1,231,311	26.00	0.018
Dec. 2004	Middletown, CT	115	0.88	101	40	402	10	N	43,167	14.00	0.010
Jan. 2005	Houston, TX	122	1.68	98	11	59	15	Y	1,953,631	27.70	0.016
Feb. 2005	Laredo, TX	102	1.03	67	35	1327	12	Y	193,117	24.00	0.004
Mar. 2005	Philadelphia ^d , PA	146	0.99	94	49	1827	25	Y	6,188,463	27.00	0.023

^a Percentage of land use within 1-km radius of the terminal designated as industrial, commercial, or transportation (ICT) in the 1992 National Land Cover Data by the U.S. Geological Survey.^b Population records are based upon 2000 census data from the U.S. Census Bureau Web site.^c AIRS values are most recent annual means for county for the nearest EPA monitor.^d Terminal located in a suburban town in the metropolitan area.

for each site; these details were used in the analysis of the effects of large local sources (Garcia et al. 2007). When each terminal was considered in detail, each was unique. For example, one terminal had a landscaping greenhouse nearby. Some had agricultural fields next to them. Several had large yard areas that were partially open fields. Several others were in an industrial park and had one or more adjacent truck terminals or warehouses. Many had commercial buildings near them, but the activities in the buildings during our visit were unknown; often even the type of activity was not clear from the company's name. Thus, although we had highly detailed data on the land uses of the surrounding areas, we could only use most of it as an indication of the potential sources of random variation observed. Our goal was to describe the variability seen in a random sample of sites. Detailed aerial maps of the terminal locations are not included here, to maintain the confidentiality of sampled terminals and trucking companies.

Selection of Terminals

The last 15 terminals to be samples for the NCI study were the terminals used for this study. These sites had been chosen from the major metropolitan areas across the United States with terminals used by the four trucking companies participating in the study. Where there was more than one terminal in an area, one was chosen at random. The order of sampling was randomized.

Selection of Yard Sampler Locations

The procedure for selecting the location of samplers was specified in the field protocol. The prevailing wind pattern at each terminal location was identified just prior to the sampling trip using an online weather source (www.weatherunderground.com) that also gave predictions about major weather changes likely during the 5-day visit. Our weather station was placed downwind of the terminal in an open area near the terminal's fence, where it made real-time measurements of wind speed and direction, temperature, and relative humidity. After a few hours of observations, using a terminal map, upwind and downwind sampling sites were chosen within about 45° of the prevailing wind but away from buildings and activities within the terminal that might unduly influence air movement. These choices could not be made precisely, because the wind direction varied over time and in some cases buildings or trailers were located along the upwind or downwind fence line. The field team did the best it could to find unobstructed locations.

The upwind sampler measured the background contribution from upwind sources, such as local industrial parks, and from regional emissions and atmospheric reaction

products. The downwind sampler measured the downwind concentrations of VOCs and PM leaving the terminal, which included the terminal's contribution plus the background contribution. Upwind–downwind sample pairs were identified during the data-analysis phase by a manual check of the individual wind rose from each 12-hour sampling session. For the last six repeat site visits, when a four-quadrant monitoring system was in place, we identified hourly upwind–downwind pairs both by a manual review of the session's wind direction data and using a spreadsheet combining the real-time wind direction and concentration data.

Time-Weighted Average Sampling During Variable Winds

Variation in wind direction and speed over time was a problem for our time-integrated upwind–downwind sampling. Short-term (~ minutes) variation in wind direction was approximately $\pm 20^\circ$ around the prevailing, or median, wind direction. Movement of weather fronts or thunderstorms in the area could cause substantial, lasting changes in prevailing wind direction. In some locations, winds during the night would frequently be described as "calm" or "light and variable." Calm conditions were defined as wind speeds less than ~ 2 mph. During these conditions, there was no meaningful wind direction or directional transport of emissions, and emissions would build up near the sources, such as in the early morning when the delivery trucks were started. In a few locations that were near rivers or the seacoast, there were diurnal cycles in wind direction caused by differences in land heating and cooling compared with the water.

As a practical matter it was not possible to move the upwind and downwind sampling systems that had the sometimes frequent small changes in wind direction. In order to simplify decision-making, we defined a wind direction that did not vary beyond $\pm 45^\circ$ of the median as stable. This definition was consistent with our focus on what might occur in nearby residential neighborhoods and area locations, without focusing on individual residences. Movements of major air masses are usually associated with fronts, and these movements can produce large changes in prevailing wind direction. These changes are noted in local broadcasts of weather reports, which were used to anticipate major changes in wind conditions. Samplers were moved when major changes were anticipated. Sometimes the shifts in wind direction were gradual, occurring over several hours. In those cases, the field team used its best judgment about when to move the samplers. Overall the samplers were moved relatively few times.

An alternative strategy would have been to change collection media every time the wind conditions changed

significantly. However, this approach was not practical, because it would have produced many samples with short durations and low sample volumes, which would usually have contained insufficient material for quantitation. As noted elsewhere, 12-hour sampling times were needed to collect sufficient material for the expected low ambient concentrations.

Changes in wind conditions (direction or speed) during sampling caused problems with assigning wind direction to integrated samples for the first phase of data analysis. The vector average wind direction and speed were calculated for each 12-hour integrated sample. A quality flag was attached to those samples with significant changes in wind direction ($> 45^\circ$) or long periods of calm winds (> 1 hour). As a result, in some cases the integrated samples did not represent wind transport of emissions from a fixed direction ($\pm 20^\circ$) and commonly included light and variable conditions. This method of assigning wind direction obscured upwind source contributions and diluted downwind contributions from the terminal's activities. However, these wind patterns are normal conditions experienced downwind in a residential area. Our goal was not to track the emissions from specific sources to specific locations, rather it was to obtain representative samples under conditions that prevailed downwind of area sources and typical commercial operations.

Selection of Repair Shop and Dock Sampling Locations

The study protocol specified that the field technician place the sampler centrally on the loading dock or in the repair shop, depending on where the loading activity or shop work was occurring. Placement of dock and shop samplers followed these guidelines: (1) the sampler should be in a location safe from damage from forklift and other vehicle traffic; (2) the sampler should be in a location away from walls and open doors and windows; and (3) when necessary, the sampler should be moved to be close to dock activity because activities sometimes shifted from one part of the dock or shop to another. Although the repair shop and dock are referred to as indoor locations, both work areas had large bay doors and could be considered semi-enclosed indoor work environments. Locations for all of the samplers used at each terminal were recorded.

In-Cab Truck Sampling

The P&D vehicles chosen for in-cab sampling were those used by drivers who volunteered to participate in the study. The dispatcher maintained a list of available vehicles and the drivers assigned to each vehicle. The study team chose which vehicles were to be tested. A driver was then identified and invited to participate in the study.

Informed consent was obtained before proceeding (see Human Subjects Protocol below). The sampling box was mounted on the dashboard of the truck in a location that did not obstruct the driver's view. The sampler measured the microenvironment inside the truck's cab in order to characterize individual drivers' exposures while driving in traffic.

PHASE 2 — REVISED STUDY PLAN — INCREASED LOCATION AND TIME RESOLUTION

Interim Data Analysis

After our first set of field tests at each of the 15 terminals in Phase 1, we conducted an interim data analysis to explore data relationships and preliminary findings and to determine how well we were meeting our objectives. As a result of these analyses, revisions were instituted to make more efficient use of the sampling resources and address the following sampling issues.

Repeat Site Sampling The revised scheme focused on defining within- and between-terminal variability by performing repeat visits during approximately the same season as the initial visit. We made repeat 5-day visits to six large terminals. The terminals selected for these repeat visits were all located in dry climates because our interim data analysis showed that at terminals in wet climates (such as Miami or Houston) a large number of aldehyde samples were lost in the summertime visits when rain or high relative humidity produced condensation in the sampling tubes. There was no way to remove excessive water vapor without also removing some of the polar hydrocarbon vapors.

Upwind and Downwind Sampling Our initial sampling plan had several limitations for clearly defining the effects of wind transport. First, only two fence-line samplers per terminal were used. These were placed in upwind and downwind locations at the start of sampling and were used to measure pollutants coming into the terminal and those leaving it, respectively. Second, wind direction was rarely stable, as noted earlier. Third, 12-hour integrated samples were intended to capture average conditions during employee work shifts, but the fixed time intervals prevented us from distinguishing the effects of shifts in wind direction within the sampling time. Although some variations in wind direction were observed, the siting of samplers was often difficult because buildings and parked trailers were near the fence.

Given the variability of wind direction and its importance for transport, our revised approach for the second

phase of the project was to place four sets of direct-reading instruments at each of the primary wind-direction quadrants and measure total VOCs and PM_{2.5} continuously. This revised approach would permit much closer temporal linkage of the measurements with the wind direction.

Driver Sampling Construction of a driver EC exposure model for the first 36 site visits of the NCI study (the 15 visits of the HEI study began after the first 21 visits [Phase 1]) was limited by a number of factors (Davis et al. 2006). First, we could not feasibly collect detailed data for each driver's route characteristics, such as actual route and number of stops, traffic conditions, and window status, all of which are known to be important predictors of driver exposure to exhaust in truck cabs. This information was necessary to closely match background conditions and in-cab characteristics with rapidly changing driver exposures. We used a variety of methods to predict these missing variables for each driver, including the use of average location characteristics in a radius around a home terminal for background conditions as well in-cab carbon dioxide (CO₂) concentrations to estimate window status. However, these methods are subject to a high degree of exposure misclassification, as evidenced by the low R^2 values obtained during exploratory multiple regression analyses ($R^2 < 0.3$). For this reason, a new strategy was used in Phase 2 of the field study that would gather more detailed and complete data on the drivers and on traffic trends during the study visits.

Revised Sampling Plan

Given the problems with the two fence-line upwind and downwind locations for integrated samplers, we chose to use four-quadrant measurements made with real-time monitors (a photo-ionization detector [PID] monitor [model ppbRAE, RAE Systems, San Jose, CA] and a DustTrak PM_{2.5} aerosol monitor [model 8520, TSI, Shoreview, MN]). We altered the yard-sampling protocol to place samplers at each of the four primary wind directions relative to the terminal center, that is, quadrants of the fence line making ~90° angles to each other, as shown in Figure 1. The "#1" sampling location was always set as the initial upwind location, and the "#3" sampling location was downwind at the beginning of sampling. These locations, set relative to the wind direction on the first day, sometimes changed as the wind shifted during our sampling program, but one of the sampling locations was always within 45° of upwind or downwind. Each of these sites also had direct-reading instruments, a ppbRAE PID monitor for total VOCs, a DustTrak aerosol monitor for PM_{2.5}, and a sampling box that contained integrated collectors.

We placed our weather station at the least obstructed of the four sampling sites. This combination of samplers gave us total hydrocarbon and aldehyde concentrations that were clearly linked to wind direction as well as matched integrated samples that provided more detailed information about composition. It was not always practical to set the secondary axis at right angles to the primary. As noted earlier, upwind and downwind sampling to determine source-category contributions from a site between the two of them was only possible when there were wind speeds sufficient for transport (i.e., speeds > 0.5 m/sec).

To deal with the limited resolution of location and time for the in-traffic measurements, at each terminal we equipped two trucks with a GPS unit and VOC and PM_{2.5} monitors in addition to the standard integrated sampling box. The GPS units provided data for truck position, speed, and direction, which could be overlaid on our GIS terminal-area maps. The time of each GPS measurement could be linked to the time of real-time VOC and PM_{2.5} data points. After each sampling session, drivers were also asked to complete a short questionnaire collecting information about traffic conditions, route, terrain, and smoking status.

Use of Photo-Ionization Detector Monitor

The PID monitor was used to detect short-term variations in total VOC exposure and to link high concentrations with general source categories associated with wind directions at the terminals and with truck activities and locations on roadways. We hypothesized that in roadway locations the emissions sources and composition would be reasonably stable. A principal component analysis was conducted on the TWA composition data to verify that this hypothesis was correct. PID data must be considered semi-quantitative, and their interpretation depends on collateral data collected at the same time as the measurements.

A PID monitor responds to all airborne chemical vapors that can be ionized by ultraviolet (UV) radiation. The monitor's response is calibrated against isobutylene. Unfortunately, all vapors do not have the same tendency to ionize. From the correction factors shown in Table 3 for UV radiation from a 10.6 eV photoionization lamp, there is a range in ionizability by UV radiation. This range would affect the relative response of the monitor for a mixture containing varying amounts of different substances. The monitor's response can be directly interpreted only if the composition of the mixture is known.

The PID monitor has been used as a survey instrument in industrial settings to identify sources of vapors. In settings where the composition of the vapors is known, such as an

Table 3. Correction Factors for ppbRAE Plus 10.6-eV Photo-ionization Lamp^a

Compound	Correction Factor 10.6 eV
Acetaldehyde	6.0
Formaldehyde	0.6
Acetone	1.1
1,3-Butadiene	0.85
Benzene	0.5
Toluene	0.5
Styrene	0.4
Hexane	4.3
Heptane	2.8
Product	
Gasoline	1.0
Diesel fuel	0.4

^a Data obtained from RAE Systems Technical Manual TN-106.

industrial operation using specific solvents, correction factors can be used to adjust the response and estimate concentrations. When the vapors are a complex mixture the relative PID response can vary as the components vary, which makes the concentration estimates approximate. Mixtures that have large numbers of components, such as gasoline, but have no dominating individual components (relative fractions are small) and do not vary by more than $\pm 10\%$ have reasonably stable PID responses per ppm.

Use of DustTrak PM_{2.5} Monitor

Real-time measurements were also made for PM_{2.5}, using the DustTrak aerosol monitor. This monitor measures the amount of 90° light scattering by airborne particles in a laser beam. A cascade-impactor precollector removes particles larger than 2.5 μm before the test chamber. The instrument is factory-calibrated with a standardized Arizona mineral dust (Kim et al. 2004). The amount of light scattering is dependent on particle size and shape distribution, absorption, and specific gravity. The monitor is designed to allow filter collection of particles after they pass through the light-scattering chamber. This characteristic of the monitor can be used to calibrate the average response of the monitor during an 8-hour work shift. It should be noted that the monitor tends to underestimate the mass of very small particles, such as those found in diesel exhaust (Kim et al. 2004).

Human Subjects Protocol

Our protocol was approved by the Institutional Review Board of the Harvard School of Public Health for research on human subjects. Subjects were paid \$25 to compensate them for their time and effort. Informed consent was obtained from all workers whose exposures were monitored. There were no formal inclusion or exclusion criteria, because all current employees of the trucking company working at the terminal were eligible. We preferred nonsmoking subjects, but we took all subjects that were available. A protocol was developed to inform them of what we were doing and why, that they could quit at any time without prejudice, and of any risks (including potential problems arising from their employer learning of their participation). We did not keep the names of individuals who agreed to participate. Nearly 100% of individuals we contacted agreed to participate. None withdrew after previously agreeing to participate. There were no adverse events associated with our human testing. A copy of the consent form used in this protocol is shown in Appendix C, which is available on the HEI Web site.

SAMPLING METHODS

SAMPLER BOX MODIFICATIONS

The first objective of our study was to modify our particle sampling system so it could also collect hydrocarbon and aldehyde vapor samples. We added a third sampling pump to our particle collection box, integrated it into the circuitry, and expanded the external battery to provide sufficient power. This third pump drew a split air stream through two external collector tubes, one for hydrocarbons and one for aldehydes and acetone. The vapor collection system is discussed in more detail in this section, and the modified sampling box is shown in Figure 2.

HYDROCARBON SAMPLING

The vapor sampling system had two legs, a triple sorbent tube for hydrocarbons and a parallel 2,4-dinitrophenylhydrazine (DNPH)-treated sorbent for aldehydes and acetone with an ozone trap. In our initial sampling proposal, the hot-spot areas had been hypothesized to have relatively high concentrations of airborne vapors, especially heavy hydrocarbons, and we therefore expected some risk of sorbent overloading. However, this overloading risk was quickly proven to not be a problem.

The U.S. EPA Compendium Method TO-17, "Determination of Volatile Organic Compounds in Ambient Air Using

Active Sampling onto Sorbent Tubes" (U.S. EPA 1999b), was used for all VOC measurements. Our basic hydrocarbon-sampling tube for this project was a triple sorbent thermal desorption tube with 200 mg Carbopack™ B (Sigma-Aldrich, St. Louis, MO) followed by 230 mg Carbo-pak™ X (Sigma-Aldrich) for 1,3-butadiene and then 170 mg Carboxen® 1001 (Sigma-Aldrich) for intermediate-molecular-weight hydrocarbons passing the first two upstream sorbents sections. This choice was based on data from a technical report from Supelco Inc. (Supelco 2001) that summarized the retention ability of 217 mg Carbo-pak™ B and 290 mg Carbo-pak™ X for a mixture of 42 analytes at 6 "challenge volumes" (0.2, 5, 10, 20, and 100 L) of pure nitrogen flowing at 50–500 mL/min through the sorbent tube after the analyte mixture was added. Using this data, we chose the above sorbents because they allowed us to minimize breakthrough and maximize both the sample volume and the range of analytes, including 1,3-butadiene. A lower flow rate, 10 mL/min, was used to extend the duration of sampling (8–12 hours for a total 6 L collected in 10 hours) to match the particle samples.

The sorbent tubes were conditioned before use by heating to 350°C for 2 hours with a flow of pure helium at 50 mL/min. Used tubes were reconditioned for 50 minutes; later this time was increased to 70 minutes when lab blanks showed some residual peaks after conditioning.

The protocol for field sampling was to place the thermal desorption tube in the sampling position and cap it. With all of the media in place and just before the system was to be placed in the field or in a truck cab, the airflow was started and measured, and the VOC flow was set to 10 mL/min. After sampling, the tube was removed from the system and recapped. Used tubes were kept refrigerated in the field, and they were kept at -20°C when they were returned to the lab. Most tubes were analyzed within 2 weeks of sample collection.

Our quality-control program is summarized in Appendix B. This program included several method-evaluation studies. In these studies we examined repeatability through duplicate samples, breakthrough tests, laboratory spiked samples, and blanks. The studies showed that we could collect hydrocarbons without breakthrough at a site with the highest exposures anticipated (i.e., city traffic during rush hours).

ALDEHYDE AND ACETONE SAMPLING

Initially we attempted to collect aldehydes by derivatization with DNPH using a commercially available sampler (Sep-Pak, Waters Associates, Milford, MA). These samplers were designed to collect ambient formaldehyde and higher-molecular-weight aldehydes using a 100 mL/min

flow rate during 24-hour sampling. The method's limit of detection (LOD) for this active sampler is about 1 ppb formaldehyde. We planned to shorten the sampling time to 8 to 12 hours to match our particle sampling time and to increase the flow rate to collect sufficient material to maintain our LOD. However, we found that our pumps could not produce a sufficient vacuum to draw through the Sep-Pak sampler for an 8-hour sampling period at a rate of ~ 0.5 L/min. A sampling cartridge with a larger grain size designed for occupational exposure sampling at a higher flow rate (Product #226-120, SKC, Eighty Four, PA) was used instead. Because of our concern about breakthrough, we analyzed both the front and back sections of the sampler but found no problems — breakthrough was within acceptable limits (< 10%) (details of these tests are given in Appendix A). After collection, aldehyde samplers were stored at -4°C. Interference from ozone was minimized with a pretreatment section that removed it. This method (Zhang et al. 2000) has been found to be inadequate for acrolein in tests at the Harvard School of Public Health.

A new method for sampling aldehydes was developed by Zhang and colleagues in 2000 using dansylhydrazine for derivatization (Zhang et al. 2000; Herrington et al. 2005). They reported two main improvements over the older DNPH technology. First, the new method was much more effective for sampling acrolein; second, it was thought to be less affected by the concentrations of ozone in the environment. At the beginning of our study, Zhang and colleagues were developing a promising new Personal Aldehydes and Ketones Sampler (PAKS) method that could efficiently measure acrolein, whereas the standard EPA reference method TO-11A could not. Although promising, further development and more testing were needed before the PAKS method would be available for general use. This method was later published by Herrington and colleagues (2005). At the time of our study, Dr. Zhang was testing a modification of the sampler to work for active sampling and to have a lower LOD, but this sampler was not available for our study (J. Zhang, personal communication, 2004).

SAMPLE ANALYSIS

Hydrocarbon Analysis

In the lab, previously used sample tubes and field blanks were placed on a dry-purge device (tubing connected to an ultra-high-purity nitrogen tank with fittings for the tubes) with a carrier flow of 75 mL/min. Tubes were purged for 25 minutes to remove moisture. In accordance with the method mentioned above, the tubes were individually spiked with a vapor-phase internal standard by

injecting the internal standard into the spiking device (ultra-high-purity nitrogen flowing at 10 mL/min into the tube) and keeping the tubes in place for 5 minutes after spiking. The internal standard (M-8260-IS: Internal Standard Mix [1,4-dichlorobenzene, 1,4-difluorobenzene, chlorobenzene, and pentafluorobenzene] 0.2 mg/mL in methanol) was obtained from AccuStandard (New Haven, CT). The vapor-phase internal standard was made from liquid standards injected as a known volume into a 2-L static dilution bottle. A volume of vapor was drawn up with a gas-tight syringe and injected into the injector-port-spiking device. The amount of internal standard used was based on the expected concentration of material in the samples. An automatic thermal desorber (Perkin-Elmer, Waltham, MA) interfaced to a gas chromatograph–mass selective detector (GC–MSD) (Hewlett-Packard, Palo Alto, CA) was used for instrumental analysis. The automatic thermal desorber transfer line connected directly to a capillary column.

Calculation of hydrocarbon air concentrations was done after field blanks were subtracted from measured values, and the values were corrected for recovery efficiency. The thermal desorption tubes gradually developed a 1–2 ng background of benzene, toluene, and xylene as the resin in the tubes broke down from repeated use. To deal with this, at least in part, the reconditioning time was increased from 50 to 70 minutes.

Analysis of adsorbent tubes was performed for the selected VOCs listed in Table 1. The thermal desorption tubes for hydrocarbons were analyzed by way of the automatic thermal desorber (Model 400, Perkin-Elmer). This was directly connected to a gas chromatograph (model HP 5890 II, Hewlett-Packard) with a mass selective detector (model HP 5971, Hewlett-Packard). Samples were analyzed in full scan or selective ion monitoring mode and quantified by an internal-standard quantification method using specialized software (EnviroQuant, Hewlett-Packard) for GC–MSD analysis. The automatic thermal desorber could be loaded with up to 50 tubes at a time. The automatic thermal desorber transfer was connected directly to a capillary column (60 m × 0.25 mm ID × 1.0 µm film thickness, DB-1, J&W Scientific) inside the gas chromatography oven.

Before the tubes were analyzed they were purged of water and an internal standard was added for quantitation. Dry-purge and addition of the internal standard were accomplished in one step. Sample tubes were placed on a spiking device (tubing connected to an ultra-high-purity nitrogen tank with a fitting for the tube) with a carrier flow of 75 mL/min. The vapor-phase internal standard was injected into the device, and the tube was kept in place for

5 minutes. The vapor-phase internal standard was made from a liquid standard in solution (usually methanol) with a known concentration that was injected as a known volume into a 2-L static dilution bottle. A volume of vapor was drawn up with a gas-tight syringe and injected into the injector-port-spiking device with flow into the sample tube. Drawing different volumes yielded different masses of analytes on the tube and thus different levels of calibration.

Aldehyde Analysis

Each sample was prepared for aldehyde analysis by desorbing the DNPH derivative from the substrate with 3 mL acetonitrile. This was done in a vacuum chamber with 12 sample positions, each fitted with a valve to control the extractant flow rate to approximately 1 mL/min. The extract was collected directly into 5-mL volumetric flasks or graduated test tubes. The vacuum was removed, and the samples were diluted to the 5-mL mark with acetonitrile. Aliquots were pipetted into special 1-mL amber vials for chemical analysis.

Samples were analyzed for aldehydes by HPLC with UV detection using a chromatograph (Model 1100, Agilent Technologies, Santa Clara, CA) equipped with a quaternary pumping system, a degassing unit, a 100-position autosampler, a thermostatted column compartment, and a UV-visible variable wavelength detector set at 360 nm. The mobile phase was a mixture of water, acetonitrile, and tetrahydrofuran at a flow rate of 1.0 mL/min. A 20-µL sample was injected into the chromatograph, and the compounds, including the DNPH derivatives, were separated in less than 30 minutes with a gradient on a 150 mm × 4 mm ID reverse-phase HPLC column (Nova-Pak C₁₀, Waters). The data were processed automatically with a software package (Chemstation software, Agilent) and could be reprocessed if required. Detailed individual chromatograms for each sample and a summary report were printed. Data and results were stored on Zip disks. The instrumentation was calibrated with standards.

Calculation of air concentrations was done after field blanks were subtracted from measured values and the values were corrected for recovery efficiency. The amount of the analyte was calculated from a standard curve.

DATA QUALITY

The quality-control methods for the integrated samples used in this study were also used for several other projects, including Dr. Deborah Bennett's study, Boston Exposure Assessment in Microenvironments (BEAM), which was funded by the American Chemistry Council, and Dr. John Spengler's HEI-funded Buffalo Peace Bridge Study. As specified by our protocol, with the aid of synchronized

watches all start and stop times (when the equipment was turned on and off) were recorded to the nearest minute on the data sheet. Our samplers did not have switches and thus could not be tampered with. Instead of switches, a separate control box was used and carried only by the field team. If a pump failed, each sampler had a timer and a back-up battery to create a record of how long the sampler had operated, so that each sampling time was known at the end of the session. Sampling times that were less than half the planned duration were flagged. Samples with values below the instrument blank LOD for the lab method were flagged by the lab. The rate of air flow through each sample medium was measured separately at the start and end of the sampling period with a 12-inch precision rotometer (calibrated against a primary standard before and after each trip). If the flow rate had changed (usually decreased) by more than 10% during the sampling period, then the measurement was flagged. The actual sampling times and measured flow rates were used to calculate the air volume that passed through the media. The specified 12-hour sampling time for the upwind and downwind samples was used to calculate the LOD because the majority of samples were within 10% of 12 hours. For personal-exposure samples, individual work times ranged from 8 to 12 hours.

Hydrocarbons Method

There were several problems with the field-blank method used to estimate the LOD for 1,3-butadiene (Table 4). First, there was very little 1,3-butadiene in the sampling media (0.01–0.02 ng with a standard deviation [SD] of 0.1–0.08 ng); as a result, the estimated LOD based on a 7.2-L 12-hour sample was unreasonably small (0.03 µg/m³) (Table 5). This was a problem for all vapors that were absent or present at very low concentrations in the sampling media, including 1,3-butadiene, MTBE, 2-methylhexane, and methylcyclohexane. To deal with this, in the lab, we spiked blank tubes with small amounts (4 ng) of these compounds. The spiking allowed quantitation of low levels of blank contamination and the variability to estimate the LOD. With this spiking method, the LOD for 1,3-butadiene was 0.86 ng, corresponding to an air concentration of 0.11 µg/m³, which was consistent with the large number of samples from the yard that were below the LOD. Thus, the SD of the repeated spiked measurements gave a more reasonable LOD for 1,3-butadiene, but the error in the spike amount also increased the SD and LOD.

As noted above, both the lab blanks and field blanks increased over time because as the tubes aged the resin in them started to break down and release benzene, toluene,

Table 4. Findings from Eight Laboratory Spiked Samples Taken to Estimate the Blank and LOD for Each Compound

Compound	Mean ^a (ng)	Bias ^b (Mean – 4)	SD	IDL ^c (SD × 3)	LOD ^d (µg/m ³)
1,3-Butadiene	4.46	0.46	0.29	0.86	0.11
MTBE	4.82	0.82	0.17	0.51	0.07
2-Methylpentane	5.43	1.43	0.16	0.48	0.06
3-Methylhexane	5.03	1.03	0.18	0.55	0.07
2-Methylhexane	5.19	1.19	0.17	0.51	0.07
2,3-Dimethylpentane	5.05	1.05	0.15	0.46	0.06
2,2,4-Trimethylpentane	5.37	1.37	0.21	0.62	0.08
Methylcyclohexane	4.90	0.90	0.17	0.51	0.07
Benzene	6.00	2.00	0.18	0.55	0.07
Toluene	5.41	1.41	0.43	1.28	0.16
<i>m&p</i> -Xylenes	7.64	3.64	0.43	1.30	0.16
Ethylbenzene	3.90	– 0.10	0.18	0.55	0.07
Styrene	3.69	– 0.31	0.28	0.84	0.11
<i>o</i> -Xylene	3.64	– 0.36	0.28	0.84	0.10

^a Sampling tubes were spiked with 4 ng of each compound.

^b The bias (contamination) is the difference between the amount spiked and that measured.

^c Instrument detection limit.

^d The LOD is the IDL divided by the volume sampled, typically 7.2 L (ng/L = µg/m³).

and *m*&*p*-xylenes. This release produced a trend in the blank data of artificial increases in the SD and LOD for these compounds (Table 5). This artifact was removed from the sample measurements by subtracting the blanks for each batch. Although the artifact was not a problem for most of the samples, because the concentrations were not close to the LOD, it was a problem for some of the outdoor samples with lower concentrations (such as the blanks measured during Phase 1 of the field work, during which 15 terminals were visited).

In Phase 2 of the field work, which entailed repeat sampling at six terminals, there was a pattern in which a few compounds (e.g., 2,2,4-trimethylpentane, methylcyclohexane, and toluene) had a high mean and high SD values for both lab and field blanks (Table 6). Because the contamination appeared to be in both types of blanks, it was unlikely to be a problem caused by field handling. Except for toluene, which is used as a solvent in some commercial cleaners for trucks, it was unlikely that these compounds were emitted by a single source in the field. In these Phase 2 samples, 1,3-butadiene was also present at the same concentrations in both the field and lab blanks, but the concentrations were much higher than in the blanks used in Phase 1.

Carbonyl Method

An internal-standard mixture was used as specified in the Methods section. The amount of internal standard was based on the expected concentration of material in the samples. The amount of the analyte was calculated from the standard curve adjusted by the ratio of the amount of internal standard added to the amount observed. This method has been used by the Harvard School of Public Health laboratory for more than 10 years to measure a range of aldehydes from formaldehyde to beyond C₁₂ aldehydes, except for acrolein and crotonaldehyde.

High relative humidity (> 90%) was a major problem when collecting aldehydes with the hygroscopic adsorbent sampling tubes. Many samples were lost when they became soaked with condensation during sampling periods with high humidity, such as sampling during rain or fog or during very humid summer weather in the South-eastern United States. Sampling during periods in summer with moderate or low humidity was not a problem. There were also no problems associated with humidity when sampling inside the truck cabs. There was no evidence of an adverse heat effect on aldehyde collection except for increases in relative humidity with decreasing evening temperatures (such as when dew formed).

Table 5. Amounts of Hydrocarbons (ng) and Oxygenated Hydrocarbons (μg) in Lab Blanks and Field Blanks and LODs during Phase 1 Sampling

Compound	Lab Blanks			Field Blanks			LOD	
	N	Mean	SD	N	Mean	SD	ng	$\mu\text{g}/\text{m}^3$
2-Methylhexane	33	0.05	0.18	49	0.07	0.27	1.59	0.22
2-Methylpentane	33	0.15	0.34	49	0.33	0.73	0.81	0.11
3-Methylpentane	33	0.07	0.23	49	0.12	0.38	1.14	0.16
2,3-Dimethylpentane	33	0.02	0.12	49	0.13	0.53	2.19	0.30
2,2,4-Trimethylpentane	33	0.07	0.16	49	0.19	0.55	0.24	0.03
Methylcyclohexane	33	0	0	49	0.04	0.16	1.65	0.23
1,3-Butadiene	33	0.02	0.1	49	0.01	0.08	0.24	0.03
Benzene	33	1.89	1.99	49	2.29	2.17	6.51	0.90
Toluene	33	1.25	1.79	49	1.33	1.15	3.45	0.48
<i>m</i> & <i>p</i> -Xylenes	33	0.88	2.06	49	0.86	1.24	3.72	0.52
<i>o</i> -Xylene	33	0.11	0.27	49	0.16	0.27	0.81	0.11
Ethylbenzene	33	0.12	0.25	49	0.15	0.20	0.60	0.08
Styrene	33	0.28	0.44	49	0.37	1.00	3.00	0.42
MTBE	33	0.02	0.1	49	0.02	0.09	0.27	0.04
Formaldehyde	34	0.03	0.02	47	0.05	0.06	0.18	0.03
Acetaldehyde	34	0.03	0.01	47	0.03	0.04	0.12	0.02
Acetone	34	0.13	0.07	47	0.12	0.16	0.48	0.07

Note: The LOD for hydrocarbon concentrations was calculated using a flow rate of 10 mL/min and a duration of 12 hours.

Table 6. Amounts of Hydrocarbons (ng) and Oxygenated Hydrocarbons (μg) in Lab Blanks and Field Blanks and LODs during Phase 2 Sampling

Compound	Lab Blanks			Field Blanks			LOD	
	N	Mean (ng)	SD	N	Mean (ng)	SD	ng	$\mu\text{g}/\text{m}^3$
2-Methylpentane	12	0.12	0.10	40	0.12	0.13	0.39	0.05
2-Methylhexane	12	0.07	0.23	40	0.07	0.22	0.66	0.09
3-Methylpentane	12	0.06	0.06	40	0.09	0.17	0.51	0.07
2,3-Dimethylpentane	12	0.02	0.03	40	0.04	0.09	0.27	0.04
2,2,4-Trimethylpentane	12	1.36	0.51	40	2.20	0.95	2.85	0.40
Methylcyclohexane	12	0.40	0.28	40	0.90	1.47	4.41	0.61
1,3-Butadiene	12	0.20	0.13	40	0.26	0.31	0.93	0.13
Benzene	12	0.35	0.26	40	0.37	0.46	1.38	0.19
Toluene	12	0.34	0.20	40	0.33	1.40	4.20	0.58
<i>m&p</i> -Xylenes	12	0.17	0.15	40	0.09	0.08	0.24	0.03
<i>o</i> -Xylene	12	0.14	0.12	40	0.39	0.30	0.90	0.13
Ethylbenzene	12	0.08	0.09	40	0.04	0.07	0.21	0.03
Styrene	12	0.38	0.43	40	0.26	0.45	1.35	0.19
MTBE	12	0.12	0.17	40	0.10	0.14	0.42	0.06
Formaldehyde	25	0.05	0.02	46	0.00	0.01	0.03	0.00
Acetaldehyde	25	0.03	0.02	46	0.01	0.01	0.03	0.00
Acetone	25	0.30	0.16	46	0.05	0.20	0.6	0.08

Note: The LOD for hydrocarbon concentrations was calculated using a flow rate of 10 mL/min and a duration of 12 hours.

METHODS USED IN THE NCI TRUCKING INDUSTRY PARTICLE STUDY

The methods used in the NCI Trucking Industry Particle Study to sample PM_{2.5}, EC, and organic compounds and the source-apportionment samples are described in detail in Appendix A.

REAL-TIME MONITORING METHODS

Photo-Ionization Detector Monitor

Total VOCs were measured by a PID monitor with a 10.6 eV lamp (model ppbRAE Plus). Our goal was to use the PID monitor to assess short-term variations, referred to as response factors, in the total amount of VOCs that might be linked to variations in source emissions. The PID monitor is highly sensitive to VOCs but has variable sensitivity to other compounds (these are expressed in terms of correction factors in Table 3). The response factors of acetaldehyde, hexane, and heptane were substantially higher than those of the other listed compounds; the response factors of higher-molecular-weight alkanes were close to or less than 1.0; and those of the aromatic compounds tended to

be close to 0.5. As a result, the response factor of gasoline vapor, which is a mixture of small amounts of hundreds of alkanes and aromatics, was an average of 1.0. Variations in refining, crude petroleum source, and blending make small changes in the average response factor. Diesel fuel, which has a higher average molecular weight and more aromatic components, has a lower response factor (0.4). Mixtures with a relatively fixed composition of many components, such as gasoline and diesel vapors, have relatively consistent response factors.

A similar situation can occur with environmental contaminants when there are mixtures of emissions from the same set of sources, such as traffic. This situation was seen in the consistency of groupings of compounds found in the principal components analysis (see below). However, variation in the relative numbers of cars and trucks and other sources affected the PID monitor response per unit of mass, shifting it closer to 1.0 or 0.5, and introduced noise into the relationship.

The output of the PID monitor could not be directly compared with the findings from the matching sorbent tube analyses. A sensitivity-weighted sum of the measured masses could not be calculated, because we did not have

the response factors for the hundreds of individual components seen in the GC–mass spectrometry (MS) analyses. Also, we expected that this would underestimate the total, because a large number of trace components in the environmental samples were at or below the LODs for gas chromatography peaks and were not individually quantified in the sorbent tube analyses.

DustTrak PM_{2.5} Monitor

PM_{2.5} was measured using the DustTrak aerosol monitor, which operated by light scattering. Each unit was sent back to the manufacturer for an annual recalibration. A small impactor on the inlet removed particles larger than 2.5 µm. The detection system had varying sensitivity by particle size, density, and composition; it was calibrated using standard inorganic dust. Urban PM_{2.5} has a smaller particle size and more than 50% organic materials, unlike the standard inorganic calibration dust; as a result, the mean DustTrak value during a concurrent TWA air sample tended to underestimate the TWA air concentration. Thus, there were errors for direct comparisons of the mean real-time concentrations with the integrated TWA gravimetric measurements. Because the composition and size of urban particles were relatively stable for 8 to 12 hours in a given location, a filter sample collected concurrently could be used as a one-point calibration for the mean DustTrak reading for that period and location.

Weather Data Monitoring

Local weather data were collected about every 12 hours during the 5-day sampling visit at each terminal. Real-time monitoring of wind direction and speed, temperature, and relative humidity were measured at 5-minute intervals for our on-site measurements using a weather monitor (Weather Monitor II, Davis, Hayward, CA). Local hourly observations at a nearby airport were also obtained from an online source (www.wunderground.com) for temperature, relative humidity, wind speed and direction, and precipitation. Data from our on-site weather station were cross-checked with the online data. They were consistently highly correlated ($R^2 > 0.9$) but not identical, which is consistent with their geographic separation of 1 to 2 miles.

DEVELOPMENT OF DATABASE AND ASSOCIATED MATERIALS

Extensive data-management and quality-control methods were used to combine the real-time and integrated sampling data and weather station data into a single format matched by time and then compiled into a database. We also performed a thorough descriptive analysis of each monitoring session, which included more than 50 12-hour sessions at

six terminals. As part of this descriptive work, potential emission sources around the terminal were identified using satellite images and GIS mapping software; wind roses were constructed and superimposed on the images for each session to determine the potential for wind transport during these time periods. To the extent possible, a time profile around each location was also constructed (using traffic data, terminal freight logs, etc.). Real-time and integrated data were added to the database and matched for time periods to test for consistency across the two sampling methods (see discussion of data processing in the Real-Time Descriptive Analyses section below).

STATISTICAL METHODS

All of the statistical analyses were performed using STATA, Version 8.2, software (StataCorp, College Station, TX). The concentration data were approximately log normal and were log-transformed where necessary to meet the normality assumptions of linear regression modeling. All initial data comparisons were made using standard nonparametric tests (the Spearman rho, Wilcoxon rank sum, and Kruskal–Wallis tests). When concentration values were below the LOD (see earlier discussion), half of the minimum value detected during sampling was substituted. With the exception of a substantial number of “nondetects” for 1,3-butadiene and MTBE (for which substitutions were not made), this affected less than 5% of the overall concentrations monitored in the yard and 1% of driver samples. Specification checks were performed with and without the substitutions, and they did not significantly alter the results. The primary statistical modeling tools applied in this study were principal components analysis and structural equation modeling.

PRINCIPAL COMPONENTS ANALYSIS

Principal components analysis is a statistical tool for data reduction that uses a correlation matrix to identify a more limited set of linear combinations that are characteristic of the entire dataset. This method has been used in environmental health applications to determine underlying source characteristics of exposures at a given location (Hopke et al. 2006). We applied this method to identify the source characteristics of particulates, VOCs, and aldehydes at the six terminal locations revisited during Phase 2. The repeat site visits were the only visits with sufficient data to perform this analysis (data from two sampling visits for each location); the fact that they were performed during the same season was consistent with the necessary principal-components-analysis assumption of homogeneity across the sampling periods.

STRUCTURAL EQUATION MODELING

Structural equation modeling was used to develop the on-site terminal exposure model. This method is becoming increasingly popular among environmental epidemiologists as a way of handling high-dimensional data (Sanchez et al. 2005). Structural equation modeling is useful in understanding causal pathways and identifying the indirect effects of intermediate variables on a primary dependent variable (such as occupational exposure). In our setting, structural equation modeling provided a way to analyze the data that reflected the natural hierarchy present in our on-site terminal sampling scheme, namely background, work area, and personal exposures. This relationship is exemplified in the pathway diagram shown in Figure 3.

In particular, the nature of our sampling plan imposed a complex covariance structure on the collected data because the concurrent measurements taken by personal sampling, stationary work area samplers, and external measurements of background conditions were not independent. Various emission sources contributed simultaneously to the measurements observed at various locations within the terminals during the same time periods. Of particular statistical concern was the correlation among the

error terms as well as the correlation among the response variables and the error terms. Both of these conditions violated necessary assumptions for linear regression modeling.

Therefore, instead of trying to fit one large model encompassing all covariates simultaneously, we fitted three related models. Using structural equation modeling, we simultaneously predicted personal exposures as a function of work-related exposure and smoking status; then work-related exposure as a function of terminal characteristics, indoor ventilation, job location, and background exposure conditions; and finally background exposure conditions as a function of weather, nearby source pollution, and other regional differences across terminal sites. This multi-layered structure allowed us to use the statistical technique known as three-stage least squares, a common structural equation modeling approach in econometrics (Zellner and Theil 1962; Goldberger 1972). The advantage of this method is that it provided coefficient estimates for all of the covariates in the model along with equation-specific R^2 values to interpret each level of exposure data.

The structural equation modeling for particulates estimated exposure for person i in job location j at terminal k , as measured by the concentration values collected from

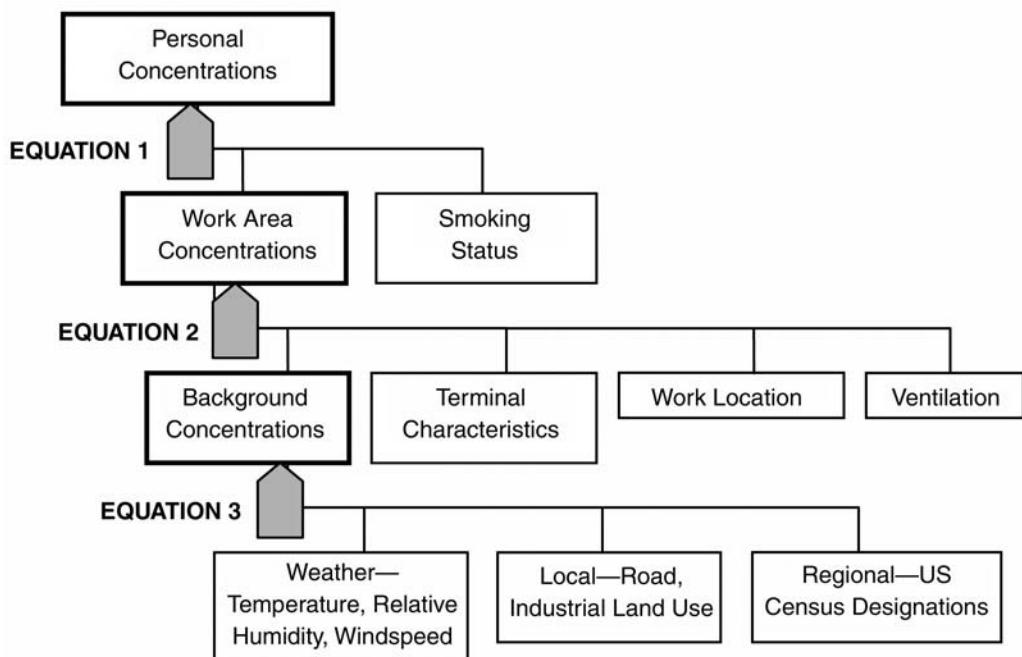


Figure 3. Diagram of structural equation model pathway for layered exposure model.

the personal sampling jackets worn by the employees during their work shifts as part of the NCI-funded study. The data were formatted to match the personal samples by time period (session) with indoor work exposures and outdoor background conditions as well as job location, terminal characteristics, smoking status, and other covariates for each subject. The structural equation modeling for VOCs and aldehydes was slightly revised to accommodate the lack of personal measurements; instead we modeled two levels of exposure (outdoor background and indoor work area). When multiple area measurements were collected during a single session, the average value was used in the statistical model. Finally, the exposure data were lognormally distributed and have been log-transformed for normality. Therefore, the estimated coefficients were interpreted within the context of a multiplicative model.

GEOGRAPHIC INFORMATION SYSTEM

Using GPS technology, a series of spatially referenced background characteristics (variables) was constructed during Phase 2 around the geocoded addresses for all terminal locations and along each driver's route. A spatial map was created with GIS (ArcGIS 9.0, ESRI, Redlands, CA) that included the location of potential point sources and air pollution monitors, land-use characteristics, road characteristics, and characteristics of other transportation networks. This approach produced a very large database of exposures and other measurements collocated by time and geographic location.

The locations of potential point sources and existing air pollution monitors were available from the EPA Web site. Our use of the point-source locations was limited to a descriptive analysis, because data on temporally relevant discharge quantities were not available for our study dates. The nearby monitors for PM_{2.5} and carbon monoxide from the Air Quality System, an EPA monitoring network (www.epa.gov/ttn/airs/airsaqs/index.htm), were temporally matched to our study periods at various spatial scales (nearest monitor and 50- and 100-kilometer buffers) to validate the exposure measurements.

Land-use characteristics were derived from satellite imagery available from the 1992 National Land Cover Data of the U.S. Geological Survey (USGS; <http://landcover.usgs.gov/natllandcover.php>). These raster datasets were extrapolated to points, and variables were then generated for the percentage of land-cover classifications designated "industrial, commercial, and transportation" (ICT), "high intensity residential," and "low intensity residential." These points represented the land-cover characteristics for any given point on the spatial surface and were spatially matched to describe both terminal and driver exposures.

The road networks were available from the U.S. Census Bureau by way of the Environmental Systems Research Institute (Census 2000 TIGER/Line Files, available at www.esri.com/data/download/census2000-tigerline). These data were matched with driver GPS data to determine route-specific location information. The same data were also used to determine road network characteristics around the terminals. For the terminal location, a variable was generated to indicate the distance to a major road (any type), but it showed little variability across terminals, because all were located near major road intersections. However, a more interesting and variable feature was the terminal's proximity to heavily trafficked interstate highways. Eleven of the initial 36 terminals were located within 500 meters of an interstate highway, and several were in close proximity to several interstate highways. We therefore generated two road-proximity measures for a terminal: continuous distance in meters to an interstate and a dummy variable for locations within 500 meters of an interstate.

REAL-TIME DESCRIPTIVE ANALYSES

During the evaluation of Phase 1 of the study, we identified two primary limitations in the study design, both of which derived from the exclusive use of integrated sampling methods. The study design was subsequently improved with the supplemental collection of real-time data in Phase 2, such as the GPS driver locations, weather data, and terminal activity data (including logs of truck traffic). The additional data collected during Phase 2 have been explored in detail for each session, which was a large task, given the complexity and size of the data sets. These largely descriptive analyses allowed us to identify important trends in exposure across the upwind and downwind locations and provided an important area of ongoing research for the study team.

Data Processing for Upwind and Downwind Contributions

Exposure data for real-time PM_{2.5} and VOCs from the four yard monitors at each terminal were collected every minute and matched by time. Nonmatching data for times at the beginning or end of each sampling period were removed. Our weather station data were recorded every 5 minutes, which was deemed the most suitable time interval for matching weather characteristics with the real-time exposure data. The 5-minute averages were separated and graphed for 12-hour periods from 10:00 AM to 10:00 PM and from 10:00 PM to 10:00 AM to enable us to look for trends and compare the real-time values with the integrated TWAs. Graphs depicting all PM_{2.5} and VOC

real-time concentrations and accompanying wind roses were generated for each sampling session. Proprietary overhead shots of the area were used to identify potential local sources contributing to terminal exposures. These data were combined in a descriptive analysis of exposure trends and terminal contributions for more than 50 sessions during the six Phase 2 sampling visits.

Only 27 TWA VOC samples were obtained that provided detailed composition data for periods with concurrent real-time measurements of total VOCs. We attempted to compare the session average of real-time total VOC data with the individual TWA component data from the analysis of the integrated samples. The correction factors were applied to 21 of the 27 samples analyzed. Correction factors from REA Systems Technical Manual TN-106 (see Table 3) for each sample were multiplied by individual concentrations, summed, and divided by the elapsed time of the session. Compounds that did not respond to a 10.6-eV lamp or did not have a correction factor were not included; these compounds were 1,3-dichloropropene, carbon tetrachloride, chloroform, methylene chloride, and trichloroethylene. That is why the ppbRAE monitor's total VOC measurements were not equal to the sum of the weighted individual measured components.

Data Processing for Driver Exposures

We imported GPS data collected for all drivers' trip routes into a GIS database and superimposed them on the U.S. Census Bureau Tiger 2000 Transportation layer to create maps with both geographic and time information. Based on time and location, the geographic and pollutant monitoring data could be combined into one data set matching the real-time location information and real-time pollutant levels information for each truck route. Local land-use layers, traffic density, point sources, weather information, and nearby EPA monitor data were added to the database. A descriptive data analysis was conducted to arrive at temporal and spatial patterns of exposure for PM_{2.5} and VOCs.

Each driver's trip data were descriptively analyzed. First, the peak concentrations (values greater than the 95th percentile for the trip) for VOC and PM_{2.5} time traces were located on the trip map. Then the characteristics of the setting and vehicle activity at that time point and location were examined. Satellite photos were retrieved for locations where the truck stopped (which showed other truck terminals, warehouses, and shopping centers) or for locations where there were broader periods of high concentrations. We also looked for systematic linkages with road types and areas coded for high industrial or residential land uses.

RESULTS

The results of this study are presented in three parts. The first part presents the results for the structural equation modeling method development. The second part presents the results associated with the two terminal exposure hot spots identified in our original hypotheses: upwind (industrial parks) and downwind (residential neighborhood) exposures. The third part presents the results associated with our third hot spot, the on-road driver exposures in P&D truck cabs. The Results sections begin with an overview of the exposure scenario and summary statistics, followed by the modeling results from Phases 1 and 2 of sampling. We also present an exposure model based on structural equation modeling, an assessment of the relationship between VOCs and PM_{2.5}, source apportionment analysis, an assessment of long-term temporal stability, and the analysis of real-time data.

DEVELOPMENT OF STRUCTURAL EQUATION MODELING APPLICATION

While the field sampling was being conducted in Phase 1, an extensive effort was made to develop the application of structural equation modeling for our data. Dr. Davis developed the necessary programming and model evaluation in STATA software. As noted earlier, structural equation modeling was expected to have major advantages, given the layered structure of our sampling strategy and data. Our sampling strategy was to concurrently collect samples representing the local outdoor area outside the terminal (upwind yard background), the work area indoors, and personal exposures indoors. Each of these had unique features, but they were not independent of each other. An existing particulate data set had already been collected from an earlier set of truck terminals, where we were using EC as a marker for diesel exposures. Additional detail on the results of the structural equation model applied to particulates is available in Appendix A.

PHASE 1 FINDINGS — TRUCK TERMINAL HOT SPOTS

One of our primary hypotheses was that samples collected at the fence line on the upwind and downwind sides of a truck terminal could indicate the distribution of VOC and PM_{2.5} exposures found in residential areas near industrial parks and commercial areas where there is light industry, other truck terminals, large retail stores, distribution warehouses, and heavy truck traffic. Designating a site as a hot spot did not mean that the concentrations in that area would always be high. Rather it meant that there was a higher likelihood that exposures would be above the

regional average, which might or might not represent a health hazard. The measurements represented the upper boundary of exposures that might occur in a residential neighborhood located near an industrial or commercial area or adjacent to a terminal. However, such nearby residential neighborhoods were not present at all of our study sites, so for those locations without residential neighborhoods the measurements represented potential exposures, but they could still indicate the range and intensity of these exposures.

The transport of locally intense emissions into a local neighborhood by local winds was the defining feature of downwind hot spots. When there was no wind or very strong winds, the concentrations of emissions at a modest distance outside the terminal were unaffected by local terminal emissions. Thus, both significant emissions and wind transport (correct direction and moderate wind speed) had to be present to produce high neighborhood concentrations.

A GIS database was used to compile spatially referenced descriptive information on each terminal visited in order

to characterize the upwind source characteristics and potential downwind neighborhood concentrations. On average, approximately 25% of the areas in the immediate vicinity (< 1 kilometer) of the monitored terminals were categorized as ICT land use by the USGS. However, this categorization was an imprecise indication of emission sources or emissions. The percentage of areas around terminals in this land-use category for the terminals we visited varied from 6% at the Maryland terminal to 92% at the Florida terminal. The terminals were often nested within dense local road networks a few kilometers from a heavily trafficked interstate. However, the terminal's proximity to an interstate was not always close; it varied from immediately adjacent to the intersection of multiple large interstates to more than 7 kilometers away from the nearest interstate. These terminal features are described in more detail in Table 2.

Figure 4 shows GIS maps of two cities visited during our study that had potential upwind sources and downwind neighborhood exposures. One map shows two terminals located in a large city surrounded by an extensive interstate

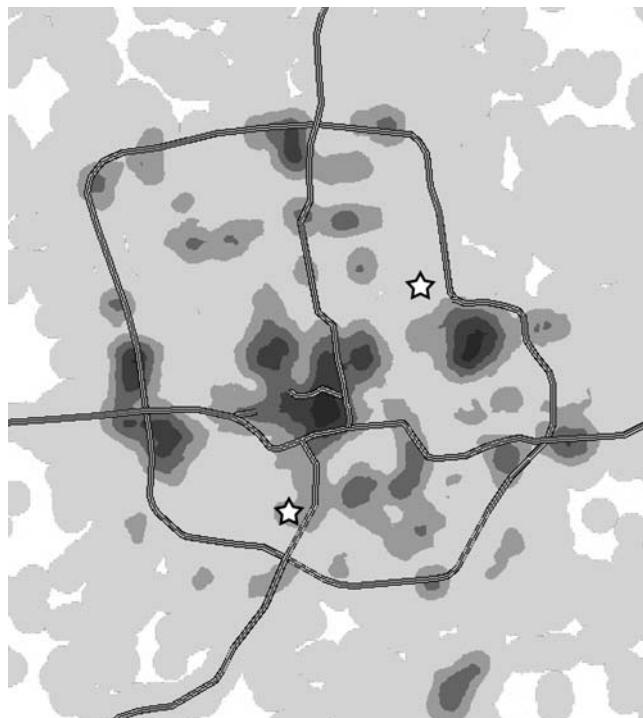
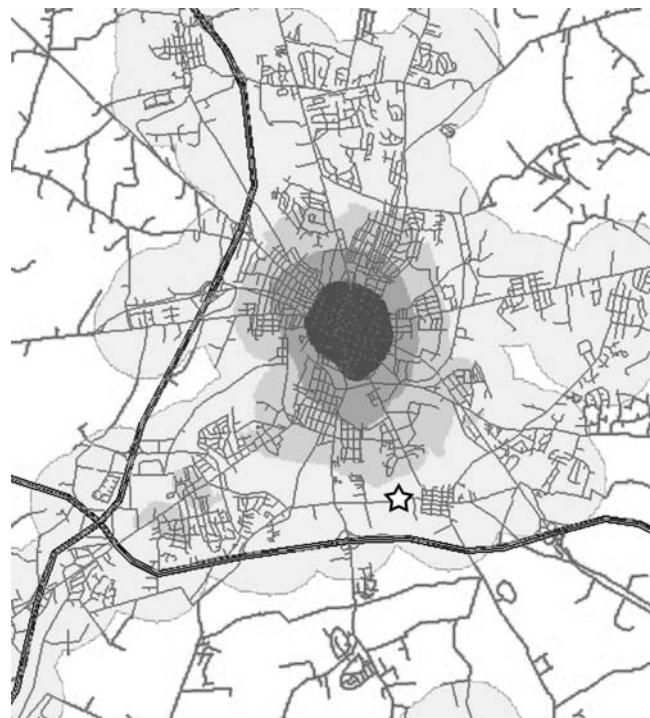


Figure 4. GIS characterization of truck terminal locations. (Left) GIS map of two terminals (stars) in one city. Darker shades indicate high-intensity industrial activity. (Right) GIS map of a terminal (star) in a medium-size city with overlays of the local road network, interstate highways, and nearby residential land uses. Darker shades indicate denser residential areas.



road network (located approximately 1 kilometer from both terminals) and by pockets of high-intensity industrial activity (indicated by the darker shaded areas). Both traffic and nearby industrial activity at these locations could be significant sources of terminal exposures if wind conditions are favorable for transport. The second map shows a terminal located in a medium-size city with overlays of the local road network, interstate highways, and nearby residential land uses (gray shades represent denser residential areas). Other than the interstate, which is located less than 1 kilometer from the terminal, there were very few industrial sources in close proximity to this terminal. However, the terminal was close to a heavily populated commercial downtown neighborhood (north-northwest of the terminal), which could be a significant source of exposure to this population if winds are favorable for transport.

Upwind Terminal Summary Statistics

Summary statistics for the integrated samples upwind in the yard and indoors for hydrocarbons, aldehydes,

PM_{2.5}, EC, and OC are compared in Table 7 and Table 8. This comparison includes semi-enclosed indoor loading dock and indoor mechanic shop work area measurements from both phases of the study. The largest quantity of total contaminants could be attributed to aromatics, with toluene accounting for the majority of these. The alkanes and aldehydes were next highest, with formaldehyde at concentrations comparable to those of toluene. With the exception of trimethylpentane and benzene, all concentrations indoors were significantly different across the locations (Kruskal-Wallis test, $P < 0.05$). Although the yard upwind background concentrations were lower than both of the sampled indoor locations, the area of highest exposures varied between the shop and the dock. For example, the shop concentrations were elevated for xylenes, alkanes, acetone, and PM; the dock, with its propane-powered forklifts, had comparatively high concentrations of 1,3-butadiene, benzene, and aldehydes. In the structural equation modeling work, discussed below, the upwind concentrations were identified as important contributors to the exposures in indoor locations.

Table 7. Comparison of Summary Statistics for Upwind and Downwind Samples

Compound ($\mu\text{g}/\text{m}^3$)	Yard Upwind			Yard Downwind				
	Observations (N)	Mean	Median	SD	Observations (N)	Mean	Median	SD
Trimethylpentane	178	0.73	0.43	0.83	144	0.63	0.41	0.70
Dimethylpentane	178	0.32	0.23	0.33	144	0.31	0.22	0.31
2-Methylhexane	178	0.52	0.41	0.43	144	0.53	0.43	0.49
Methylpentane	178	1.67	1.06	2.99	144	1.43	1.06	1.30
3-Methylhexane	178	0.61	0.43	0.63	144	0.64	0.50	0.62
Methylcyclohexane	178	0.50	0.27	0.67	144	0.49	0.29	0.87
Hexane	37	1.23	0.82	1.20	37	1.42	1.03	1.19
1,3-Butadiene	178	0.15	0.05	0.36	144	0.14	0.10	0.19
Benzene	178	1.29	1.05	0.91	144	1.25	1.06	0.89
Ethylbenzene	178	0.60	0.42	0.55	144	0.55	0.39	0.53
<i>m&p</i> -Xylenes	178	1.99	1.50	1.68	144	1.76	1.34	1.60
<i>o</i> -Xylene	178	0.72	0.51	0.66	144	0.62	0.45	0.60
Styrene	178	0.26	0.15	0.37	144	0.25	0.17	0.33
Toluene	178	3.65	2.82	3.03	144	3.62	2.65	4.14
MTBE	178	0.27	0.01	0.50	144	0.27	0.02	0.46
Acetone	137	2.81	1.68	3.69	110	2.17	1.59	2.27
Acetaldehyde	137	2.60	2.15	2.97	110	2.41	1.95	2.67
Formaldehyde	137	3.32	3.17	1.91	110	3.06	3.21	1.68
EC	179	0.76	0.52	0.78	142	0.79	0.57	0.71
OC	179	5.15	3.77	9.94	142	4.63	4.20	2.48
PM _{2.5}	179	10.74	8.50	8.01	139	10.83	9.72	6.58

Table 8. Comparison of Summary Statistics for Yard, Dock, and Shop Samples

Compound ($\mu\text{g}/\text{m}^3$)	Yard Upwind			Dock			Shop					
	Observations (N)	Mean	Median	SD	Observations (N)	Mean	Median	SD	Observations (N)	Mean	Median	SD
Trimethylpentane	432	0.71	0.43	0.79	64	0.87	0.53	0.99	19	2.57	1.04	4.50
Dimethylpentane	432	0.38	0.24	0.47	64	0.54	0.31	0.51	19	2.21	0.95	3.09
2-Methylhexane	432	0.64	0.42	0.95	64	0.90	0.59	0.88	19	3.98	0.82	7.13
Methylpentane	432	1.60	1.04	2.28	64	2.34	1.33	2.47	19	7.42	2.56	13.26
3-Methylhexane	432	0.76	0.48	1.28	64	1.04	0.72	1.08	19	4.91	1.06	8.73
Methylcyclohexane	432	0.46	0.27	0.68	64	0.63	0.36	0.91	19	1.77	0.65	2.97
Hexane	161	1.70	1.01	1.86	44	3.05	1.59	4.13	15	2.49	1.58	2.35
1,3-Butadiene	432	0.20	0.12	0.45	64	0.75	0.60	0.57	19	0.30	0.24	0.28
Benzene	432	1.24	1.01	0.87	64	1.54	1.15	1.26	19	1.14	0.90	0.92
Ethylbenzene	432	0.56	0.40	0.53	64	1.03	0.71	0.92	19	7.28	2.33	8.87
<i>m&p</i> -Xylenes	432	1.85	1.36	1.69	64	2.85	1.98	2.54	19	23.01	8.50	26.89
<i>o</i> -Xylene	432	0.66	0.46	0.63	64	0.90	0.64	0.74	19	8.44	2.50	10.71
Styrene	432	0.26	0.15	0.36	64	0.74	0.51	0.62	19	0.60	0.49	0.56
Toluene	432	3.67	2.77	3.43	64	10.54	7.08	11.20	18	5.56	3.78	4.75
MTBE	432	0.27	0.01	0.53	64	0.29	0.04	0.66	19	0.62	0.03	1.65
Acetone	345	2.62	1.75	3.07	65	9.44	4.93	17.55	17	19.19	12.09	14.98
Acetaldehyde	345	2.42	2.06	2.45	65	6.27	4.84	4.27	17	45.00	3.64	100.51
Formaldehyde	345	3.33	3.22	1.75	65	25.44	18.96	19.03	17	13.72	5.05	16.63
EC	432	0.83	0.57	0.77	67	1.12	0.94	0.74	15	1.75	1.26	1.14
OC	432	4.93	4.15	6.60	67	7.93	7.55	2.75	15	7.87	6.97	3.27
PM _{2.5}	427	11.67	9.30	14.63	66	13.88	10.87	8.22	15	19.58	18.43	7.97

Within the set of VOCs measured, there were some strong correlations with PM components among the individual components in both the upwind yard and indoor work location data (Table 9). In the upwind VOCs, all of the aliphatic and aromatic components were strongly correlated with EC (0.54–0.62) and less so with PM_{2.5} (0.32–0.49). A similar pattern was seen in the indoor loading dock data. However, the shop indoor VOC data showed more inconsistent correlations with the individual aliphatic and aromatic components and PM components. For example, benzene, styrene, and toluene showed reasonable correlations with EC (0.51–0.67) but not with PM_{2.5} (0.22–0.34), but ethylbenzene and the xylenes had negative correlations with both. The aliphatics in the shop were correlated with EC (0.34–0.61) but not with PM_{2.5} (0.04–0.35). Acetone, acetaldehyde, and formaldehyde were unevenly correlated with EC (0.80, 0.18, and 0.91, respectively) and PM_{2.5} (0.18, 0.57, and 0.47, respectively). The shop had lower correlations because of larger and different sources in the shop, such as low-volatility aromatic

cleaning solvent, spilled fuel, and low-temperature exhaust from lightly loaded diesel engines (when truck tractors were driven into the shop). When we examined VOCs in the principal component analysis there were similar patterns of associations within the chemical groups, as expected.

Are these upwind concentrations an indication of the presence of hot spots? To investigate this possibility, we compared our measurements with measurements of separately monitored EPA levels in several locations. Table 10 shows a comparison of data from an EPA Air Toxics Monitoring Program (2006) with data from our measurements. The means and medians of these two data sets were generally very similar; the means for styrene and acetone were significantly higher in our study, but the increase was rather small, and many of the other measures were a little larger in the EPA data set. Table 11 compares our data set with four data sets from other studies of neighborhood air toxics concentrations, including inner-city locations in New York and Los Angeles. In most cases the data for 1,3-butadiene were similar to our measurements, all the data for the aromatics

Table 9. Correlations^a of VOC Data with EC and PM_{2.5} Data

Compound	Loading Dock		Mechanic Shop		Yard Upwind	
	EC	PM _{2.5}	EC	PM _{2.5}	EC	PM _{2.5}
Trimethylpentane	0.53 ^b	0.56 ^b	0.41 ^b	0.21	0.52 ^b	0.37 ^b
Dimethylpentane	0.57 ^b	0.43 ^b	0.60 ^b	0.04	0.54 ^b	0.32 ^b
2-Methylhexane	0.54 ^b	0.40 ^b	0.56 ^b	0.06	0.60 ^b	0.35 ^b
Methylpentane	0.59 ^b	0.46 ^b	0.47	0.19	0.56 ^b	0.32 ^b
3-Methylhexane	0.57 ^b	0.45 ^b	0.56 ^b	0.04	0.59 ^b	0.35 ^b
Methylcyclohexane	0.54 ^b	0.50 ^b	0.61 ^b	0.19	0.63 ^b	0.44 ^b
Hexane	0.23	0.52 ^b	0.34	0.35	0.71 ^b	0.24 ^b
1,3-Butadiene	0.34 ^b	0.44 ^b	0.66 ^b	0.24	0.39 ^b	0.53 ^b
Benzene	0.57 ^b	0.63 ^b	0.67 ^b	0.34	0.59 ^b	0.35 ^b
Ethylbenzene	0.33 ^b	0.39 ^b	-0.13	-0.33	0.61 ^b	0.39 ^b
<i>m&p</i> -Xylenes	0.40 ^b	0.38 ^b	-0.13	-0.33	0.60 ^b	0.37 ^b
<i>o</i> -Xylene	0.42 ^b	0.40 ^b	-0.13	-0.33	0.62 ^b	0.40 ^b
Styrene	0.53 ^b	0.21	0.51 ^b	0.30	0.61 ^b	0.49 ^b
Toluene	0.09	0.32 ^b	0.55 ^b	0.22 ^b	0.61 ^b	0.39 ^b
MTBE	-0.13	0.51 ^b	0.44	0.06	0.39 ^b	0.53 ^b
Acetone	0.40 ^b	0.38 ^b	0.80 ^b	0.18	0.28 ^b	0.40 ^b
Acetaldehyde	0.12	0.70 ^b	0.18	0.57 ^b	0.24 ^b	0.27 ^b
Formaldehyde	0.24 ^b	0.51 ^b	0.91 ^b	0.47	0.28 ^b	0.26 ^b

^a Nonparametric Spearman correlation coefficients.

^b Significant at the 5% level.

Table 10. Comparison of Data from EPA Air Toxics Monitoring Program^a and Yard Upwind Sites

Pollutant ($\mu\text{g}/\text{m}^3$)	EPA Air Toxics Monitoring Program				Yard Upwind Sites			
	Detects (N)	Mean	Median	SD	Observations (N)	Mean	Median	SD
1,3-Butadiene	789	0.18	0.13	0.15	432	0.20	0.12	0.45
Benzene	1291	1.69	1.12	2.68	432	1.24	1.01	0.87
Ethylbenzene	1223	0.78	0.48	1.09	432	0.56	0.40	0.53
Styrene	961	0.42	0.21	0.81	432	0.26	0.15	0.36
Toluene	1294	3.96	2.56	5.28	432	3.67	2.77	3.43
<i>m&p</i> -Xylenes	1260	1.82	1.13	2.56	432	1.85	1.36	1.69
<i>o</i> -Xylene	1201	0.82	0.52	1.00	432	0.66	0.46	0.63
Acetaldehyde	1606	2.39	1.82	2.23	345	2.42	2.06	2.45
Acetone	1606	2.07	1.62	1.67	345	2.62	1.75	3.07
Formaldehyde	1600	6.37	2.51	21.83	345	3.33	3.22	1.75

^a U.S. EPA 2006.

Table 11. Comparison of Yard Upwind Values with Other Studies of Neighborhoods

Pollutant ($\mu\text{g}/\text{m}^3$)	Yard Upwind	RIOPA Study Outdoor Samples ^a			Minneapolis Neighborhoods ^b			New York Inner City Neighborhood ^{c,d}			Los Angeles Inner City Neighborhood ^c			Brisbane Eagle Farm Light Industry ^e			
		N	Median	SD	N	Median	SD	N	Median	SD	N	Median	SD	N	Median	SD	
1,3-Butadiene	432	0.12	0.45	—	—	—	—	31	0.1	0.2	35	0.2	0.4	—	—	—	
Benzene	432	1.01	0.87	554	2.15	2.11	113	1.3	2.2	31	0.1	0.4	32	0.01	—	—	
Ethylbenzene	432	0.40	0.53	554	1.29	1.87	113	0.6	0.8	31	1.1	1.2	35	4.3	1.7	28	10.5
Toluene	432	2.77	3.43	554	7.09	6.47	113	2.6	4.2	31	0.5	0.7	32	2.3	0.8	31.9	31.9
<i>m&p</i> -Xylenes	432	1.36	1.69	554	3.57	4.15	113	2.3	3.3	31	1.1	1.7	35	2.9	1.4	28	5.9
<i>o</i> -Xylene	432	0.46	0.63	554	1.48	3.90	113	0.8	1.1	31	0.7	0.9	36	1.7	0.5	32	40.3
Acetaldehyde ^f	345	2.06	2.45	395	3.21	1.65	—	—	—	36	2.8	0.9	40	3.7	1.2	28	2.9
Formaldehyde ^f	345	3.22	1.75	395	3.02	3.00	—	—	—	36	2.1	0.9	35	4.1	1.6	35	4.0

^a Weisel et al. 2005.^b Adgate et al. 2004.^c Sax et al. 2004.^d Kinney et al. 2002.^e Hawas et al. 2002.^f Aldehydes were collected by passive sampling using the DNSH method (Herrington et al. 2005).

were higher, and most of the aldehydes data were comparable. Therefore, the upwind data showed no evidence of a toxic hot spot in the upwind neighborhoods.

Downwind Terminal Contributions

Our goal was to determine the relative contribution of within-terminal emissions to the TWA contaminant levels leaving the truck terminals and moving into the surrounding area. Clearly, this contribution is strongly dependent on the upwind contribution and wind transport (which depends on wind direction, stability, and speed). The comparison of all upwind (yard background) and downwind TWA samples collected is shown in Table 7.

Overall, the TWA samples for upwind and downwind measurements were not significantly different from each other in a series of robust comparison tests.

As noted earlier, wind direction was rarely stable for an entire integrated sampling period, which reduced the differences between upwind and downwind concentrations. Table 12 shows the ratios between the mean TWA downwind and upwind concentrations by terminal site. These ratios show that the terminals' VOC contributions were overall slightly greater than one. For the particulate contributions, the ratios were consistently greater than one, indicating a modest terminal contribution of 2%–3%. For the five repeat visits with available paired upwind and downwind data, when there was better spatial sampling to

Table 12. Ratios of Mean Downwind and Upwind Pollutant Concentrations by Terminal

Terminal ^a	EC	OC	PM _{2.5}	Butadiene	Benzene	Formaldehyde	Acetaldehyde
Oklahoma City (23)	0.29	0.57	0.37				
Columbus (24)	0.42	0.64	1.35		0.86	0.66	2.33
Milwaukee (25)	1.09	1.02	0.78	0.88	1.00	0.93	1.26
Memphis (26)	1.14	1.22	1.23		0.85	0.71	1.84
Phoenix (27)	0.80	1.25	—		0.72	2.67	1.88
Portland (28)	—	0.98	1.17		0.76	1.05	1.90
Denver (29)	1.58	1.18	1.44		1.35	1.34	1.06
Miami (30)	0.94	1.07	0.52		1.13		
Hagerstown (31)	0.93	1.40	1.58	0.71	0.82	0.65	0.58
Nashville (32)	0.72	0.82	0.64	1.11	0.84	0.98	0.94
Middletown (33)	0.93	0.84	1.53	0.56	1.11	1.16	1.36
Laredo (35)	1.20	0.92	1.10	0.89	1.01		
Philadelphia (36)	1.12	1.17	1.03	1.39	1.76	1.25	0.93
Repeat-Visit Terminals							
Philadelphia (37)	1.31	1.21	1.12	3.22	2.25	1.42	1.11
Columbus (38)	0.55	0.88	0.79	0.47	0.74	0.78	0.85
Milwaukee (39)	1.50	1.39	1.25				
Phoenix (40)	1.08	0.88	1.18	0.91	0.82	0.84	0.80
Denver (42)	2.93	1.07	1.20	1.18	1.63	1.11	0.68
All Visits^b							
Mean	1.03	1.03	1.02	1.13	1.10	1.11	1.25
SD	0.62	0.23	0.42	0.79	0.44	0.51	0.54
Median	1.01	1.04	1.14	0.90	0.93	1.01	1.08
Repeat Visits^c							
Mean	1.47	1.09	1.11	1.45	1.36	1.04	0.86
SD	0.89	0.22	0.19	1.22	0.72	0.29	0.18
Median	1.31	1.07	1.18	1.04	1.22	0.98	0.82

^a Number indicates the order in which the terminals were visited. Visits 1–21 were part of the NCI-funded study. No ratios are provided for Elizabeth (22), Houston (34), and Portland (41) because of sampler failures.

^b Ratios for all visits to terminals for which there were data.

^c Ratios for 6 repeat visits to terminals for which there were data. Portland visit (41) contributed no data for ratios because the downwind samples failed.

capture the differences, the mean and median differences were larger but still modest. The hydrocarbons and aldehydes showed more mixed effects. For all of the visits, all four VOCs had means greater than one, but the medians were much lower. Increased ratios were seen for 1,3-butadiene and benzene, 45% and 36% higher downwind, respectively, but not for the aldehydes for these five visits. This result indicated that the terminal activities had limited impact on the surrounding neighborhoods overall or that the sources, such as aldehydes, were from outside the terminal. Contributions from the internal emission sources were also probably obscured by within-session wind variations that were not captured by the integrated measurement strategy. A better approach would have been to measure the within-session variability for VOCs and PM using real-time monitoring, which is discussed in more detail in the next section, on Phase 2 investigations.

Source Characteristics

Two types of source evaluations were performed. First, a principal components analysis was performed to determine the relationships among the air contaminants. Those that occurred together tended to come from the same source categories, such as traffic emissions. The second analysis was a molecular-marker study performed in collaboration with Dr. James Schauer's laboratory (University of Wisconsin-Madison, Madison, WI). This analysis could provide a wider and yet more specific identification of emission sources contributing to the observed contaminants.

In the principal component analysis, the upwind background VOC and PM exposures could be collapsed into three primary factors that explained 80% to 92% of the total variability at each of the sites. Because of their relatively high percentages of missing samples, 1,3-butadiene and MTBE were excluded from these analyses. The principal component analysis results from the six terminal sites (Table 13) generally suggested a strong primary factor for hydrocarbon (evenly associated with the individual alkanes and aromatics) in these microenvironments, which was responsible for the majority of explained variability (46%–75%) in the entire dataset. A second smaller independent contribution, 11%–22%, usually came from formaldehyde and acetaldehyde and less so from acetone that loaded onto the second component. A weak variable third component, 5%–11%, had no consistent contributors, but some locations had formaldehyde and benzene, and others had some alkane and styrene contributions, probably reflecting local sources, such as gasoline filling stations and body shops. These results are consistent with the correlations noted in the previous section. The alkanes and aromatics in factor 1 were most likely from traffic sources upwind of the terminals. The aldehydes in factor

2, and sometimes in factor 3, were probably from photochemical atmospheric reactions in the region of the terminal. The principal component analysis was limited by relatively small sample sizes (between 30 to 50 observations per location for 15 different compounds). We expected that the source characteristics would have been more distinguishable using this method had a longer time period been observed at each location and more samples been collected.

A small set of high-volume particulate samples collected at a truck terminal in St. Louis, Missouri, were sent to Dr. James Schauer's lab for detailed characterization of OC and EC constituents. The findings are summarized in Table 14 and Table 15 (both tables are from Sheesley et al. 2009). These detailed results were used in a mass balance analysis to estimate the amounts of materials from various sources, in which the individual source categories have sets of molecular markers. Unfortunately, the patterns of markers overlapped for vehicle emissions, which made it difficult to distinguish between cars with spark ignitions and both light- and heavy-duty diesel trucks. Another problem was that engine emissions change with load and driving conditions. EC predominated in diesel emissions (Table 15) under high loads and acceleration, when there was less OC; under light loads or when idling, OC dominated. The contribution of lubricating oil to the emissions was noted in exhaust from both cars and trucks and was found in the fractions of OC comprising high-molecular-weight hydrocarbons (shown in Table 15). The various PAHs, hopanes, cholestanes, sitostanes, acosenes, and triacotanes are all associated with the lubricating oils used in motor vehicles.

As expected, each location did not have the same source contributions, which was most evident in the OC compounds. Cigarette smoke, which contains large amounts of OC, varied by location but was found in the highest concentrations in the driver and shop samples. Even some nonsmoking drivers had smoke-related OC in their personal-exposure samples, perhaps because the truck tractor had been previously used by a smoker (surface-deposited VOCs are off-gassed during truck use). *n*-Alkanes are important markers for cigarette smoke but can also be emitted from a variety of sources in the ambient or work environment. Given the upwind placement of the yard samplers, no cigarette smoke should have been present in the yard samples; this source was not calculated for the yard or urban background averages. However, this probably resulted in a small overestimation of the cigarette smoke in some of the other samples, such as the dock area, because the environmental *n*-alkanes would have been assigned to cigarette smoke.

The EC concentrations associated with diesel emissions that P&D drivers were exposed to were substantially higher

Table 13. Principal Component Analysis Results for Upwind Concentrations at Six Truck Terminals with Repeat-Site Visits^a**Philadelphia, PA (59 Observations)**

Component	Eigenvalue	Difference	Proportion	Cumulative
1	10.41148	8.72907	0.6941	0.6941
2	1.68240	0.56236	0.1122	0.8063
3	1.12004	0.60590	0.0747	0.8809
4	0.51414	0.15714	0.0343	0.9152
Eigenvectors: Variable		1	2	3
Trimethylpentane	0.29303	-0.08129	0.12258	
Dimethylpentane	0.30119	-0.04845	0.04001	
2-Methylhexane	0.29746	-0.03064	-0.09313	
Methylpentane	0.28291	-0.06777	0.03750	
3-Methylhexane	0.29599	-0.03356	-0.07716	
Methylcyclohexane	0.28915	0.06748	-0.21077	
Benzene	0.22826	-0.05374	0.38971	
Ethylbenzene	0.29374	-0.11086	-0.01200	
<i>m&p</i> -Xylenes	0.29325	-0.13020	0.02429	
<i>o</i> -Xylene	0.29020	-0.12509	-0.02509	
Styrene	0.23589	-0.11354	0.17484	
Toluene	0.27730	0.02761	-0.25639	
Acetone	0.04900	0.33342	0.78699	
Acetaldehyde	0.13029	0.64569	-0.21651	
Formaldehyde	0.15222	0.62475	-0.08452	

Columbus, OH (32 Observations)

Component	Eigenvalue	Difference	Proportion	Cumulative
1	8.06425	5.91380	0.5376	0.5376
2	2.15045	0.51269	0.1434	0.6810
3	1.63776	0.66599	0.1092	0.7902
4	0.97177	0.29168	0.0648	0.8549
Eigenvectors: Variable		1	2	3
Trimethylpentane	0.32007	0.07282	-0.01997	
Dimethylpentane	0.26094	0.30355	-0.12524	
2-Methylhexane	0.26046	0.27159	-0.15337	
Methylpentane	0.09789	-0.01519	0.73713	
3-Methylhexane	0.17612	0.24369	0.55146	
Methylcyclohexane	0.30933	0.09364	-0.22378	
Benzene	0.32060	0.01536	0.16374	
Ethylbenzene	0.31362	-0.21091	-0.03663	
<i>m&p</i> -Xylenes	0.32407	-0.20009	-0.01163	
<i>o</i> -Xylene	0.31756	-0.21312	0.00709	
Styrene	0.29831	-0.07938	-0.06648	
Toluene	0.28533	-0.03521	-0.14613	
Acetone	-0.17283	0.43333	-0.05872	
Acetaldehyde	0.01017	-0.45945	-0.03063	
Formaldehyde	0.13398	0.47310	-0.06598	

Table continues next page

^a Each factor has an eigenvalue (also called its latent root) associated with it that is related to the amount of variability it explains; these are ranked from the highest to the lowest. The eigenvectors or factor loadings represent the importance of each individual compound to each factor (only the primary three eigenvalues are reported here). Compounds with eigenvectors closer to one are more strongly related to a given factor, whereas values closer to zero show weak associations.

Table 13 (Continued). Principal Component Analysis Results for Upwind Concentrations at Six Truck Terminals with Repeat-Site Visits^a

Milwaukee, WI (41 Observations)				
Component	Eigenvalue	Difference	Proportion	Cumulative
1	9.23479	7.53026	0.6157	0.6157
2	1.70452	0.66138	0.1136	0.7293
3	1.04314	0.08008	0.0695	0.7988
4	0.96306	0.23954	0.0642	0.8630
Eigenvectors: Variable	1	2	3	
Trimethylpentane	0.31610	-0.07041	-0.01190	
Dimethylpentane	0.29324	-0.15527	-0.05706	
2-Methylhexane	0.31451	-0.09587	0.02867	
Methylpentane	0.29862	-0.22527	0.05575	
3-Methylhexane	0.31609	-0.05347	0.03297	
Methylcyclohexane	0.27616	0.08393	-0.19560	
Benzene	0.29981	-0.12261	0.13578	
Ethylbenzene	0.26641	0.37402	-0.08243	
<i>m&p</i> -Xylenes	0.29839	0.15380	-0.07171	
<i>o</i> -Xylene	0.29422	0.11832	-0.08677	
Styrene	0.05586	0.66751	-0.16334	
Toluene	0.29305	0.11518	0.08584	
Acetone	0.01958	0.25005	0.74578	
Acetaldehyde	0.12571	-0.42539	-0.11561	
Formaldehyde	0.09045	-0.07871	0.55841	
Phoenix, AZ (36 Observations)				
Component	Eigenvalue	Difference	Proportion	Cumulative
1	11.26329	9.34352	0.7509	0.7509
2	1.91977	1.22864	0.1280	0.8789
3	0.69113	0.31270	0.0461	0.9249
4	0.37843	0.09569	0.0252	0.9502
Eigenvectors: Variable	1	2	3	
Trimethylpentane	0.28558	-0.02203	-0.23326	
Dimethylpentane	0.28752	-0.09283	-0.20793	
2-Methylhexane	0.26749	-0.12707	-0.23521	
Methylpentane	0.24485	-0.00478	-0.39223	
3-Methylhexane	0.27090	-0.11620	-0.22928	
Methylcyclohexane	0.27959	-0.01475	-0.05949	
Benzene	0.28807	-0.02361	-0.04127	
Ethylbenzene	0.27549	-0.17208	0.30997	
<i>m&p</i> -Xylenes	0.27259	-0.13325	0.40123	
<i>o</i> -Xylene	0.26820	-0.20424	0.34675	
Styrene	0.27555	-0.03287	0.35122	
Toluene	0.28328	-0.07054	-0.14151	
Acetone	0.16989	0.51784	-0.20786	
Acetaldehyde	0.19567	0.50786	0.21358	
Formaldehyde	0.15580	0.58244	0.16081	

Table continues next page

^a Each factor has an eigenvalue (also called its latent root) associated with it that is related to the amount of variability it explains; these are ranked from the highest to the lowest. The eigenvectors or factor loadings represent the importance of each individual compound to each factor (only the primary three eigenvalues are reported here). Compounds with eigenvectors closer to one are more strongly related to a given factor, whereas values closer to zero show weak associations.

Table 13 (Continued). Principal Component Analysis Results for Upwind Concentrations at Six Truck Terminals with Repeat-Site Visits^a

Portland, OR (50 Observations)

Component	Eigenvalue	Difference	Proportion	Cumulative
1	9.14751	6.97432	0.6098	0.6098
2	2.17318	0.92761	0.1449	0.7547
3	1.24557	0.38155	0.0830	0.8378
4	0.86402	0.08821	0.0576	0.8954
Eigenvectors: Variable	1	2	3	
Trimethylpentane	0.31347	0.06429	-0.01009	
Dimethylpentane	0.28401	0.19744	-0.29654	
2-Methylhexane	0.31445	0.06256	-0.06918	
Methylpentane	0.26424	0.20242	-0.38733	
3-Methylhexane	0.31318	0.10114	0.01309	
Methylcyclohexane	0.24131	-0.13917	0.40271	
Benzene	0.29442	-0.01268	0.28767	
Ethylbenzene	0.31637	-0.14423	0.07562	
<i>m&p</i> -Xylenes	0.30686	-0.15987	-0.04215	
<i>o</i> -Xylene	0.30939	-0.12220	-0.05931	
Styrene	0.12952	-0.39503	0.29146	
Toluene	0.31573	0.01045	-0.10893	
Acetone	0.03383	0.39158	0.58616	
Acetaldehyde	0.03915	0.33411	0.24623	
Formaldehyde	0.01627	0.63232	0.00268	

Denver, CO (58 Observations)

Component	Eigenvalue	Difference	Proportion	Cumulative
1	6.89480	3.57083	0.4597	0.4597
2	3.32397	1.66795	0.2216	0.6813
3	1.65602	0.57094	0.1104	0.7917
4	1.08508	0.38902	0.0723	0.8640
Eigenvectors: Variable	1	2	3	
Trimethylpentane	0.30564	0.10609	0.02605	
Dimethylpentane	0.14512	0.47277	0.25268	
2-Methylhexane	0.09940	0.48869	0.25914	
Methylpentane	0.25088	0.21168	-0.27846	
3-Methylhexane	0.08364	0.48412	0.29259	
Methylcyclohexane	0.31545	-0.04321	-0.04290	
Benzene	0.35347	-0.03896	-0.10308	
Ethylbenzene	0.35215	-0.14362	0.02057	
<i>m&p</i> -Xylenes	0.35577	-0.08746	-0.12541	
<i>o</i> -Xylene	0.35677	-0.09249	-0.10090	
Styrene	0.18614	-0.11281	0.00024	
Toluene	0.35595	-0.00617	-0.10613	
Acetone	0.07476	-0.24213	0.43245	
Acetaldehyde	0.05974	-0.28182	0.56485	
Formaldehyde	0.18133	-0.23706	0.38606	

^a Each factor has an eigenvalue (also called its latent root) associated with it that is related to the amount of variability it explains; these are ranked from the highest to the lowest. The eigenvectors or factor loadings represent the importance of each individual compound to each factor (only the primary three eigenvalues are reported here). Compounds with eigenvectors closer to one are more strongly related to a given factor, whereas values closer to zero show weak associations.

Table 14. Molecular Markers of Personal Exposure Sample Averages by Job Title

Molecular Markers ($\mu\text{g}/\text{m}^3$)	Dockworker (n = 14)			Long Haul Drivers (n = 21)			P&D Drivers (n = 18)		
	Average ^a	Unc ^b	SD ^c	Average	Unc	SD	Average	Unc	SD
OC	13.00	1.02	2.92	24.05	1.66	8.75	17.98	1.30	6.80
EC (ng/m ³)	1.12	0.16	0.41	1.55	0.20	0.42	2.71	0.25	1.37
Benzo[b&k]fluoranthene	0.04	0.01	0.03	0.22	0.03	0.21	0.27	0.04	0.39
Benzo[e]pyrene	0.19	0.04	0.29	0.11	0.02	0.09	0.30	0.06	0.30
Benzo[a]pyrene	0.04	0.01	0.01	0.12	0.03	0.16	0.19	0.05	0.37
Indeno[1,2,3-cd]pyrene	0.08	0.02	0.10	0.07	0.02	0.03	0.11	0.03	0.11
Benzo[g,h,i]perylene	1.15	0.11	NA ^d	ND ^e			0.81	0.26	0.38
22,29,30-Trisomeohopane	0.20	0.03	0.13	0.48	0.07	0.39	0.31	0.04	0.15
17 α (H)-21 β (H)-29-norhopane	0.92	0.14	0.51	3.07	0.46	2.40	1.40	0.21	0.67
17 α (H)-21 β (H)-hopane	0.50	0.06	0.29	1.40	0.17	1.10	0.68	0.08	0.32
12S-17 α (H),21 β (H)-30-Homohopane	0.25	0.03	0.14	0.78	0.09	0.68	0.29	0.04	0.18
22R-17 α (H),21 β (H)-30-Homohopane	0.18	0.02	0.11	0.57	0.07	0.48	0.21	0.03	0.13
20R,5 α (H),14 β (H),17 β (H)-Cholestane	0.30	0.04	0.24	0.95	0.12	0.64	0.41	0.05	0.19
20S,5 α (H),14 β (H),17 β (H)-Cholestane	0.14	0.02	0.11	0.40	0.06	0.27	0.21	0.03	0.09
20R,abb-sitostane	0.18	0.02	0.16	0.55	0.07	0.41	0.22	0.03	0.14
20S,abb-sitostane	0.22	0.03	0.18	0.59	0.08	0.45	0.22	0.03	0.14
Tetracosane	4.90	0.46	3.44	6.84	0.64	4.16	11.27	1.05	6.15
Pentacosane	4.23	0.56	3.21	5.69	0.76	4.55	7.17	0.96	3.14
Hexacosane	2.16	0.33	2.24	3.25	0.53	1.73	2.93	0.44	2.20
Heptacosane	3.11	0.59	1.99	14.60	2.77	22.34	4.37	0.83	6.75
Octacosane	1.24	0.23	0.98	3.27	0.62	4.13	1.47	0.28	1.48
Nonacosane	3.00	0.44	1.70	14.40	2.09	25.73	4.55	0.66	8.29
Triacotane	1.20	0.17	0.67	6.70	0.97	10.14	2.22	0.32	3.55
Hentriacontane	7.04	1.21	6.47	46.90	8.05	84.24	16.19	2.78	37.10
Dotriacontane	1.51	0.26	0.89	9.82	1.70	18.10	3.11	0.54	5.56
Tritriacotane	3.70	0.85	3.77	30.01	6.87	54.86	8.02	1.84	18.66
Tetratriacotane	0.21	0.06	0.21	3.54	1.00	2.92	0.93	0.26	0.78
Pentatriacotane	0.03	0.01	0.02	0.24	0.09	0.17	0.07	0.02	0.05

Note: This table was adapted from Sheesley et al. 2009.

^a Average indicates average of all job or area samples.

^b Unc indicates average analytical uncertainty for organic speciation.

^c SD indicates standard deviation of all job/area samples.

^d NA indicates that standard deviation not available because there was only one value.

^e ND indicates not detected.

Table 15. Molecular Markers of Area Exposure Sample Averages by Location

Molecular Markers ($\mu\text{g}/\text{m}^3$)	Dock ($n = 14$)			Shop ($n = 8$)			Yard Upwind ($n = 6$)			Urban Background ($n = 9$)		
	Average ^a		Unc ^b	SD ^c	Average		Unc	SD	Average		Unc	SD
	OC	0.90	2.21	12.76	0.95	2.49	8.62	0.76	1.77	8.43	0.74	2.83
EC (ng/m^3)	1.18	0.15	0.38	1.97	0.18	0.82	1.19	0.15	0.38	0.94	0.14	0.34
Benzo[<i>b</i> & <i>k</i>]fluoranthene	0.06	0.01	0.19	0.03	0.21	ND ^d	0.01	0.10	0.07	0.01	0.05	0.05
Benzo[<i>e</i> pyrene	0.04	0.01	0.07	0.10	0.20	0.10	0.04	0.05	0.05	0.01	0.04	0.04
Benzo[<i>a</i> pyrene	0.03	0.01	0.10	0.07	0.02	0.10	ND	0.00	0.01	0.003	0.001	0.01
Indeno[1,2,3- <i>cd</i>]pyrene	0.08	0.02	0.21	0.10	0.02	0.10	0.00	0.00	0.01	0.03	0.01	0.05
Benzo[<i>g,h,i</i>]perylene	0.12	0.04	0.34	0.00	0.00	0.01	0.07	0.02	0.16	0.00	0.00	0.01
22,29,30-Trisnorneohopane	0.18	0.03	0.09	0.24	0.03	0.07	0.05	0.01	0.04	0.03	0.00	0.03
17 α (H)-21 β (H)-29-norhopane	0.84	0.13	0.27	0.92	0.14	0.21	0.57	0.09	0.31	0.30	0.04	0.14
17 α (H)-21 β (H)-Hopane	0.45	0.05	0.18	0.49	0.06	0.19	0.34	0.04	0.18	0.21	0.02	0.09
12S-17 α (H)-21 β (H)-30-Homohopane	0.23	0.03	0.11	0.24	0.03	0.13	0.19	0.02	0.12	0.09	0.01	0.06
22R-17 α (H)-21 β (H)-30-Homohopane	0.17	0.02	0.09	0.18	0.02	0.10	0.15	0.02	0.10	0.07	0.01	0.05
20R, 5 α (H),14 β (H),17 β (H)-Cholestan e	0.25	0.03	0.13	0.36	0.05	0.11	0.12	0.02	0.07	0.03	0.00	0.05
20S, 5 α (H),14 β (H),17 β (H)-Cholestan e	0.11	0.02	0.05	0.15	0.02	0.05	0.06	0.01	0.04	0.01	0.00	0.02
20R,abb-sitostane	0.15	0.02	0.06	0.16	0.02	0.06	0.08	0.01	0.07	0.06	0.01	0.04
20S,abb-sitostane	0.18	0.02	0.11	0.20	0.03	0.09	0.09	0.01	0.07	0.01	0.07	0.01
Tetracosane	5.31	0.49	3.08	7.86	0.73	2.82	1.44	0.13	1.05	1.54	0.14	1.31
Pentacosane	4.59	0.61	1.67	5.46	0.73	2.15	2.26	0.30	1.14	1.62	0.22	0.83
Hexacosane	2.29	0.35	1.67	2.09	0.32	0.85	0.98	0.15	0.63	0.48	0.07	0.50
Heptacosane	2.95	0.56	2.18	1.68	0.32	0.95	0.70	0.13	0.60	0.57	0.11	0.57
Octacosane	1.60	0.30	3.01	0.52	0.10	0.36	0.51	0.10	0.34	0.29	0.06	0.34
Nonacosane	2.80	0.41	2.84	1.36	0.20	0.92	0.54	0.08	0.21	0.47	0.07	0.49
Tricocene	1.28	0.19	1.92	0.65	0.09	0.89	0.08	0.01	0.07	0.22	0.03	0.27
Hentriacontane	6.05	1.04	6.79	3.36	0.56	2.42	0.64	0.11	0.63	0.45	0.08	0.52
Dotriacontane	0.97	0.17	0.90	0.66	0.11	0.82	0.05	0.01	0.07	0.17	0.03	0.22
Tririacotane	3.31	0.76	3.81	1.72	0.39	1.64	0.53	0.12	0.45	0.21	0.05	0.27
Tetraacotane	0.35	0.10	0.38	0.18	0.05	0.24	0.04	0.01	0.10	0.11	0.03	0.21
Pentraacotane	0.04	0.01	0.05	0.02	0.01	0.02	0.01	0.00	0.03	0.00	0.00	0.01

Note: This table was adapted from Sheesley et al. 2009.

a Average indicates average of all job or area samples.

b Unc indicates average analytical uncertainty for organic speciation.

c SD indicates standard deviation of all job/area samples.

d ND indicates not detected.

than the EC concentrations in the urban background (2.5-fold higher) and the yard background (twofold higher). Approximately 10% of the EC was from nondiesel sources. The OC concentrations that the drivers were exposed to were about twofold higher than either the OC concentrations in the urban or yard backgrounds, but more than half of the OC was unapportioned. It is important to note that, for the P&D drivers, concentrations of polycyclic aromatic hydrocarbons were consistently higher than those for the upwind yard and urban backgrounds and those for workers in other jobs, indicating a source specific to these drivers, such as something in traffic exposures. The concentrations for the upwind yard background were higher than those for urban background measured at a local Supersite monitoring station run by the EPA, which might reflect the industrial park location of the truck terminal.

BTEX Ratios

The ratios of benzene, toluene, ethylbenzene, and xylenes (BTEX) can be useful in identifying sources of aromatic

hydrocarbon emissions. The BTEX ratios observed in the monitoring locations in this study are shown in Table 16. The median concentration values were used to construct the ratios presented in the table to reduce the influence of a small number of extreme outliers. Table 17 shows BTEX ratios from other studies of similar exposure settings, including settings near major roadways, in vehicles (cars and buses), and at bus stations. One factor affecting this comparison is that the benzene content of gasoline ("petrol") in the United Kingdom and Europe is approximately twice that in the United States (~ 4% benzene by volume versus ~ 1%–2% by volume, respectively). There might be other systematic differences in the toluene, xylene, and ethylbenzene content of gasoline that also affected these comparisons.

The upwind background BTEX ratios observed at the truck terminals were similar to those observed along major roadways in comparable studies, because truck terminals are located near major roadways, and concentrations therefore share a common traffic source. The strong

Table 16. BTEX Ratios from the Current Study

Ratios	Yard Upwind	P&D Driver (Nonsmokers)	Mechanic Shop	Loading Dock
Toluene:Benzene	3.3	3.1	4.0	4.0
Toluene:Xylene	1.5	1.2	0.3	2.7
Toluene:Ethylbenzene	6.9	5.3	1.6	10.0
Benzene:Xylene	0.6	0.4	0.1	0.4
Benzene:Ethylbenzene	2.5	1.7	0.4	1.6
Xylene:Ethylbenzene	4.6	4.5	4.7	3.7

Table 17. Comparison of BTEX Ratios from Other Studies

Ratios	U.S. Roadways ^a	U.K. Roadways ^b	U.K. Cars and Buses (In-Vehicle) ^b	Spanish Public Buses (In-Vehicle) ^c	U.K. Bus Station ^b
Toluene:Benzene	[~1.6–1.9]	2.2	2.4–3.4	[~3.4–2.5]	2.4
Toluene:Xylene	[~0.9]	1.9	1.9–2.1	[~0.5]	2.8
Toluene:Ethylbenzene	3.6–5.6	8.7	8.7–9.5	5.7–6.3	12.4
Benzene:Xylene	[~0.5]	0.9	0.6–0.9	[~0.6]	1.2
Benzene:Ethylbenzene	2.3–2.9	4.0	2.5–3.9	1.7–2.5	5.3
Xylene:Ethylbenzene	4.3–6.0	4.6	4.5–4.6	2.6–3.1	4.5

Note: Values in brackets were calculated from other ratio values in the Table, such as Toluene:Benzene = Toluene:Ethylbenzene divided by Benzene:Ethylbenzene. These are only approximate because contributors to the ratios may not be the same in the numerator and denominator.

^a Muhamed et al. 2002.

^b Kim et al. 2001.

^c Parra et al. 2008.

comparability across studies suggested that emissions from motor vehicles were the dominant source of hydrocarbons in the ambient air across locations. BTEX ratios for non-smoking P&D driver were comparable to those from similar studies of in-vehicle bus and car exposures, with differences potentially attributable to unknown concentrations of tobacco smoke in the comparison studies. Although there were no directly comparable data available to match the ratios from the specific indoor work environments from our trucking study (i.e., the mechanic shop and loading dock), rough comparisons were made using the reported ratios in bus stations in the United Kingdom (Kim et al. 2001). Although some of the ratios were similar, the unique mix of fuel sources (e.g., propane and diesel emissions present on the loading dock) in the trucking settings and ventilation characteristics made a direct comparison difficult.

Finally, the concentration ratio of toluene to benzene can be used to predict proximity and intensity of traffic source emissions (Gelencser et al. 1997) because toluene is more reactive in the open air and decays at a much faster rate than benzene as distance from the mobile source increases. Table 17 shows the ratios of BTEX components for several sites from various studies. A toluene–benzene

ratio of approximately 2:1 was found at sampling locations near traffic sources; the ratio trended toward one as distance increased. Our results showed higher ratios than those found in other studies of U.S. roadways (at the roadside, not for drivers in traffic), suggesting a pattern of intense proximate traffic sources. The ratios were 3.3 for the terminal upwind background and 3.1 for the non-smoking drivers. The highest ratios were observed in the indoor work areas (4.0 in the mechanic shop and 4.0 in the loading dock), likely attributable to the lack of photochemical destruction, intensity of indoor sources, evaporating fuel in the shop, propane-powered forklifts on the dock, and lower ventilation rates. The values outdoors compared well with those found in other studies (Table 17).

Structural Equation Modeling for Time-Weighted Average VOCs

Table 18 shows the structural equation modeling results for a representative set of four VOCs: benzene, 1,3-butadiene, toluene, and formaldehyde. These four VOCs were chosen for a priori interest (1,3-butadiene, benzene, and formaldehyde are carcinogens) or because they accounted for the majority of their class of compounds (toluene and

Table 18. Structural Equation Modeling Regression Results for Four TWA VOCs

	Benzene (n = 76)	1,3-Butadiene (n = 75)	Toluene (n = 75)	Formaldehyde (n = 70)
Equation 1: $\ln(\text{WorkAreaConc}_{ij}) = \beta_{10} + \beta_{11}\ln(\text{YardConc}_{ij}) + \beta_{12}\text{JobLoc}_{ij} + \epsilon_{ij}$				
Yard upwind	0.79 ^a	0.48 ^a	0.46	0.35
Job	0.53 ^a	0.99 ^a	1.19 ^a	0.99 ^a
Constant	– 0.26	– 0.81 ^a	0.34	1.55
Root mean square error	0.63	0.71	1.22	0.84
R ²	0.38	0.25	0.20	0.18
Equation 2: $\ln(\text{YardConc}_{ij}) = \beta_{21}(\text{Temp}_{ij}) + \beta_{22}(\text{Windspeed}_{ij}) + \beta_{23}(\text{RoadDist}) + \beta_{24-6}(\text{RegDummy}_{ij}) + \eta_{ij}$				
Temperature	0.01	– 0.05 ^a ($\downarrow 41\%$)	0.01	0.02 ^a ($\uparrow 23\%$)
Windspeed (mph)	– 0.09 ^a	– 0.09 ^a	– 0.15 ^a	– 0.04 ^a
Interstate distance ^b	– 0.0004 ^a	0.0002	– 0.0003 ^a	– 0.0001
Region 1 (midwest)	0.99 ^a	– 0.05	2.15 ^a	1.22 ^a
Region 2 (northeast)	1.03 ^a	– 1.04 ^a	2.07 ^a	1.37 ^a
Region 3 (west)	1.45 ^a	0.06	2.78 ^a	1.08 ^a
Constant ^c	NA ^d	NA	NA	NA
Root mean square error	0.40	0.68	0.53	0.37
R ²	0.45	0.83	0.80	0.90

^a Indicates significant at 5% level.

^b Because no sites were < 500 m from an interstate, the variable represents continuous distance to an interstate in meters.

^c The constant has been dropped from Equation 2 because all three regional dummy variables are included in the model. This does not affect the coefficients for the other variables, only the interpretation of the regional variables (hypothetical null used as baseline).

^d NA indicates not available.

formaldehyde). High upwind background concentrations significantly elevated work area exposures for benzene and 1,3-butadiene; the model predicted elevated exposures in the loading dock over the mechanic shop for all exposures. Higher ambient temperatures significantly increased expected ambient concentrations of formaldehyde with the opposite effect for 1,3-butadiene. Wind speed was significantly negative (higher wind speed predicts lower levels) for all concentrations. Formaldehyde was a component of some vehicle emissions; it is also formed in the atmosphere, and its concentration increases with temperature. In contrast, 1,3-butadiene is removed from the atmosphere by atmospheric reactions that increase with temperature. The distance to a major road was only significant for toluene and benzene; but was observed also for formaldehyde with a negative correlation. The census region dummy variables (regions 1–3 in the table) provided evidence of significant regional variability across the United States, with significantly higher exposures to benzene observed in the West (region 3) and significantly higher exposures to formaldehyde observed in the Northeast (region 2). No site visits were conducted in the South, the fourth census region.

PHASE 1 FINDINGS — DRIVER HOT SPOTS

To study the third potential hot-spot location, the cabs of P&D trucks while driving in traffic, we measured VOCs and PM using integrated samplers. Measurements of VOCs and PM in truck cabs were assumed to reflect exposures of the drivers. P&D drivers typically drive tractors towing trailers and smaller single-bodied trucks in cities and in suburban and metropolitan areas. They work primarily during the day. They are exposed to a range of driving conditions, including rush-hour stop-and-go traffic in the city, heavy traffic on urban highways, and lighter traffic on suburban roads and highways. Driving conditions also include the processing of freight P&D orders around the terminal location (typically within a ~50-kilometer radius). We attempted to sample only nonsmoking drivers. More than 70% of the samples obtained from drivers were from nonsmokers.

Driver Summary Statistics

Summary statistics for PM and VOCs sampled in truck cabs of P&D drivers according to smoking status are shown in Table 19. Compared with the measured on-site terminal concentrations for upwind yard background and indoor work locations, concentrations for nonsmoking drivers were higher on average for benzene, MTBE, styrene, and hexane. Furthermore, the measured PM concentrations were much lower than those reported in an earlier study of

the industry done in the 1980s (Zaebst et al. 1991) that used an identical sampling protocol and analytic methods to collect EC and PM_{2.5} at six large U.S. truck terminals. During the earlier time period of the study by Zaebst and colleagues, in-cab EC concentrations were much higher. For P&D drivers the geometric mean (GM) was 4.0 µg/m³ and the geometric SD (GSD) 2.0; for long-haul (LH) drivers the GM was 3.8 µg/m³ and the GSD 2.3 compared with the concentrations observed in our study (P&D drivers GM = 1.2 µg/m³ and GSD = 2.8; LH drivers GM = 1.1 µg/m³ and GSD = 2.3) (not shown in table).

Because we made a concerted effort to sample non-smoking drivers, the sample size for smokers was relatively small. Different components of the VOCs were present in the two groups (Table 19). Most alkane concentrations were slightly higher or the same for nonsmokers versus smokers. As expected, 1,3-butadiene concentrations were significantly higher for the smokers. For aromatics, the concentrations for smokers tended to be slightly higher. Also as expected, MTBE concentrations were very low and about the same for both smokers and nonsmokers. Acetone concentrations were lower for smokers, but concentrations of aldehydes were somewhat higher, as expected. PM_{2.5} concentrations detected from smoking drivers were significantly higher than those observed for nonsmoking drivers, but the difference was small for EC. Because cigarette smoke has a large amount of OC there was a large difference in OC for smoking drivers.

Relationships Between PM_{2.5} and VOCs

Table 20 lists the correlations of TWA VOCs with EC and PM_{2.5} for nonsmoking drivers. EC and PM_{2.5} correlations with VOC components were relatively low, with the exception of those for a few aromatics, primarily benzene ($r = 0.4\text{--}0.5$). The pattern of correlations was consistent within chemical groupings associated with traffic emissions, with a few exceptions. Although the lower-molecular-weight alkanes generally tracked with the EC and PM_{2.5}, hexane did not, which suggested a different source. Similarly, the aromatics showed consistent associations with EC and PM_{2.5}. 1,3-Butadiene was somewhat less correlated with EC and PM_{2.5}. The aldehydes and acetone were not related to EC or PM_{2.5}. However, MTBE was associated with PM_{2.5}, which might reflect an association with car emissions because there is no MTBE in diesel fuel. At the time of the study, MTBE was being phased out in the United States; it was only detected in some of the samples from areas where MTBE was still being used. The aromatics and alkanes, except for hexane, had consistent and modest correlations with EC and lower and less consistent correlations with PM_{2.5}. Benzene and MTBE were more

Table 19. VOC Summary Values for P & D Drivers

Compound ($\mu\text{g}/\text{m}^3$)	Nonsmokers			Smokers				
	Observations (n)	Mean	Median	SD	Observations (n)	Mean	Median	SD
Trimethylpentane	235	1.22	0.72	2.29	62	0.83	0.51	0.99
Dimethylpentane	234	0.68	0.35	1.22	62	0.64	0.32	1.04
2-Methylhexane	234	1.15	0.67	2.65	61	1.21	0.59	2.04
Methylpentane	234	2.33	1.58	4.40	61	1.83	1.39	1.57
3-Methylhexane	234	1.46	0.79	3.74	61	1.60	0.68	2.95
Methylcyclohexane ^a	235	0.78	0.58	0.81	62	7.80	0.62	50.08
Hexane	64	2.24	1.66	2.07	18	1.53	1.19	0.85
1,3-Butadiene ^a	235	0.34	0.28	0.47	62	1.26	0.80	1.50
Benzene ^a	235	1.56	1.43	0.78	62	2.36	1.84	1.76
Ethylbenzene	235	6.91	0.85	84.36	62	2.74	0.95	11.41
<i>m&p</i> -Xylenes	235	24.16	2.79	301.40	62	7.59	3.14	27.03
<i>o</i> -Xylene	235	9.56	1.00	123.53	62	3.22	1.05	14.66
Styrene ^a	235	8.63	0.55	112.13	62	1.46	0.66	2.39
Toluene	235	5.83	4.53	4.27	62	6.66	4.89	6.99
Toluene:Benzene	235	3.7	3.2	NA	62	2.8	2.7	NA
MTBE	235	0.61	0.04	1.36	61	0.51	0.07	0.93
Acetone	234	10.30	7.58	18.97	62	6.92	5.73	7.16
Acetaldehyde ^a	234	5.63	4.63	4.23	62	8.78	6.07	7.98
Formaldehyde	234	8.30	7.11	5.67	62	9.59	8.16	5.65
EC ^a	223	1.23	1.05	0.87	61	2.00	1.50	1.83
OC ^a	223	11.79	10.66	5.46	61	27.65	20.71	30.90
PM _{2.5} ^a	207	15.98	13.88	10.22	55	37.83	29.48	29.50

^a Concentrations were significantly higher for smokers than nonsmokers ($P < 0.05$) using Wilcoxon rank sum nonparametric comparison tests.

NA indicates not available.

highly correlated with PM_{2.5}. The pattern for the aldehydes anticipated the findings of the principal components analysis (below), which found that aldehydes and PM measures tended to represent different sources, as was seen for the upwind and downwind yard samples.

Although the Phase 1 analysis of driver exposures was limited by the issues described earlier, a number of important trends were identified during the exploratory analyses of the first 36 sampling visits in the NCI study using univariate methods and pairwise comparisons. Median driver exposures were significantly higher for smokers than for nonsmokers for PM and certain VOCs, including methylcyclohexane, 1,3-butadiene, benzene, styrene, and acetaldehyde ($P < 0.01$). Concentrations observed in the truck cabs of nonsmoking drivers (Table 21) were significantly correlated (Spearman $r = 0.4$ – 0.6 ; $r = 0.9$ for MTBE; $P < 0.01$) with yard upwind concentrations measured at the driver's home terminal during that sampling session.

When the windows of the truck were predicted to be open (based on in-cab CO₂ concentrations), concentrations of PM and 1,3-butadiene were observed to be significantly higher than when the windows were closed, based on Wilcoxon rank sum tests ($P < 0.05$). Open windows had an opposite effect on the concentrations of aldehydes, which were significantly lower than when the window was closed (data not shown). The effects of whether the windows were open or not on the concentrations of other compounds were not statistically significant. The results provided evidence of an external source for PM and 1,3-butadiene and an internal source for aldehydes.

Weather data (i.e., wind speed, relative humidity, and temperature) were collected from the closest monitoring station to the main terminal and matched to the sampling time periods using an online source (Weather Underground, available at www.wunderground.com). Average wind speed was significantly related to EC and VOCs

Table 20. Correlations^a of TWA VOCs with EC and PM_{2.5} for Nonsmoking P & D Drivers

Compound	EC	PM _{2.5}
Trimethylpentane	0.29 ^b	0.18 ^b
Dimethylpentane	0.30 ^b	0.18 ^b
2-Methylhexane	0.27 ^b	0.23 ^b
Methylpentane	0.21 ^b	0.17 ^b
3-Methylhexane	0.28 ^b	0.23 ^b
Methylcyclohexane	0.30 ^b	0.34 ^b
Hexane	-0.01	0.01
1,3-Butadiene	0.15 ^b	0.22 ^b
Benzene	0.42 ^b	0.47 ^b
Ethylbenzene	0.38 ^b	0.35 ^b
<i>m&p</i> -Xylenes	0.37 ^b	0.37 ^b
<i>o</i> -Xylene	0.42 ^b	0.34 ^b
Styrene	0.23 ^b	0.17 ^b
Toluene	0.31 ^b	0.23 ^b
MTBE	0.12	0.45 ^b
Acetone	0.07	-0.18
Acetaldehyde	0.01	-0.10
Formaldehyde	0.18 ^b	-0.09

^a Nonparametric Spearman correlation coefficients.^b Significant at the 5% level.**Table 21.** Correlations of Nonsmoking P & D Driver Exposure Concentrations with Terminal Yard Upwind Concentrations^a

Compound	Correlation Coefficient
Trimethylpentane	0.6
Dimethylpentane	0.6
2-Methylhexane	0.5
Methylpentane	0.5
3-Methylhexane	0.5
Methylcyclohexane	0.4
Hexane	0.4
1,3-Butadiene	0.4
Benzene	0.5
Ethylbenzene	0.5
<i>m&p</i> -Xylenes	0.5
<i>o</i> -Xylene	0.5
Styrene	0.4
Toluene	0.5
MTBE	0.9
Acetone	0.5
Acetaldehyde	0.5
Formaldehyde	0.4
PM _{2.5}	0.5
EC	0.5

^a All Spearman correlations were significant at $P < 0.01$.

(higher wind speeds were associated with lower in-cab concentrations; Spearman $r = 0.1\text{--}0.5$, $P < 0.05$) but not PM_{2.5} and aldehydes. Temperature was significantly positively related to in-cab PM and VOC concentrations (higher temperatures were associated with higher in-cab concentrations; Spearman $r = 0.2\text{--}0.3$, $P < 0.05$), with the exception of 1,3-butadiene, which was significantly negatively correlated ($r = 0.3$, $P < 0.01$). 1,3-Butadiene is reduced by atmospheric reactions, so an inverse correlation with temperature would be expected with an outdoor source. Relative humidity had a much weaker relationship with in-cab concentrations; it was only significantly (negatively) correlated with a few compounds (2,3-dimethylpentane, acetone, and acetaldehyde).

For some of the trucks sampled, vehicle characteristics were obtained, including production year, make, and model. Make and model did not have a significant effect on in-cab particle concentrations, but EC concentrations increased significantly with truck age ($R^2 = 0.34$; $P < 0.01$). This relationship is shown in Figure 5 for P&D drivers with the group median EC value graphed by production year (group medians are more representative of the central tendency because

of the non-normality of exposure data). The results for VOCs were more mixed, with higher concentrations of styrene, toluene, and MTBE observed in the newer models ($r = 0.1\text{--}0.3$, $P < 0.01$) in contrast to lower concentrations of formaldehyde ($r = 0.2$, $P < 0.01$) (data not shown).

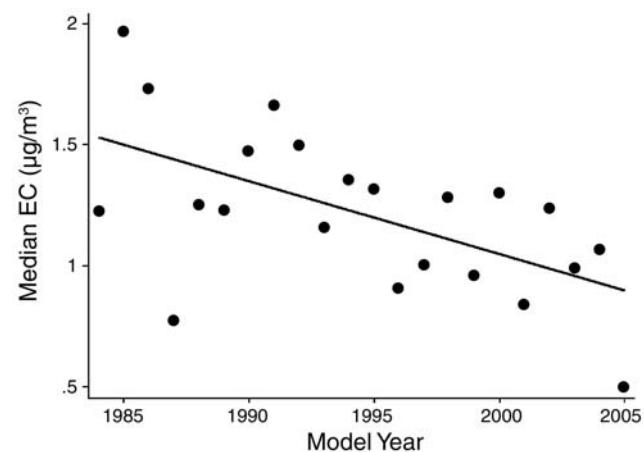


Figure 5. P&D drivers' median EC exposure by truck model year.

PHASE 2 FINDINGS — REAL-TIME DATA ON UPWIND AND DOWNWIND EXPOSURES

After extensive data management and quality control efforts, we constructed a combined database of the real-time data, the integrated data, and weather station data in a single format matched by time. We performed a thorough descriptive analysis of each monitoring session, which included more than 50 12-hour sessions at six terminals. As part of this descriptive work, potential emission sources at each location around the terminal were identified using satellite images, telephone directories, and the local knowledge of terminal workers. GIS mapping software was used to superimpose maps on these images. Wind roses were constructed and overlaid on the maps for each sampling session to

determine the potential sources for wind transport during these time periods. To the extent possible, a time profile of traffic activity around each location was also constructed (using, for example, traffic data and terminal freight logs). Averages of real-time data were matched by time to TWA integrated data to test for consistency across the two sampling methods. These maps have not been reproduced here, because of a confidentiality agreement with the monitored terminals and Google's new policy on publication of its maps. An example of the real-time trends observed for total VOCs and PM_{2.5} across four yard sampling locations is shown in Figure 6. Overall, the analysis of the sessions provided evidence of the influence of emissions from high traffic periods at the terminal on downwind sites.

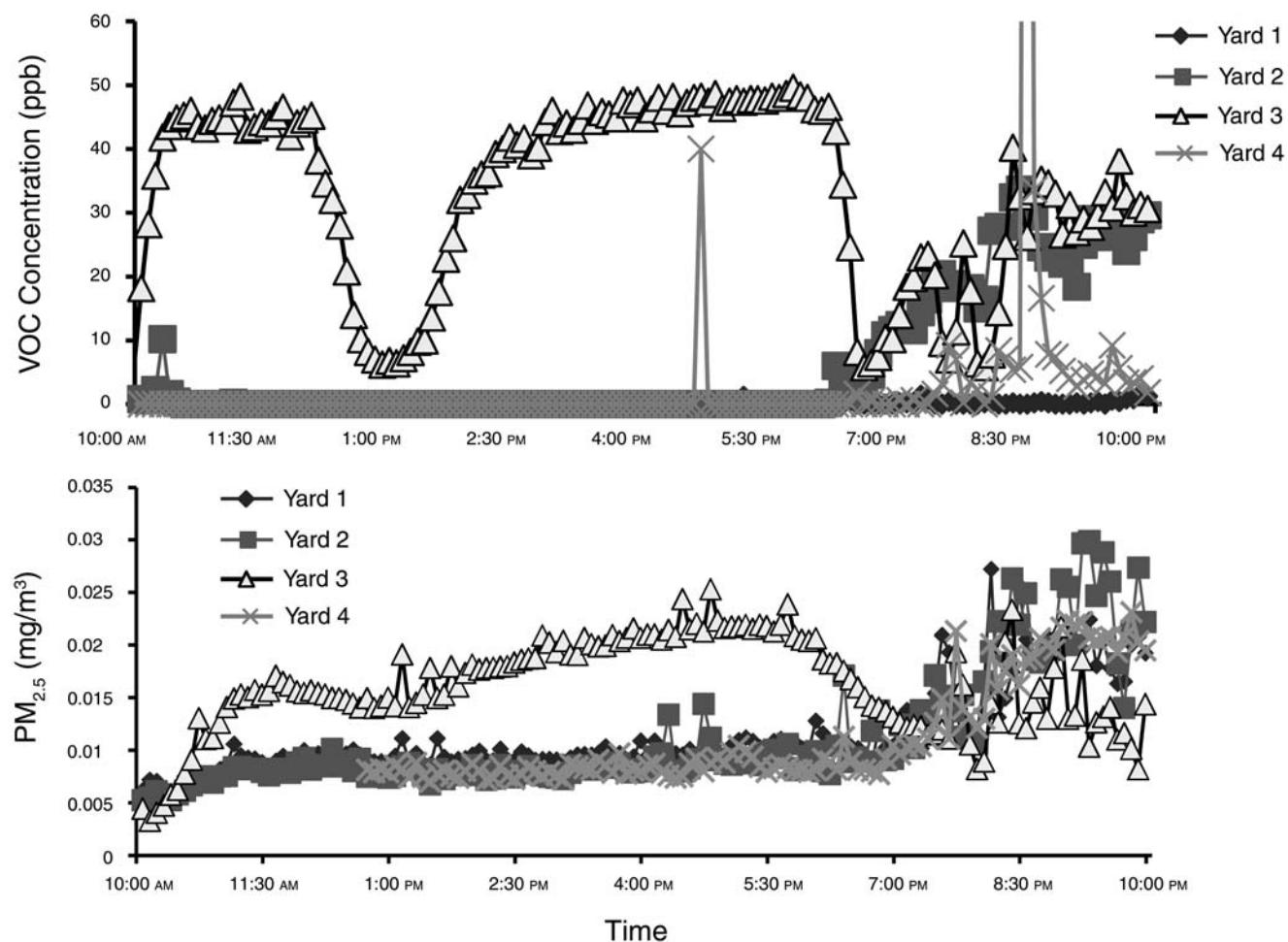


Figure 6. Example of temporal patterns across sampling stations for (top) VOCs and (bottom) PM_{2.5}.

This level of detailed analysis by session was necessary to accurately characterize the terminal contribution to background exposures in the context of our two hypothesized hot spot locations, industrial areas upwind of terminals and neighborhoods downwind of terminals. Our results from the real-time measurements suggested that VOCs and PM concentrations were most elevated when wind direction and speed favored transport without much dilution (i.e., ~ 10 m/sec). Although the terminal contribution to background exposures at the fence line varied considerably across the sampling locations by wind and local source characteristics, we observed a terminal contribution to downwind exposures during approximately 70% of the total sessions and a significant upwind-to-downwind differential during nearly 60% of the total sessions. Because the terminal emissions were incompletely mixed, these differences were not maintained as steady exposures throughout a session; instead, exposure varied with changes in the source operations and wind parameters. This variation is illustrated for a representative session in Figure 7. Histograms of the real-time VOC and PM_{2.5} terminal contribution (downwind minus upwind) for a representative session, where a consistent differential was present throughout the session, are shown.

We are continuing our work on this type of analysis to provide a more quantitative description of the terminal's contributions to the neighborhood exposures in subsequent published articles.

PHASE 2 FINDINGS — REAL-TIME DATA FROM DRIVERS' SAMPLES

The addition of GPS tracking to the protocol for collecting the P&D truck samples provided a tremendous

amount of additional information. GPS tracking captured truck speed and location every minute during a driver's work shift. This information was entered into the GIS database. As a result, we could track a driver's route and match the PM_{2.5} and total VOC real-time data to specific locations and their characteristics. Figure 8 and Figure 9 show representative examples of a driver's route and the associated exposures. Figure 9 also shows the time profile for VOCs during a driver's work shift. The upward curve in Figure 9 at the end of the VOC profile (denoting the end of the trip) was common. It corresponded to the time after the truck was parked at the terminal. This was seen in many of the time profiles. It appeared to represent the accumulation of in-cab emissions when the vehicle stopped moving and ventilation stopped as well.

The link between location and the time profile that GPS tracking made possible allowed us to identify where the exposures spiked. Interestingly, the brief VOC spikes that represent the top 5% of values tended to occur when the truck was in a terminal, stopped at delivery sites, or stopped in traffic — not while the truck was moving. In most cases, these spikes did not make a major contribution to the overall average, because they were too brief. Also, VOC values decreased with increasing truck speed and increased with increasing road traffic.

Much smaller effects in the opposite direction were seen for PM_{2.5}. Figure 8 shows that two individual PM_{2.5} peaks occurred along highways at higher speeds, perhaps associated with following other vehicles or trucks. Sometimes they were associated with industrial land uses. Neither PM_{2.5} nor total VOC (Figure 9) time profiles showed clear evidence of rush-hour effects — the trucking companies try whenever possible to route deliveries and pickups to

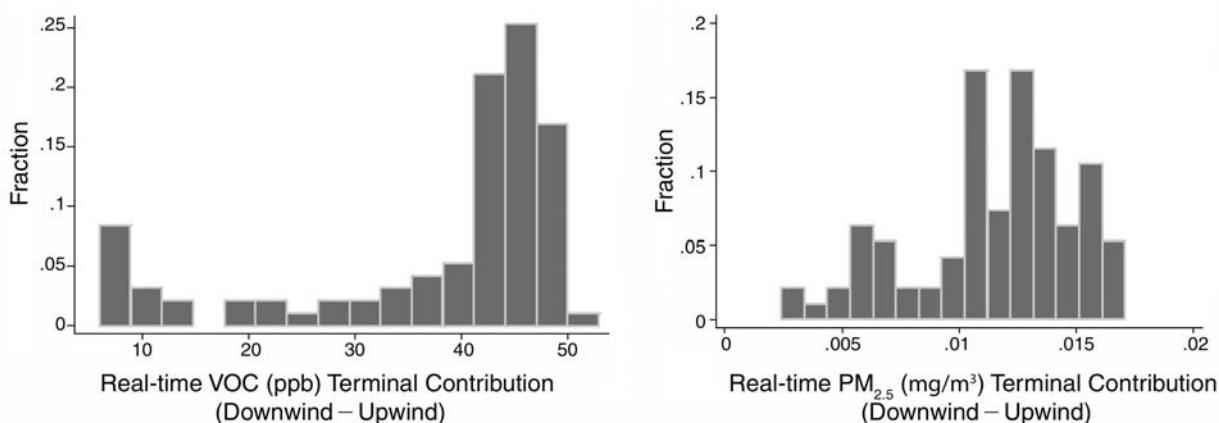


Figure 7. Example of real-time data on downwind minus upwind differences for (left) VOCs and (right) PM_{2.5}. Fraction indicates the number of observations in the bin divided by the number of the total observations.

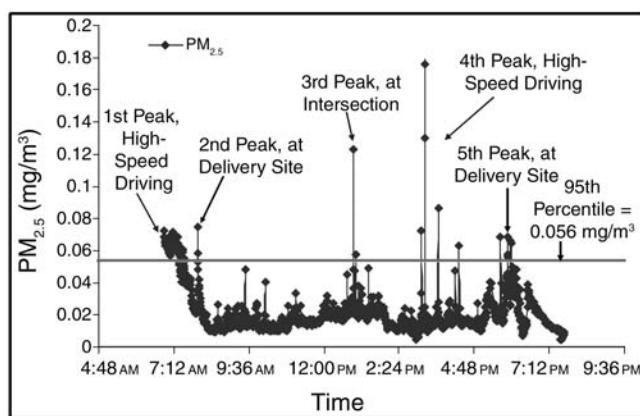
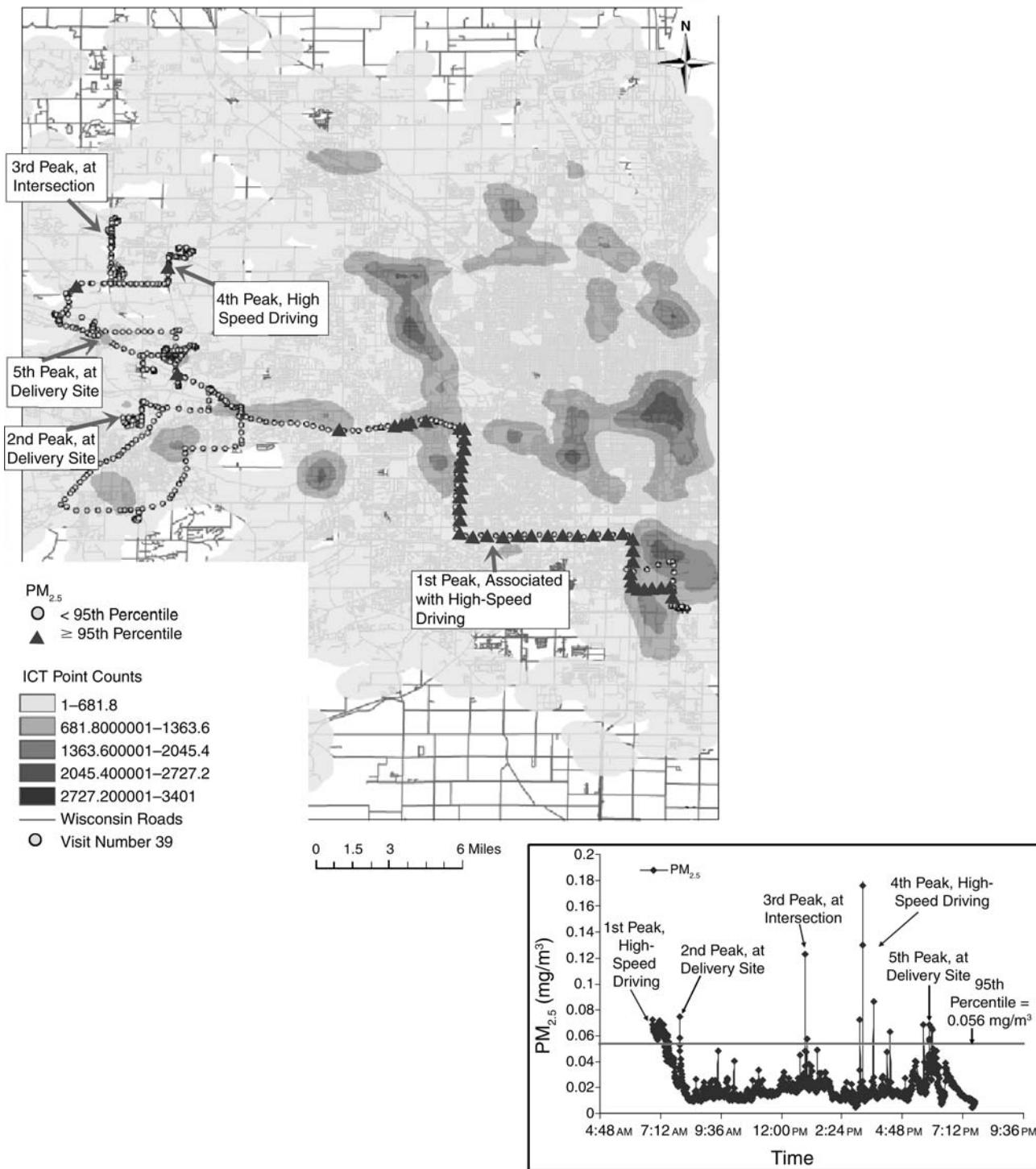


Figure 8. (Top) GIS map of a P&D driver's trip in Milwaukee, Wisconsin (Visit Number 39), showing PM_{2.5} exposures over time. Exposures exceeding the 95th percentile are indicated by triangles. ICT values shown are point counts. Higher point counts (darker shades) indicate more industrial land use. **(Bottom)** PM_{2.5} time trace for a P&D driver over a full shift.

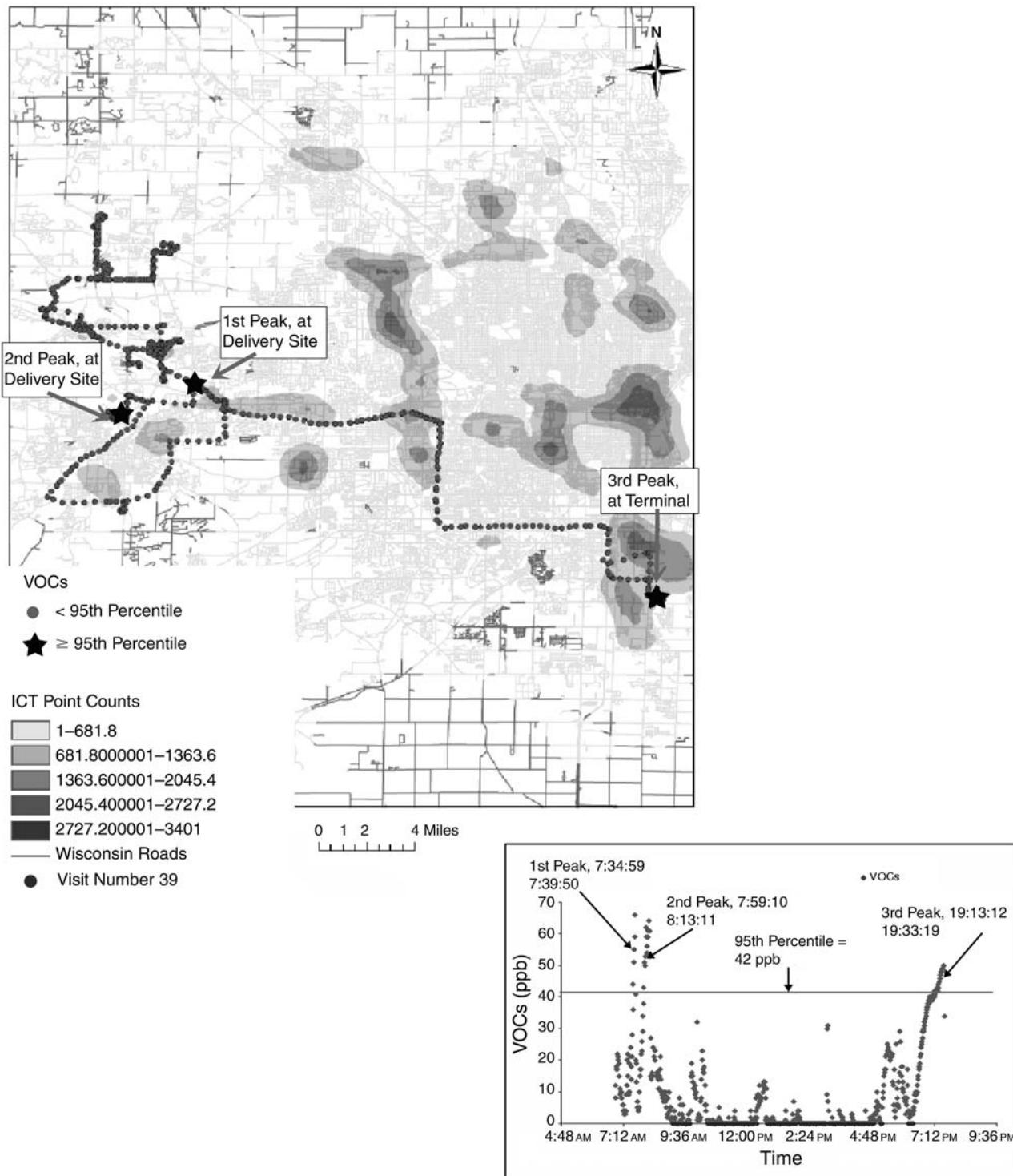


Figure 9. (Top) GIS map of a P&D driver's trip in Milwaukee, Wisconsin (Visit Number 39), showing VOC exposures over time. Exposures exceeding the 95th percentile are indicated by stars. ICT values shown are point counts. Higher point counts (darker shades) indicate more industrial land use. (Bottom) VOC time trace for a P&D driver over a full shift.

minimize passage through areas prone to stop-and-go traffic.

The analysis of the drivers' data is still ongoing. As shown for PM_{2.5} and total VOCs in Figures 8 and 9, we have developed a set of overlays and ways of linking the timed measurements of exposure and vehicle speed so they can be displayed on the area map with roads, population, and industrial-use densities. We are exploring analytic strategies to allow us to move beyond descriptive analyses.

REPEAT-VISIT ANALYSIS

Table 22 and Table 23 show the concentrations of PM for loading docks and of VOCs and PM for yards (upwind yard background) for two repeat visits to terminal locations in Phase 2 of our study. Overall, concentrations were relatively stable across the repeat visits, generally within

± 30%, with few significant differences. Background concentrations observed in the yard tended to be higher during the second visit (with the exception of a few VOCs that had significantly lower concentrations); these concentrations reflected area-wide background differences.

The structural equation model developed to predict upwind yard values of TWA VOCs (Table 18) was tested against both time periods. Where significant differences in background concentrations existed between the first and second visits, they could be attributed in large part to changes in the set of exogenous variables used to predict concentrations in the exposure model. Table 24 shows summary statistics (with significant differences between visits indicated) for the predictor variables of weather during both visits. Although both repeat site visits were made during the same time of year in order to limit the impact of weather and seasonality, weather differences

Table 22. Summary Statistics for EC and PM_{2.5} Measurements at Loading Docks

Location / Compound	First Visit			Second Visit			Significant Difference ^a and Direction
	Observations (n)	GM (µg/m ³)	GSD (µg/m ³)	Observations (n)	GM (µg/m ³)	GSD (µg/m ³)	
Philadelphia							
EC	20	0.7	1.7	10	1.1	1.8	
PM _{2.5}	20	12.7	2.0	10	18.9	1.8	
Columbus							
EC	23	0.8	1.5	9	0.9	2.2	
PM _{2.5}	23	12.8	1.7	9	16.5	1.6	
Milwaukee							
EC	24	0.2	2.1	17	0.4	1.6	+ ^a
PM _{2.5}	24	8.8	1.9	17	11.1	1.4	
Phoenix							
EC	6	1.2	1.9	9	1.7	1.4	
PM _{2.5}	6	13.1	1.5	9	17.1	1.5	
Portland							
EC	18	0.6	1.3	10	0.8	1.5	
PM _{2.5}	18	6.5	1.4	9	6.8	1.3	
Denver							
EC	23	0.7	1.7	12	1.6	1.4	+ ^a
PM _{2.5}	23	10.6	1.5	12	8.4	1.3	

^a Significant differences ($P < 0.05$) in median values using Wilcoxon rank sum non-parametric comparison test. + indicates increase in GM.

Note: VOCs were not measured in the shop during the initial site visits.

Table 23. Summary Statistics for VOC and PM Measurements in Yard Upwind Locations

Location / Compound	First Visit			Second Visit			Significant Difference ^a and Direction
	Observations (n)	GM ($\mu\text{g}/\text{m}^3$)	GSD ($\mu\text{g}/\text{m}^3$)	Observations (n)	GM ($\mu\text{g}/\text{m}^3$)	GSD ($\mu\text{g}/\text{m}^3$)	
Philadelphia							
EC	24	0.5	1.9	38	0.8	2.1	
PM _{2.5}	24	10.7	1.5	38	13.9	1.8	+ ^a
Benzene	26	1.3	1.5	36	0.9	1.5	- ^a
Toluene	26	2.0	1.6	36	2.2	2.3	
Ethylbenzene	26	0.3	1.6	36	0.2	2.1	
<i>m&p</i> -Xylenes	26	1.1	1.6	36	0.8	2.2	- ^a
<i>o</i> -Xylene	26	0.4	1.6	36	0.3	2.1	
Formaldehyde	29	2.0	5.0	34	3.3	1.6	
Columbus							
EC	20	0.6	1.7	24	1.1	2.3	+ ^a
PM _{2.5}	22	9.1	2.2	23	12.5	1.6	
Benzene	13	0.9	3.5	22	1.0	2.3	
Toluene	13	2.6	6.0	22	2.4	4.3	
Ethylbenzene	13	0.5	5.9	22	0.4	3.9	
<i>m&p</i> -Xylenes	13	1.5	6.4	22	1.1	4.3	
<i>o</i> -Xylene	13	0.6	6.6	22	0.4	4.3	
Formaldehyde	12	0.9	11.3	20	3.1	1.5	
Milwaukee							
EC	29	0.1	3.4	31	0.2	5.6	+ ^a
PM _{2.5}	27	3.0	2.5	32	8.5	1.7	+ ^a
Benzene	28	0.6	1.9	28	0.6	1.5	
Toluene	28	0.8	3.0	28	1.5	1.9	+ ^a
Ethylbenzene	28	0.1	5.6	28	0.2	1.9	+ ^a
<i>m&p</i> -Xylenes	28	0.4	4.8	28	0.7	2.0	
<i>o</i> -Xylene	28	0.1	5.1	28	0.2	1.9	
Formaldehyde	20	3.0	1.9	21	1.9	4.0	
Phoenix							
EC	6	0.8	2.1	36	1.2	1.7	
PM _{2.5}	3	15.3	1.4	36	16.5	1.4	
Benzene	6	0.9	1.5	30	0.9	1.7	
Toluene	6	3.7	1.5	30	4.0	1.7	
Ethylbenzene	6	0.6	1.5	30	0.6	2.0	
<i>m&p</i> -Xylenes	6	2.3	1.5	30	1.8	2.1	
<i>o</i> -Xylene	6	0.3	13.7	30	0.7	2.0	
Formaldehyde	6	7.9	1.4	32	3.1	2.8	- ^a

Table continues next page

^a Significant differences ($P < 0.05$) in median values using Wilcoxon rank sum non-parametric comparison test. + indicates increase in GM; - indicates decrease in GM.

Table 23 (Continued). Summary Statistics for VOC and PM Measurements in Yard Upwind Locations

Location / Compound	First Visit			Second Visit			Significant Difference ^a and Direction
	Observations (n)	GM ($\mu\text{g}/\text{m}^3$)	GSD ($\mu\text{g}/\text{m}^3$)	Observations (n)	GM ($\mu\text{g}/\text{m}^3$)	GSD ($\mu\text{g}/\text{m}^3$)	
Portland							
EC	18	0.4	1.4	36	0.4	2.3	
PM _{2.5}	17	4.2	1.4	35	5.8	1.6	+ ^a
Benzene	17	1.0	2.0	30	0.6	1.8	- ^a
Toluene	17	2.4	1.7	30	1.8	2.1	
Ethylbenzene	17	0.4	1.7	30	0.3	2.0	
<i>m&p</i> -Xylenes	17	1.2	1.6	30	0.9	2.2	
<i>o</i> -Xylene	17	0.3	4.3	30	0.3	2.1	
Formaldehyde	15	2.5	3.5	25	3.7	1.8	
Denver							
EC	32	0.6	1.8	32	0.7	2.0	
PM _{2.5}	32	7.1	1.5	29	7.4	2.2	
Benzene	30	1.6	1.5	28	1.5	1.4	
Toluene	30	3.7	1.7	28	4.0	1.4	
Ethylbenzene	30	0.7	1.8	28	0.5	1.7	- ^a
<i>m&p</i> -Xylenes	30	1.9	1.8	28	1.5	1.8	
<i>o</i> -Xylene	30	0.6	1.9	28	0.5	1.7	
Formaldehyde	32	3.4	1.3	30	2.9	1.3	- ^a

^a Significant differences ($P < 0.05$) in median values using Wilcoxon rank sum non-parametric comparison test. + indicates increase in GM; – indicates decrease in GM.

Table 24. Average Summary Weather Statistics for First and Second Visits at Six Terminals

	Philadelphia		Columbus		Milwaukee		Phoenix		Portland		Denver	
	First	Second										
Relative humidity (%)	39.1	41.9	70.4 ^a	54.1	78.8 ^a	57.7	13.3 ^a	16.1	57.1	53.5	51.4	49.3
Temperature (°C)	3.7 ^a	11.2	2.3 ^a	11.7	9.4 ^a	15.8	34.0 ^a	36.0	23.2 ^a	20.6	19.9 ^a	22.4
Wind speed (kph)	12.9 ^a	9.8	11.4	12.1	20.9 ^a	14.8	11.7	11.4	11.4 ^a	13.4	11.3 ^a	16.3

^a Differences are statistically significant at $P < 0.05$ level using the Wilcoxon rank sum test.

Note: Proximity to an interstate and industrial activity did not change.

had a significant impact on observed upwind background concentrations. For example, there were significantly lower wind speeds and humidity (less rain) during the second site visit in Milwaukee, both of which would support the expectation of higher PM concentrations. However, these elevated predictions were damped somewhat by higher temperatures during the second visit.

The average difference between the actual and predicted values was 40% for yard background. This margin of error is smaller than predicted by the original structural equation model, which left 50% (upwind yard background) of

the variability unexplained (based on R^2 values). The only site visits with predictions outside these expected ranges were the second site visits to Phoenix and Denver and the second site visit to Columbus for yard background. When the actual yard background observations were inserted into the model in place of the predicted values, the differences between the predicted and observed values declined dramatically: from 1.19 to -0.03 (58% to 1%) in Phoenix and from 1.07 to 0.46 (67% to 29%) in Denver.

Table 25 lists the concentrations of VOCs and PM for nonsmoking driver exposures across the repeat site visits

Table 25. Summary Statistics for Nonsmoking Driver Exposures for Two Visits at Six Terminals^a

Location / Compound	First Visit			Second Visit			Significant Difference ^a and Direction
	Observations (n)	GM ($\mu\text{g}/\text{m}^3$)	GSD ($\mu\text{g}/\text{m}^3$)	Observations (n)	GM ($\mu\text{g}/\text{m}^3$)	GSD ($\mu\text{g}/\text{m}^3$)	
Philadelphia							
EC	12	0.8	1.8	18	1.0	1.5	
PM _{2.5}	12	18.1	1.8	18	17.8	1.6	
Benzene	12	1.7	1.3	15	1.2	1.4	-b
Toluene	12	4.2	1.9	15	4.1	1.7	
Ethylbenzene	12	0.7	1.5	15	0.5	1.4	-b
<i>m&p</i> -Xylenes	12	2.8	1.6	15	1.8	1.4	-b
<i>o</i> -Xylene	12	0.9	1.6	15	0.6	1.4	
Formaldehyde	13	7.1	1.3	15	4.4	3.9	
Columbus							
EC	9	1.1	1.6	11	0.8	1.6	
PM _{2.5}	9	12.2	1.5	11	13.8	1.5	
Benzene	2	2.1	1.3	10	1.2	1.3	-b
Toluene	2	4.5	1.4	9	5.0	1.6	
Ethylbenzene	2	1.2	1.2	10	0.9	1.7	
<i>m&p</i> -Xylenes	2	3.9	1.1	10	3.1	1.8	
<i>o</i> -Xylene	2	1.5	1.1	10	0.9	1.6	-b
Formaldehyde	2	12.1	1.4	10	6.2	1.3	-b
Milwaukee							
EC	13	0.7	1.6	17	0.8	1.7	
PM _{2.5}	13	6.7	1.5	17	12.8	1.5	+b
Benzene	17	0.8	1.7	16	0.9	1.4	
Toluene	17	2.0	1.9	16	3.3	1.6	+b
Ethylbenzene	17	0.4	2.2	16	0.5	1.7	
<i>m&p</i> -Xylenes	17	1.7	2.3	16	1.6	1.7	
<i>o</i> -Xylene	17	0.5	2.1	16	0.5	1.7	
Formaldehyde	17	12.3	1.7	16	7.4	1.3	-b

Table continues next page

^a Summary statistics were limited to nonsmoking drivers to eliminate the effect of smoking on the comparisons.

^b Significant differences ($P < 0.05$) in median values using Wilcoxon rank sum non-parametric comparison test. - indicates decrease in GM; + indicates increase in GM.

to six locations. Overall, concentrations were relatively stable across the 1- to 2-year time period between the first and second visits. Although there was a tendency toward lower driver exposures during the second visit, only about 25% of the samples were significantly different ($P < 0.05$). There were no significant differences in EC concentrations among driver exposures.

HOT-SPOT DETERMINATION

To provide some guidance about hot spots, we assumed that a location that has a median concentration that

exceeds the screening value for a compound in Table 26 is a hot spot for that compound. Further, six compounds — 1,3-butadiene, benzene, xylenes, acetaldehyde, formaldehyde, and diesel exhaust particulate — had observed values that compared with EPA screening values based on cancer risks. Because the observed median values for alkanes, acetone, MTBE, and most aromatics were well below the screening values, we did not consider these compounds.

Examining the upwind yard means for the terminals, we found that 100% exceeded the screening value for formaldehyde and 93% exceeded the screening value for acetaldehyde. Only 6% of the means exceeded the screening

Table 25 (Continued). Summary Statistics for Nonsmoking Driver Exposures for Two Visits at Six Terminals^a

Location / Compound	First Visit			Second Visit			Significant Difference ^a and Direction
	Observations (n)	GM (µg/m³)	GSD (µg/m³)	Observations (n)	GM (µg/m³)	GSD (µg/m³)	
Phoenix							
EC	10	1.3	1.5	16	1.3	1.2	
PM _{2.5}	10	23.3	3.1	16	13.5	1.6	
Benzene	9	1.4	1.3	17	1.2	3.1	
Toluene	9	5.7	1.3	17	4.4	5.4	
Ethylbenzene	9	1.2	1.5	17	1.4	8.7	
<i>m&p</i> -Xylenes	9	4.5	1.5	17	6.0	6.0	
<i>o</i> -Xylene	9	1.4	1.4	17	2.4	5.9	
Formaldehyde	9	13.2	1.4	17	9.4	1.4	–b
Portland							
EC	13	1.7	1.9	12	1.1	1.6	
PM _{2.5}	13	15.6	1.4	12	11.4	1.5	–b
Benzene	12	1.7	1.5	12	0.9	1.2	–b
Toluene	12	4.6	1.6	12	3.3	1.5	
Ethylbenzene	12	0.9	1.4	12	0.7	1.8	
<i>m&p</i> -Xylenes	12	3.1	1.3	12	2.7	1.9	
<i>o</i> -Xylene	12	1.1	1.3	12	1.0	1.9	
Formaldehyde	12	8.6	1.8	12	5.5	1.3	–b
Denver							
EC	16	0.9	2.1	12	0.6	7.0	
PM _{2.5}	16	9.3	1.5	12	11.7	1.6	
Benzene	11	1.8	1.5	13	1.9	1.3	
Toluene	11	6.8	1.8	13	7.6	1.6	
Ethylbenzene	11	0.9	1.6	13	1.4	3.6	
<i>m&p</i> -Xylenes	11	2.7	1.6	13	4.7	3.5	
<i>o</i> -Xylene	11	0.9	1.7	13	1.6	3.3	
Formaldehyde	14	10.6	1.5	13	4.6	7.1	–b

^a Summary statistics were limited to nonsmoking drivers to eliminate the effect of smoking on the comparisons.

^b Significant differences ($P < 0.05$) in median values using Wilcoxon rank sum non-parametric comparison test. – indicates decrease in GM; + indicates increase in GM.

Table 26. EPA Screening Values Based on Cancer and Non-cancer Risks^a

Compounds / (CASRN ^b)	EPA Screening Values ($\mu\text{g}/\text{m}^3$)		Hazard Index Target(s)
	Non-cancer	Cancer	
n-Hexane (110-54-3)	200	—	Nervous system
Butadiene (106-99-0)	0.2	0.03	Reproductive system; hematopoietic system cancer
Benzene (71-43-2)	3	0.13	Hematopoietic system cancer; development; nervous system
Ethylbenzene (100-41-4)	100	—	Development; alimentary system
Styrene (100-42-5)	100	—	Nervous system
Toluene (108-88-3)	40	—	Nervous system; respiratory system; development
Xylenes (mixed) (multiple CASRNs)	10	—	Nervous system; respiratory system development
MTBE (1634-04-4)	300	3.8	Respiratory irritation; nervous system; cancer (?)
Acetaldehyde (75-07-0)	0.9	0.45	Respiratory system; cancer
Formaldehyde (50-00-0)	0.98	180	Respiratory system; cancer; eyes
Diesel exhaust particulate (no CASRN)	0.5	—	Respiratory system; cancer

^a EPA screening values from February 2006.^b CASRN indicates the Chemical Abstracts Service Registry Number.

value for benzene; 61% exceeded the screening value for 1,3-butadiene. Based on these criteria, a large number of the sites were hot spots for upwind concentrations. Because the downwind contributions were small compared with the upwind contributions, the terminal-added contributions were small, and the upwind setting overall was the determining factor.

Although diesel exhaust particulate has a low chronic non-cancer screening value ($0.5 \mu\text{g}/\text{m}^3$), our data did not include direct measurements of diesel exhaust particulate. Our source apportionment analyses showed that most of the EC and a portion of the OC were from diesel emissions, depending on how the engines were operated (this issue is discussed in more detail in the section on source apportionment measurements). Given the magnitude of our observed EC and $\text{PM}_{2.5}$ exposures, it was likely that a large fraction of the samples had values that exceeded the chronic screening value for diesel exhaust particulate, because many EC values were greater than $0.5 \mu\text{g}/\text{m}^3$.

DISCUSSION

THE NATURE OF EXPOSURES

The total exposure in potential hot spots can be described as the sum of material contributions from various spatial scales: regional background plus the local area plus personal components. Each of these contributions has a distinct spatial character and time scale of variation. The yard background concentrations of PM and VOCs in our study were assumed to represent regionally stable (low reactivity) components of emissions (such as aged urban traffic emissions in a city with added components from photochemical and other reactions) and losses from reactions (such as those for toluene and 1,3-butadiene). Regionwide background particles were in the accumulation size mode, which remains airborne, plus particulate nitrates and sulfates formed from gaseous nitrogen oxides and sulfur dioxide emissions. The local area components

were emitted by sources within approximately 500 meters of the monitor, such as nearby industrial activities, commercial operations, or highways. The personal components were emitted by sources within the immediate area of the individual, such as the vehicle being driven or the smoking of a cigarette. Each of these layers of contributors has a different time scale of variation, and each is affected by different environmental factors. Because these layers affect each other, they are not independent in space or time, which imposes a complex covariance structure in a regression analysis.

Large variations in exposure intensity and composition were found across the terminals as well as within the terminals and surrounding areas. Our hypothesis was that the locations we chose to measure would have exposures generally higher than regional averages and perhaps at times even high enough to suggest possible health risks.

METHODOLOGY ISSUES

Measurement and analytic methods define what we can observe. In this study, there were several important methodologic issues that limited our observations. First, substantial effort was invested in developing our methodology to describe the layered exposures. We found that the problem of the layered exposures could be effectively addressed by using structural equation modeling. Each layer had its own regression relationship, which fed into the relationship for the next layer above. For example, the yard background concentration was a function of the census region, weather, etc. The next level was the local area model, which had a term for the background concentration.

The second methodologic issue was dealing with the short time scale variations in wind direction and speed. Our Phase 1 design did not provide a good match between the integrated (TWA) samples and the wind variations — the sampling times were too long, and we could not shorten them enough to match the temporal variations without a major loss in sensitivity. During Phase 2 testing, we used real-time monitors for total VOCs and PM_{2.5} that had averaging times of 10 seconds to 1 minute. The wind direction measurements showed variations on the same time scale. This also allowed us to deal separately with the frequent periods of calm (when there was no meaningful directional transport). However, the use of real-time monitors introduced another problem: they did not measure exactly the same environmental components as the TWA sampling. The VOC monitor, the ppbRAE, had a PID that responded strongly to some materials, such as the aromatic vapors, and weakly to alkane vapors heavier than heptane. Similarly, the DustTrak monitor for PM_{2.5} had a light-scattering response that varied with particle size and

composition. If it was assumed that the composition was approximately fixed, then the variation in the real-time monitor's signal could be interpreted as being proportional to the change in the total concentration.

Our principal components analysis of the TWA VOC samples showed that the aromatic and aliphatic vapors were well correlated with each other and that formaldehyde, acetaldehyde, and acetone were correlated with each other but that the oxygenated hydrocarbons were not correlated with the aromatic or aliphatic groups. This implied different sources for the oxygenated hydrocarbons. The PID response could only be interpreted if there were companion data to define the relative composition, which we had from the TWA samples. If the composition did not vary much over 12 hours at a site, then the TWA information could be used to approximate what the PID was measuring. However, we could not test this assumption.

The third methodologic issue was how to handle the variations in space and time for the drivers and to link them to the exposure modifiers and sources. This was done with the use of GPS to measure precisely the locations of the terminals, surrounding sources, and drivers' trucks during their trips. The position information was integrated with geographic data on road locations, industrial activity, and population density as well as with other data using a GIS. The real-time measurement data for the drivers represented a concentration at a location and time, so the GIS approach allowed us to fully integrate the measurement data with all of the descriptors.

TERMINAL UPWIND AND DOWNDOWN HOT SPOTS

Contributions from upwind sources were an important dimension of local exposures at truck terminals. Our interest was twofold. First, for this general type of location, how often was it a hot spot? Second, and more specifically, which source types were associated with relatively high concentrations? As shown by the GIS maps, we found that highways and roads, other truck terminals, warehouses, light industry, greenhouses, and even farm fields can make significant contributions to concentrations downwind. However, a range of contributions was affected by the weather, turbulent dilution by wind, rainfall, and prevailing wind directions. As a result, there were significant differences across the terminal sites because of the variation in their settings, weather, and upwind sources.

The composition of the upwind contributions was variable and did not generally match the yard measurements in the terminal area. Traffic contributions were a constant part of the air contaminants, which makes sense, given the required proximity of the terminals to roads and highways.

As seen in other studies, EC and OC were major components of the particles.

The concentrations in the loading docks and shops of the terminals were nearly all higher than data from the EPA Air Toxics Monitoring Program; only mean benzene concentrations were lower.

An important part of our analysis of samples was directed toward source apportionment. The source apportionment samples were analyzed in Dr. Schauer's lab. The yard background sample was somewhat higher in EC and vehicle OC emissions than the urban background sample measured at a local Supersite monitoring station run by the EPA. As expected from the nature of the sites, EC was primarily from diesel engines. However, the origin of the organic components was not as obvious. A small portion of EC could be separately assigned to diesel and spark combustion engines, but a large portion of "fuel-oil impacted exhaust" could not be assigned to a source, because the relative amount of unburned fuel and oil in the exhaust varies for both cars and trucks depending on their modes of operation. Dr. Schauer and his associates are working to resolve this problem.

Community complaints about irritation and odor from diesel emissions in locations with high volumes of truck traffic were not addressed by our study. We noted that short, intense emissions were present as spikes in real-time data at all three target locations, but we did not attempt to determine whether they were or could have been the sources of the community complaints.

STRUCTURAL EQUATION MODELING FOR TERMINAL VOC SAMPLES

Four structural equation model relationships were developed, one each for benzene, 1,3-butadiene, toluene, and formaldehyde, to describe the effects of various factors in a two-level model: work area and yard background. Work area concentrations were not strongly predicted by job location or yard concentrations ($R^2 = 0.18\text{--}0.38$). However, the yard levels for 1,3-butadiene, toluene, and formaldehyde were strong functions of temperature, wind speed, distance to an interstate highway, and regional location ($R^2 = 0.80\text{--}0.90$). Benzene was less a function of those variables ($R^2 = 0.45$). These factors had effects consistent with our understanding of the behavior and environmental chemistry of these VOCs.

Terminal contributions could only be detected by subtracting the upwind concentration from the downwind concentration when the wind was obviously blowing from one sampler to another. Because of the wind variation, the TWA integrated samples were poor indicators of short-duration upwind-downwind differences; they only

showed a small average difference, equaling only a few percent. The real-time data indicated much higher percentage differences in some cases, as high as 75% higher than the upwind value, when the wind had moderate velocity and was clearly directed from the upwind to the downwind monitor. A major part of the total contaminants came from upwind in the settings where there were highways, other terminals, warehouses, and light industry upwind, that is, industrial park settings.

On average, approximately 25% of the land use of areas within 1 kilometer of the monitored terminals fell into the ICT category as defined by the USGS. However, there was a high degree of heterogeneity for the percentage of areas in this land-use category, with percentages ranging from 6% at the Maryland terminal to 92% at the Florida terminal. The category of land use itself was not a significant predictor of elevated air toxic levels. However, the transportation part of land use, indicated by distance to a highway and road type, was an important predictor. The terminals were typically nested within dense local road networks a few kilometers from a heavily trafficked interstate, but some were more than 7 kilometers from the nearest interstate.

Where a truck terminal (or other facility with regular truck traffic, such as a highway truck stop or a large retail store) was located in a residential area without other pollution sources nearby — an area with a low background — the traffic or individual truck emissions could be quite noticeable to the residents. In general, individuals do not notice the TWA concentration; they do notice the brief periods when there is sensory stimulation: eye, nose, or throat irritation; odors; or visible smoke. Thus, the real-time data provided a more useful picture of this type of exposure, the brief peaks that might be noticed by the residents.

DRIVER HOT SPOTS

Exposures during driving were generally higher than background, especially on major highways. The temporal variation in real-time measurements showed the presence of brief, high-intensity peak exposures. The peaks we observed were generally orders of magnitude above the yard background levels and were generally associated with incompletely mixed concentrated emissions and sources close to the monitor. Because the real-time VOC monitor only measured total VOCs (weighted by the responsiveness of the PID), the composition of materials present had to be extrapolated from local source data or obtained from the analysis of TWA samples collected concurrently, which only provided average composition. The most useful data was obtained when we could reasonably assign the VOC peaks to recognized sources, such as cars and trucks in traffic. We found that the VOC and PM_{2.5}

real-time data were not well correlated with each other; their peaks generally occurred at different times, which strongly indicated that some VOCs were from different sources than the PM_{2.5} or from different operating conditions of an individual source.

Drivers had exposures that changed through space and time. Trucks moved at different speeds, in different directions, in varying traffic densities, and were subjected to the weather and local emissions in the areas that they passed through. Our study required a sophisticated system that allowed us to track all of these dimensions at the same time. In our Phase 2 testing we used a GPS monitor to track the location of the truck as the driver made deliveries and picked up new freight from customers. These data were entered into our GIS, which allowed us to superimpose the trip information on maps that showed roads and residential and industrial land-use densities. With the GIS, we could also link the data with temporal data on PM_{2.5} and total VOC observations and with weather data. The resulting very large data set allowed us to identify when and where the exposures spiked. Interestingly, short spikes in total VOCs represented 5% of values on top of background levels, and they occurred when the truck was stopped in the terminal, at delivery sites, or in traffic, which appeared to allow the trucks' own VOC emissions to accumulate around the truck cab. These emissions quickly dissipated when the truck started moving again. In most cases, these spikes did not make a major contribution to the overall average, because their durations were too short.

PM_{2.5} levels were not well correlated with VOCs and showed a different pattern of associations with driving activities. PM_{2.5} levels usually declined when the truck was stopped and were highest when the truck was moving at highway speeds. These differences in the temporal patterns of total VOCs and PM levels strongly suggested different sources as well as behaviors of the two types of emissions. For example, VOCs are much more mobile than particles; they have much higher diffusion rates and are more readily diluted with turbulent mixing than are particles. As a result, there were fewer and lower peaks and higher average concentrations for VOCs. VOCs can also evaporate from surfaces where they have been deposited, such as materials deposited by driver smoking.

The GIS is a powerful way to present very complex spatial and temporal data and to show associations among them. There is far more work to be done to develop methods to analyze this very complex data set. Our descriptive analysis has shown a number of interesting associations between the driver's location and speed data and the real-time VOC and PM_{2.5} data, which would be very difficult to observe by any other means. For example,

when a truck was parked with the windows up, we saw that the total VOCs increased sharply inside the cab. We believe this was the result of adsorbed or condensed materials off-gassing, which might be cigarette smoke condensate, fuel tracked in, or cleaning solvents. Nonsmoking drivers often complained of cigarette smoke odors and residues on the windshield when they took a truck. The combination of the GPS showing where the truck was and that it was stationary, with satellite photographs showing the characteristics of the location, and with the VOC monitor showing a steady increase in vapor concentrations was very informative. We are exploring statistical methods using time series and mixed modeling approaches to characterize on-road exposures more accurately. These techniques should allow us to construct a detailed exposure model for drivers.

REPEAT SITE VISITS

One of our major concerns was the representativeness of the measurement data. Each site was observed for only one week, and conditions could vary substantially over time because of weather, changes in terminal business activity levels, equipment breakdowns, and other factors. Our study considered how much conditions changed from year to year and how stable the environmental pollutant conditions were. The repeated-measures analysis showed that the pattern of what was high and low was reasonably consistent from year to year. Also, the quantitative differences were modest — most were ± 27% — and only a few were statistically significant.

COMPARISON WITH OTHER STUDIES

One of our goals was to determine if industrial parks were hot spots. We found that the upwind contributions showed a wide distribution of concentrations, which indicated that upwind areas were definitely not consistently hot spots. However, there were combinations of conditions that led to a shift in the distribution of exposures to higher values. Terminal locations that were dense with emission sources or close to major highways, other terminals, warehouses, and light industry had higher than average concentrations and more upper-tail high concentrations. The distributions across locations appeared to represent a continuum with no evidence of a bimodal distribution — meaning there was no clear, distinct group of locations that could be identified as consistently being hot spots.

The EPA Air Toxics Monitoring Program (U.S. EPA 2006) offered a point of comparison for our sampling data. This sampling program was designed to characterize the magnitude and composition of potentially toxic air pollution in

or near urban locations. However, the program made no attempt to collect a statistically representative sample of locations in the United States. Most of the locations tested were chosen because of their proximity to local sources, such as highways or point sources. Samples were collected in vacuum canisters for hydrocarbons, chlorinated hydrocarbons, and selected polar compounds. Carbonyl compounds, including aldehydes, were collected with cartridge samplers (U.S. EPA 1999a). The EPA did not measure every contaminant that we measured, but it did measure the majority and all of compounds that were associated with acute or chronic toxicity. Overall, all of our yard measurements were highly consistent with those obtained by the EPA, as shown in Table 10. The primary difference was that our data were less variable, except for 1,3-butadiene, acetaldehyde, and acetone. The smaller SDs might have been a reflection of a more homogeneous set of sites in our data set; the EPA data set had more low-level background sites. There appeared to be an outlier or two in the EPA formaldehyde data because, although our medians were comparable with those of the EPA, the EPA's mean and SD were much larger. Within the two data sets there were substantial differences, especially for the EPA's rural and small-town measurements, which would be expected.

The EPA data also had some specific point-source measurements that were much higher than those in our data. However, the latter sampling situations were a small portion of the EPA data set and generally did not affect the overall medians.

Although a number of studies have measured concentrations of VOCs and aldehydes, none have focused specifically on industrial parks or settings in which drivers might experience intense and prolonged exposure to diesel exhaust and exhaust from other combustion sources. A few studies have focused on VOC and aldehyde exposures in microenvironments similar to those observed in our study, including background, traffic, and transportation depots. These studies included exposure data from the United Kingdom (Kim et al. 2001, 2002), Mexico (Serrano-Trespalacios et al. 2004), and Hong Kong (Ho et al. 2002, 2004; Lee et al. 2002), which are provided in Table 27 for comparison. Our driver exposures were lower than the least exposed microenvironment in the U.K. study (a bus station), and exposures to formaldehyde and acetaldehyde were somewhat higher for our drivers compared with roadside observations made in Mexico and Hong Kong. However, our measurements were made in trucks that mainly traveled in American cities during non-rush-hour

Table 27. Comparison of Mean VOC and Aldehyde Concentrations^a

Compound / VOC	Birmingham, U.K. ^b				Mexico ^c Back-ground	Hong Kong ^d Roadside	Current Study ^e	
	Bus Station	Major Roads	Cars	Buses			Drivers	Yard Upwind
1,3-Butadiene	0.9 (0.7)	1.8 (0.9)	7.9 (4.7)	1.7 (0.9)			0.5 (0.9)	0.2 (0.5)
Benzene	20.0 (16.1)	49.6 (22.4)	203.7 (152.3)	20.2 (7.8)			2.0 (1.3)	1.2 (0.9)
Toluene	47.3 (33.8)	108.1 (50.3)	494.0 (283.6)	69.3 (30.9)			6.6 (6.0)	3.7 (3.4)
Ethylbenzene	3.8 (1.3)	12.4 (8.6)	51.9 (30.8)	8.0 (3.9)			1.7 (6.5)	0.6 (0.5)
p-Xylene	3.5 (1.0)	11.6 (8.4)	52.5 (31.5)	7.6 (3.9)			5.1 (15.6)	1.9 (1.7)
m-Xylene	10.1 (3.7)	32.3 (21.2)	127.2 (76.8)	20.3 (10.2)				
<i>o</i> -Xylene	3.5 (1.3)	13.2 (9.7)	54.2 (33.6)	8.6 (4.1)			2.0 (8.3)	0.7 (0.6)
Styrene	0.6 (0.4)	1.7 (1.5)	4.3 (3.1)	0.7 (0.3)			1.6 (7.1)	0.3 (0.4)
Toluene:	2.4	2.2	2.4	3.4			3.3	3.1
Benzene								
Formaldehyde					5.5 (2.2)	4.7 (2.5)	9.2 (6.7)	3.3 (1.8)
Acetaldehyde					4.3 (2.1)	2.1 (1.4)	6.5 (6.0)	2.4 (2.5)

^a Mean (SD) expressed in $\mu\text{g}/\text{m}^3$.

^b Kim et al. 2001, 2002.

^c Serrano-Trespalacios et al. 2004.

^d Ho et al. 2002, 2004; Lee et al. 2002.

^e Davis et al. 2007.

times; traffic density was higher in the non-American comparison sites and consequently more likely to be associated with greater emissions.

Table 11 shows comparisons of our study values with those of other studies that looked at neighborhood exposure levels. The distribution of values observed in our upwind background samples was comparable to or lower than those seen in inner-city locations in Los Angeles and New York City (Sax et al. 2004). Los Angeles had much higher levels of aromatics, nearly tenfold higher, presumably associated with the heavy influence of vehicle traffic near the neighborhoods. Higher values were seen in a study in Brisbane, Australia, that evaluated a neighborhood that had intense traffic and light industry and that was next to a petroleum distribution station (Hawas et al. 2002). The Relationships of Indoor, Outdoor, and Personal Air (RIOPA) study values from Houston, Texas; Los Angeles, California; and Elizabeth, New Jersey (Weisel et al. 2005) were also higher than our study values. Measurements in residential areas of Minneapolis, Minnesota, were more nearly comparable (Adgate et al. 2004). All of the sites evaluated in these comparisons were in the central part of a city or near industrial sites and heavy traffic, whereas the truck terminal sites in our study were often located on the outskirts of major metropolitan areas near highways and light industry but usually not in areas affected by dense traffic. This comparison clearly indicates the higher exposures of inner-city residents.

CONCLUSIONS

The principal objective of our hot-spot study was to develop a descriptive picture of the sources and of the statistical exposure distributions at three potential hot-spot locations: upwind and downwind of large truck terminals and inside truck cabs while the trucks were being driven. The terminal samplers were placed at positions along fence lines. The upwind location was chosen to represent conditions near industrial parks and other concentrations of light industry, commercial activity, and trucking operations. The downwind location was chosen because concerns had been expressed that residential areas located near truck terminals might have higher-than-average exposures to truck emissions. In this study of conditions near a random set of truck terminals, we were not attempting to measure conditions in actual communities but to explore the potential for high exposures in those settings. We were also not attempting to perform air transport modeling from sources upwind of the terminals, which would have required a different type of study design.

Our primary conclusions are:

1. The industrial, commercial, and truck activities that took place upwind of the terminals produced modest but significant increases in the concentrations of individual VOCs and PM components above regional background, depending on the proximity of roads, land use, and weather conditions. Sets of alkanes (6- to 8-carbon branched chains) and aromatic compounds (benzene and substituted single-ring compounds) were highly correlated within groups. Traffic emissions were a major component of observed VOC contaminants. Aldehydes were not well correlated with the hydrocarbons or PM_{2.5}, but they were related to EC particles.
2. Downwind fence-line concentrations near the terminals, on average, were not significantly elevated compared with upwind concentrations in the 12-hour TWA samples. A few terminals showed small significant elevations for downwind compared with upwind concentrations, but overall the downwind TWA contributions were small. The downwind VOC components and their relationships were very similar to those of the upwind samples. Preliminary findings from real-time total VOC measurements indicated that during short periods with fixed wind directions there were higher downwind VOCs, but these differences were averaged out of full-period TWA measurements because of wind direction fluctuations.
3. In-cab conditions during driving showed the largest overall increases in exposures compared with regional backgrounds. Exposure levels for some VOCs and PM were associated with driver smoking, road type, age of the truck tractor, and window status (open or closed). In general, the truck being driven was not a major contributor to the driver's exposure. Having windows open resulted in higher levels than having windows closed for EC, PM_{2.5}, and traffic-associated VOCs.
4. The composition of VOCs and PM across terminals was dominated by traffic-related air contaminants, namely low-molecular-weight aromatic and alkane compounds. The BTEX aromatic compounds were generally highly correlated with each other. 1,3-Butadiene and formaldehyde both showed evidence of environmental reactions: 1,3-butadiene declined with increasing temperature because of losses; formaldehyde increased because of photochemical formation.
5. Pollutant levels in the hypothesized hot spots we studied were not dramatically elevated compared with typical urban background levels, although there were few data to compare with our observations. Because our data were collected at truck terminals at randomized city locations and in random order, we

believe they are representative of conditions at such terminals across the United States. However, they were small samples, which might not give a precise estimate of the overall distribution. Nevertheless, these are useful data for assessing the frequency of various concentration levels in these locations.

IMPLICATIONS OF FINDINGS

The term hot spot is an intuitive label for a high-exposure setting. However, it is difficult to define precisely. More important, it carries an implication that a person in a hot spot will be excessively exposed and will be at increased risk for adverse health effects. But what effects represent this risk? We chose to use the EPA's screening value concept to define when exposures at a location were sufficiently high for it to be considered a hot spot. The advantage of these values was that they represented a conservative approach to the risk of everyday exposures for the general population. Clearly, other values could be chosen, such as occupational limits or levels associated with acute danger to life, which are generally much higher. However, these higher levels seemed inappropriate for our concern for daily exposures. In this study, we found that exposures at our hot-spot locations could be high on occasion but were certainly not high most of the time. For one of our terminal-related hot spots to have high exposures, two conditions would have to have been met: high emissions and suitable wind transport to the point of exposure. If either of these conditions was not met, there would be no hot spot, as is well known for stationary point sources. Consequently, the description of exposure at a hot spot is a probability distribution for the concentrations likely to occur.

We set out to determine if three types of exposure locations appeared likely to be hot spots: industrial parks, neighborhoods near trucking operations, and truck driving in heavy traffic. We concluded that under some circumstances high concentrations can occur in these settings.

- Industrial parks are areas with aggregate light industrial activities, such as truck terminals, large warehouses, light manufacturing, and heavily traveled roads, which can together create a hot spot downwind when the weather is suitable.
- Neighborhoods immediately downwind of an individual truck terminal appear to receive a generally modest contribution from the terminal. However, the terminal does contribute to local conditions and can be one factor in forming a local hot spot.
- Driving in heavy urban traffic can put someone in a local hot spot of traffic emissions. Diesel emissions from trucks are a major contributor to these conditions,

especially short-term, high-intensity peak exposures in a vehicle following a high-emitting truck.

- Formaldehyde, acetaldehyde, and their precursors are emitted by vehicular traffic and are formed photochemically in the atmosphere. In our study they were commonly above the EPA screening values.
- Diesel emissions indicated by EC frequently exceeded the EPA screening value of $0.5 \mu\text{g}/\text{m}^3$.

These findings imply that car drivers with long commutes on major highways can have exposures to traffic emissions that match occupational exposures, which are associated with increased risks of lung cancer and cardiovascular disease.

UNRESOLVED SCIENTIFIC QUESTIONS

Where do hydrocarbon and aldehyde vapors come from? Our findings support the view that they come predominantly from traffic emissions. It was clear that VOCs and PM did not vary together; their correlation was low. PM came with exhaust, and some VOCs were present in exhaust, but exhaust did not appear to be the predominant source. VOC values increased when the vehicle stopped. Particle and 1,3-butadiene exposures came primarily from outside the vehicle. Closing the vehicle's windows reduced EC, $\text{PM}_{2.5}$, and 1,3-butadiene levels but increased aldehyde levels. Aldehyde levels were high for both smoking and nonsmoking drivers but a little higher for the former (10%–30%), indicating that smoking was not the primary source. This uncertainty could be resolved by sampling simultaneously inside and outside the vehicle, which would more precisely define emission sources and the degree to which the driver's own vehicle was a source of the interior exposures.

More long-term sampling is required at a single site to better characterize upwind and downwind exposures. Data collected over an extended time period would enable us to accurately identify and track the many variable source contributors to terminal upwind exposures, allowing us to clearly estimate when the terminal contributes significantly to downwind exposures. Although we were able to identify the existence of a terminal contribution and some significant downwind contributions from some truck terminals, further data would allow us to build a more detailed exposure model that incorporated these contributions. Also, long-term sampling is necessary before factor analytic methods such as principal component analysis can provide a robust estimate of source characteristics at an individual location.

Although the EPA screening values suggested that 1,3-butadiene, benzene, formaldehyde, and acetaldehyde

exposures were above the levels that merit some concern, what actual risk was present was not clear. Communities downwind of major transportation facilities, such as bus depots, truck stops, warehouses, major retail outlets, and truck terminals, have been found to have relatively high levels of these contaminants. Community studies targeting these contaminants could clarify the risks.

This study produced clear evidence that vehicle emissions contribute significantly to driver exposures, but it was unable to distinguish the diesel and spark-emission engine sources. More work is needed to develop methods, such as molecular tracers, to make this distinction — not because we expect that the health effects would be different, but because it will affect how interventions are chosen to reduce exposures.

POTENTIAL FOR FUTURE EPIDEMIOLOGIC STUDIES

In this study we have demonstrated the feasibility of measuring VOCs in various work environments in the trucking industry and noted differences in exposure patterns that suggested different source characteristics across the work environments. Background exposures (upwind) proved to be the lowest exposures; the highest exposures varied between the mechanic shop (alkanes, ethylbenzene, xylenes, and acetone), the loading dock (toluene and formaldehyde), and the truck cabs (hexane, 1,3-butadiene, benzene, styrene, MTBE, and acetaldehyde).

Although there is concern about exposure to VOCs and cancer in humans, previous epidemiologic studies have mainly focused on the relationship between exposure to air toxics and respiratory symptoms, including the worsening of asthma syndromes (Dales and Raizenne 2004; Morello-Frosch and Jesdale 2006). Delfino and colleagues (2003b), for example, conducted a panel study in 22 Hispanic children with asthma in Los Angeles and found positive associations between asthma symptoms and increases in ambient benzene and formaldehyde. In an additional analysis, associations were noted between symptoms and exhaled-breath VOCs, including toluene, xylene, and benzene (Delfino et al. 2003a). In a study by Arif and Shah (2007), a cohort of 669 subjects ages 20 to 59 years was drawn from the National Health and Nutrition Examination Survey (NHANES) (Centers for Disease Control and Prevention 1999–2000) and wore passive organic vapor monitors. Subjects were identified who had previously indicated that they had asthma diagnosed by a health professional and whether they had attacks of wheezing in the past 12 months. Factor analysis identified subjects with increased effects among those exposed to aromatic compounds. The odds of physician-diagnosed asthma were greater in subjects with this exposure, and in

subjects without asthma there was a greater risk of attacks of wheezing. Analyses were adjusted for smoking, body-mass index, household smoking, and poverty level. Other studies assessing indoor exposures have reported similar associations (Wieslander et al. 1997; Rumchev et al. 2004).

Little attention has been paid to air toxics in the assessment of the health effects that have been attributed entirely to PM and, in particular, to PM from mobile combustion sources. There is a large body of emerging literature, primarily in children, relating proximity to traffic to respiratory symptoms (Weiland et al. 1994; Duhme et al. 1996; Van Vliet et al. 1997; Guo et al. 1999; Venn et al. 2001) and, in some studies, to reduced pulmonary function (Brunekreef et al. 1997) and asthma (Nicolai et al. 2003). There have been fewer traffic studies for adults (Nitta et al. 1993; Oosterlee et al. 1996). We (Garshick et al. 2003) studied male U.S. veterans drawn from the general population of southeastern Massachusetts and — adjusting for cigarette smoking, age, and occupational exposure to dust — found that subjects living within 50 meters of a major roadway were more likely to report persistent wheezing, particularly those living within 50 meters of heavily trafficked roads ($\geq 10,000$ vehicles/24 hr; OR = 1.71, 95% CI = 1.22–2.40); the risk of chronic phlegm in those living within 50 meters of heavily trafficked roads was of borderline significance (OR = 1.40, 95% CI = 0.97–2.02). These results suggest that residential exposure to vehicular emissions near busy roadways results in respiratory disease symptoms and asthma in adults and children and that VOCs associated with traffic might contribute. Some of the highest VOC exposures we measured were for formaldehyde and acetaldehyde, which are well-known respiratory irritants.

Our current study supports the conclusion that locations with aggregations of light industry, warehouses, and truck terminals have significantly higher background levels of VOCs and PM. Thus, neighborhoods downwind of these aggregations can experience hot-spot conditions; these neighborhoods are not uncommon. Contributions from individual truck terminals were modest and did not dominate the exposures. It was the aggregation of many sources and high volumes of local traffic that could make these areas hot spots at times. The contribution of these exposure conditions to increased risk of respiratory or other effects is not known, but the studies noted above suggest that they could be important. We believe that useful studies could be designed to explore this hypothesis. Our structural equation model of background exposures linked to residential address could be used to predict historical exposures and to study relationships with selected health effects that might include chronic respiratory disease or cancer. Children are a particularly sensitive population for

these kinds of exposures. Other outcomes might include the occurrence of chronic respiratory symptoms and asthma, assessed using a community-questionnaire survey linked to exposure models. Individuals with long daily commutes to work are another target population that could be studied with the exposure-assessment techniques developed in our study.

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APPENDIX A. NCI Trucking Industry Particle Study

DESCRIPTION OF THE NCI TRUCKING INDUSTRY PARTICLE STUDY

Our study was an add-on to the ongoing NCI Trucking Industry Particle Study, a national study of lung cancer and particulate exposures in the U.S. trucking industry. In the latter study, 36 large terminals (> 100 employees) across the country were selected (Figure A.1) to represent the occupational particulate exposures of trucking industry employees in various terminal work areas, specifically the outdoor yard and freight dock areas and their associated workers; the repair shop and their mechanics; and the truck drivers, including P&D drivers as they drive in city traffic picking up consignments and making deliveries and long-haul (LH) drivers as they drive between large terminals in various cities. The terminals were located in most of the major metropolitan areas of the United States, some smaller cities, and a few rural areas. Where there was more than one terminal in an area, such

as in Greater Los Angeles, one was chosen at random. The order in which terminals were visited for testing was randomized across the terminals; the 15 terminals in our hot-spot study were a random subset of the original 36.

The overall goals of the NCI Trucking Industry Particle Study were to describe the distribution of fine particulate exposures at randomly chosen terminals, to identify the factors that affected the levels of exposure at work locations in and around the terminals, and to define driver exposures and factors affecting them. In that study, we measured the work shift (8–12 hours) exposures to $\text{PM}_{2.5}$ and to EC and OC in $\text{PM} \leq 1 \mu\text{m}$ aerodynamic diameter (PM_1). PM_1 was chosen to represent freshly generated combustion particles, and more than 90% of $\text{PM}_{2.5}$ was PM_1 , based on concurrent size-selective measurements. VOC measurements for our add-on hot-spots study were collected concurrently with the particulate measurements. When the NCI study ended, concurrent $\text{PM}_{2.5}$, EC, and OC measurements continued to be collected as part of the add-on study to obtain a complete and matching data set of particles and VOCs.

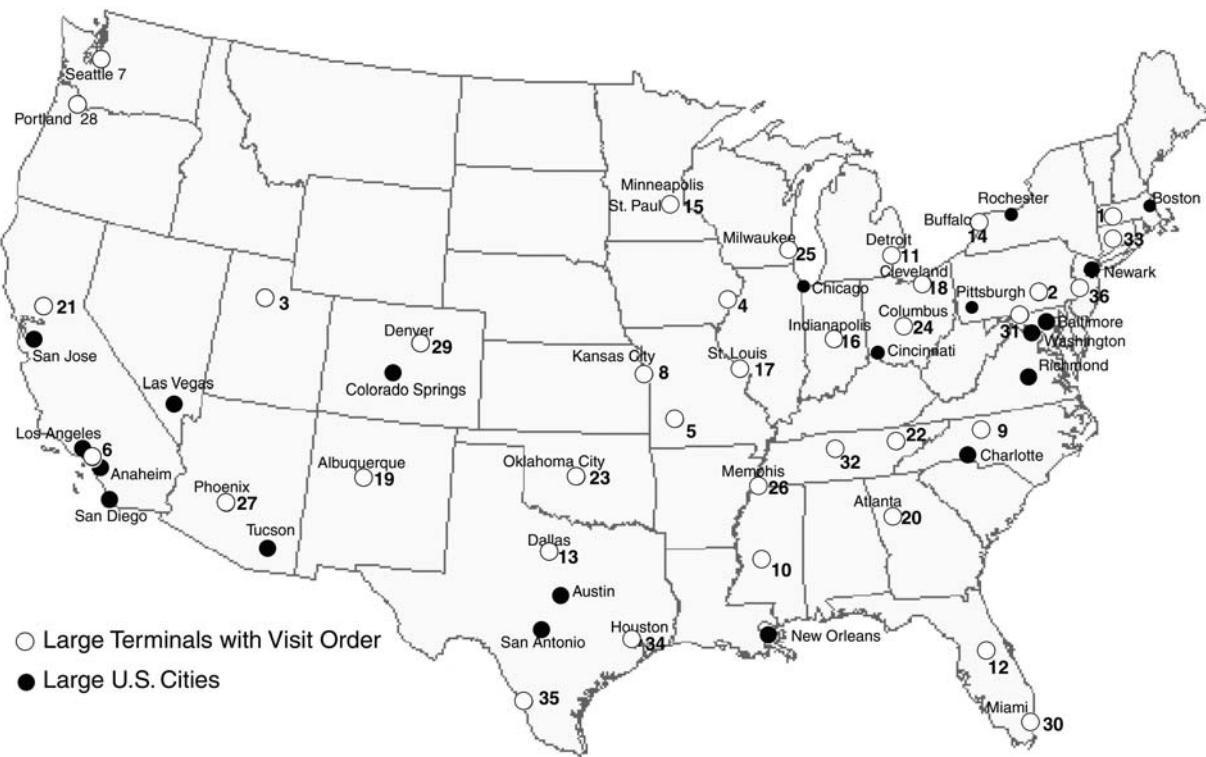


Figure A.1. Map of the United States, showing locations of all 36 terminals visited. Numbers indicate the order in which the terminals were visited.

METHODS USED IN THE NCI TRUCKING INDUSTRY PARTICLE STUDY

PM_{2.5}

PM_{2.5} was measured using a slightly modified sampling pump (Vortex Timer 2, Casella, Amherst, NH), a GK2.05 SH (KTL) cyclone pre-selector (BGI, Waltham, MA), and a 37-mm Teflo filter (Pall, Ann Arbor, MI) with a pore diameter of 1 µm. The filters were placed in 37-mm three-piece polystyrene cassettes (Millipore, Bedford, MA). The pumps were calibrated to 3.5 L/min, the flow rate at which the cyclone conforms to the EPA (PM) standard for a 50% cut point of 2.5 µm. The mass collected on the filters was determined with an analytic balance (Micro-Gravimetric M5, Mettler Instruments, Hightstown, NJ). The filters were weighed before and after sampling in a room controlled for temperature and humidity. Before each weighing, the filters were conditioned at 21 ± 1.5°C and 40 ± 5% relative humidity (EPA specifications) for at least 48 hours. The filters were archived at -20°C for analysis in a future project.

A quality assurance–quality control (QA–QC) program was run for all of the samples, including field and lab blanks, replicate samples, and determination of LODs. Sample batches included periodic introduction of blanks and duplicate samples that were not identified to the lab. Any sample with a potential problem in the field or lab was flagged in the data set.

Elemental Carbon and Organic Carbon

EC and OC were measured using a Harvard field monitor, equipped with a slightly modified sampling pump (Casella), an SCC1.062 Triplex cyclone pre-selector (BGI), and a 25-mm quartz tissue filter (Omega Specialty Instruments, Chelmsford, MA) with a pore diameter of 1.2 µm. The filters were placed in 25-mm three-piece polystyrene cassettes (Millipore). The pumps were calibrated to 3.5 L/min, the flow rate at which the cyclone conforms to the EPA (PM) standard for a 50% cut point of 1.0 µm. Prior to field sampling, the quartz tissue filters were pre-fired at 900°C for 5 hours, and the foil used to line the Petri dishes for filter storage was pre-fired at 550°C for 15 hours. This was to prevent off-gassing from the Petri dish material onto the filters. Upon returning from the field, the filters were stored at -20°C and analyzed for EC and OC using the National Institute for Occupational Safety and Health 5040 thermo-optical analyzer method (Birch and Cary 1996; National Institute for Occupational Safety and Health 1998) at the laboratory of Dr. James Schauer (University of Wisconsin, Madison, WI).

A QA–QC program was run for all of the samples, including field and lab blanks, replicate samples, and

determination of LODs. Sample batches included periodic introduction of blanks and duplicate samples that were not identified to the lab. Any sample with a potential problem in the field or lab was flagged in the data set.

Source Apportionment Samples

Our analytic scheme for the source apportionment samples required larger samples of the materials. These were therefore collected at 16.7 L/min using a higher-volume sampling pump (Vortex Ultra Flow, Casella), a 2000-30EH cyclone pre-selector (URG, Chapel Hill, NC), and a 47-mm quartz tissue filter (Omega Specialty Instruments) with a pore diameter of 1.2 µm. The filters were placed in 47-mm 2000-30FG filter cassette packs (URG). The pumps were calibrated to 16.7 L/min, the flow rate at which the cyclone conforms to the EPA (PM) standard for a 50% cut point of 2.5 µm. The high-volume samplers collected PM_{2.5} on quartz tissue filters that were pre-fired at 900°C for 5 hours, and the foil used to line the Petri dishes for filter storage was pre-fired at 550°C for 15 hours. Upon returning from the field, the filters were stored at -20°C and then shipped frozen to Dr. Schauer's lab.

The source apportionment analyses were done by GC-MS in Dr. Schauer's lab to quantify a wide range of specific OC compounds. This detailed chemical profile was used in a chemical mass-balance algorithm for source identification and apportionment to characterize the sources of the PM_{2.5} and to assess the chemical nature of the organic constituents of vehicle exhaust (Schauer et al. 1999). The analysis involved extensive sample preparation, including spiking the filters with seven deuterated internal recovery standards, and five extraction steps. The extracts were combined and reduced in volume, and finally half of the combined extract was derivatized with diazomethane to esterify organic acids. Both the derivatized and underivatized extracts were separated on a GC (model 5890, Hewlett-Packard) using a capillary column 30 m × 0.25 nm in diameter (HP-1701, Hewlett-Packard) and then analyzed with a mass spectrometer (model 5972, Hewlett-Packard). More than 100 compounds were quantified with a relative error of 20%. The following chemical compound groups were evaluated as source markers: n-alkenoic acids, alkane dicarboxylic acids, aromatic carboxylic acids, resin acids, levoglucosan and other sugars, and other OC compounds. The cost and complexity of the analysis limited its application to composited samples from work locations where the types of sources were likely to be approximately constant.

The sampling box was equipped with a HOBO H8 data logger (Onset Computer, Bourne, MA) to obtain real-time data on temperature and relative humidity. The monitor

was also equipped with a GMW25 CO₂ monitor (Vaisala, Woburn, MA), which used a silicon-based infrared absorption sensor to measure CO₂. Output from the temperature, relative humidity, and CO₂ monitor was also stored in the HOBO data logger.

Structural Equation Modeling of Particulate Exposures

As shown in Table A.1, personal exposures to EC were significantly predicted by work area concentrations and personal smoking status ($R^2 = 0.64$). Based on the coefficient and summary statistics for work area EC, an increase of one SD in exposure above average work area levels led to an increase in estimated personal exposures of 32.7% in the shop and 79% in the dock. The predicted value of EC exposures for smokers increased more modestly, by 19% compared with nonsmokers when work area exposures were held constant, which was consistent with the small amount of EC in cigarette smoke (Birch and Cary 1996). Thus, if smoking status was not known, we could still make a good prediction of EC exposure.

Work area exposures to EC were predicted by terminal characteristics, work location (dock or shop), indoor ventilation, and upwind yard concentrations observed in the yard ($R^2 = 0.64$). An increase of one SD in yard size increased work area EC concentrations by 12.6%; the effect of increased numbers of P&D drivers was much smaller, at 3.7% (the number of mechanics was not statistically significant). Ventilation rates, or the degree of closure of the buildings where the source activity was occurring, as measured by an interaction term between job location and outdoor temperature levels, significantly predicted that work area EC would be 57.1% lower in warmer versus colder outdoor temperatures in the shop; the effect of ventilation rates on the dock was negligible (doors in the dock area were rarely fully closed). An increase of one SD in upwind yard concentrations in the yard significantly increased predicted work area EC levels by 78.8%. Also, the large differential between shop and dock exposures was in line with work area source strength differences, and the results showed that area exposures were more than 700% higher in the shop than in the dock.

Differences in upwind yard exposures at the terminal locations were significantly predicted by a number of weather conditions as well as location-specific factors, such as the distance to a major road and the percent of ICT land use nearby ($R^2 = 0.51$). The percent of ICT land use within a 1-kilometer radius was significant in the model, where an increase of one SD above the mean led to a 28.1% increase in the amount of EC observed in the terminal yard. A dichotomous variable representing a cutoff distance of 500 meters from an interstate highway was a

Appendix Table A.1. Regression Results for EC

$$\text{Equation 1: } \log(\text{PersonalEC}) = \beta_{10} + \beta_{11}\log(\text{WorkAreaEC}) + \beta_{12}(\text{Smoking}) + \epsilon_{ijk}$$

Work Area	0.99 ^a
Smoking	0.17 ^a
Constant	0.05
Equation R²	0.64

$$\text{Equation 2: } \log(\text{WorkAreaEC}) = \beta_{20} + \beta_{21}(\text{Terminal Size}) + \beta_{22}(\text{P\&D}) + \beta_{23}(\text{Shop}) + \beta_{24}(\text{Ventilation}) + \beta_{25}\log(\text{YardEC}) + \beta_{26}(\text{Job}) + \gamma_{ijk}$$

Terminal Size	0.01 ^a
P&D Drivers	0.002 ^a
Mechanics	0.002
Ventilation	-0.09 ^a
Yard Background	0.71 ^a
Job	2.11 ^a
Constant	-0.21 ^a
Equation R²	0.64

$$\text{Equation 3: } \log(\text{YardEC}) = \beta_{30} + \beta_{31}(\text{Relative Humidity}) + \beta_{32}(\text{Temperature}) + \beta_{33}(\text{Wind speed}) + \beta_{34}(\text{Interstate}) + \beta_{35}(\text{Industrial}) + \beta_{36}(4 \text{ Regional Dummies}) + \eta_{ijk}$$

Relative Humidity	-0.004 ^a
Temperature	-0.01 ^a
Wind speed (kph)	-0.11 ^a
Interstate Distance (0-1)	-0.30 ^a
Industrial Land Uses	0.01 ^a
Region 1 (Midwest)	0.51 ^a
Region 2 (Northeast)	0.84 ^a
Region 3 (South)	1.08 ^a
Region 4 (West)	0.68 ^a
Constant	NA ^b
Equation R²	0.51

^a Indicates significant at 5% level.

^b NA indicates not available.

significant predictor of upwind yard EC concentrations, and upwind yard concentrations were 35.1% higher at terminals located closer to an interstate. Relative humidity, temperature, and wind speed were included in the final model to control for weather effects on pollution dispersal and removal, all of which had a negative impact on predicted upwind yard exposure levels. In particular, for predicted upwind yard EC, an increase of one SD in relative humidity decreased it by 7.9%, an increase of one SD in temperature decreased it by 9.2%, and an increase of one SD in wind speed decreased it by 45.5%. Precipitation was excluded from the model because of a lack of variability — 80% of sampling sessions had no precipitation, and more than 90% had less than a tenth of an inch.

Census region designations showed significant differences in background (i.e., upwind yard) EC levels across the United States. The constant term was excluded from the background equation because all four regional dummy variables were represented (*P* values tested the hypothesis stated as “the constant for region *x* is zero”), with the result that each regional coefficient was a multiplicative constant elevating background exposures to various degrees above the hypothetical zero scenario. The biggest regional increases in upwind yard EC exposure levels were seen at truck terminals in the South, followed by the Northeast, the West, and finally the Midwest.

These findings clearly show that our structural equation modeling approach had some major advantages when a stratified sampling approach was used (Davis et al. 2006; Smith et al. 2006). The high degree of correlation among the upwind yard, work area, and personal samples did not produce the problems with collinearity in the structural equation modeling analysis that are common in applications of normal linear regression with this type of sampling data. High *R*² values for the model give it good predictive power. We applied this analytic approach to the VOC and other particulate data collected as part of our study.

APPENDIX B. Data Management — QA–QC Procedures

A rigorous QA–QC program was set up that was compatible with the EPA’s VOC method TO-17 to ensure accuracy, precision, and reproducibility of findings. There are dated written protocols for all field and lab activities. All instruments were calibrated before and after use. Logs were kept on each instrument’s routine maintenance, repairs, and calibrations. Similar procedures were applied to both lab instruments and field equipment, such as sampling pumps. Values of blanks and standards were tracked over time to detect developing problems with analytic instruments and field equipment. There was a requirement to have 10% random lab and field blanks interspersed with samples, replicate analyses, and blinded spiked samples. For example, quality control for filter-weighing included weighing a tare weight and re-zeroing after every five filters and reweighing a control filter after every 10 filters. Our weighing precision for a recent field study was $\pm 3 \mu\text{g}$ for each filter, with an average field blank value of $-2.02 \mu\text{g}$ (*SD* = 3.26) for eight blanks. In addition, we collected routine duplicate field samples, which were not identified as such when submitted for analysis. Analyses were performed blind, although when needed an analyte range was identified to avoid wasting samples. In all lab

analyses, when possible, we used the addition of internal standards to ensure correction for losses during analysis.

At a review of the project in 2005, we presented a series of quality control data tables that we had developed during the previous 15 months. We have updated these tables and provided some additional data that was requested. After reviewing our records, we find that our double-entry system for the database had an error of 2%.

SAMPLE LOSSES

Table B.1 shows data for aldehyde samples attempted and successfully analyzed by visit to a terminal; Table B.2 shows comparable data for hydrocarbon samples. The overall success rate was 77% for aldehydes, 64% for the upwind and downwind yard samples, and 94% for the in-vehicle samples. The yard samples attempted in Memphis, Miami, Hagerstown, Houston, and Laredo had substantial losses because of high moisture in the air (high humidity, heavy rain, or both). The analyses of the in-vehicle samples were much more successful, because the air was drier. As shown in Table B.2 the success rates for the hydrocarbon samples averaged 88% overall, 85% for yard samples, and 92% for in-vehicle samples. Only one terminal visit (Houston) had a success rate of less than 80%, which again was evidence of the effects of high humidity. A revision to the sampling protocol, designed to solve the moisture problem, is discussed below.

FORMALDEHYDE AND 1,3-BUTADIENE LOSSES

At the review of the project in 2005, concerns were raised about possible losses of formaldehyde and 1,3-butadiene during our sampling. Our hot-spot study data are shown in Table B.3. Formaldehyde levels were generally higher than acetaldehyde levels for both the yard and drivers, but the concentrations were lower in the yard compared with the truck cabs. As expected from other data, the 1,3-butadiene and benzene concentrations and their respective ratios were also in the appropriate ranges. Because the seasonal and diurnal averages have not been separated out, the overall averages include both. The yard data have approximately equal numbers of day and night values, but all of the driver data are daytime values. Our study covered the United States and included urban areas and predominantly rural locations with various mixes of sources, which would be expected to affect the concentrations and ratios. In other studies, the values of concentrations and ratios from various city and roadside locations were comparable to those of our study. However, none of the other studies had the same mix of locations, especially as many suburban and rural sites. Our modeling included the

Appendix Table B.1. Number of Aldehyde Samples Attempted and Successfully Analyzed and the Percentage of Success by Visit to Terminal

Terminal (Date)	Yard Upwind		Yard Downwind		In-Vehicle		Total	Samples Obtained (%)	Field Blanks	Lab Blanks
	Attempted	Analyzed	Attempted	Analyzed	Attempted	Analyzed				
Elizabeth, NJ (1/2004)	0	0	4	3	2	2	6	5	83	1
Oklahoma City, OK (2/2004)	1	1	2	2	3	3	6	6	100	0
Columbus, OH (3/2004)	12	11	2	2	4	2	18	15	83	2
Milwaukee, WI ^a (4/2004)	14	10	15	11	19	19	48	40	83	5
Memphis, TN ^b (5/2004)	10	5	4	2	18	16	32	23	72	2
Phoenix, AZ ^c (6/2004)	5	5	1	1	10	10	16	16	100	2
Portland, OR (7/2004)	20	17	13	11	18	18	51	46	90	6
Denver, CO (8/2004)	18	18	14	14	19	18	51	50	98	4
Miami, FL ^b (10/2004)	4	0	3	0	18	18	25	18	72	2
Hagerstown, MD ^b (10/2004)	13	1	13	2	20	19	46	22	50	6
Nashville, TN (11/2004)	12	8	12	7	20	20	44	35	80	5
Middletown, CT (12/2004)	14	13	14	13	20	16	48	42	88	3
Houston, TX ^b (1-2/2005)	10	2	11	3	20	19	41	24	56	4
Laredo, TX ^b (2/2005)	15	1	13	0	14	12	42	13	31	1
Philadelphia, PA (3/2005)	16	16	13	13	18	17	47	46	98	6
Total	164	108	134	84	223	209	521	401	77	43

^a The full complement of modified sampling boxes was not ready until the fourth terminal visit.^b Upwind and downwind samples were lost in Memphis, Miami, Hagerstown, Houston, and Laredo because of high moisture in the air (i.e., high humidity and heavy rain).^c Delivery of the sampling equipment in Phoenix was delayed until Wednesday of the sampling week.

Appendix Table B.2. Numbers of Hydrocarbon Samples Attempted and Successfully Analyzed and Percentage of Success by Visit to Terminal

Terminal (Date)	Yard Upwind		Yard Downwind		In-Vehicle		Total		Samples Obtained (%)	Field Blanks	Lab Blanks
	Attempted	Analyzed	Attempted	Analyzed	Attempted	Analyzed	Attempted	Analyzed			
Elizabeth, NJ (1/2004)	0	0	4	3	2	2	6	5	83	1	2
Oklahoma City, OK (2/2004)	1	1	2	2	3	3	6	6	100	0	2
Columbus, OH (3/2004)	12	11	2	2	4	2	18	15	83	2	2
Milwaukee, WI ^a (4/2004)	14	13	15	15	19	19	48	47	98	5	2
Memphis, TN (5/2004)	12	12	4	4	18	18	34	34	100	2	2
Phoenix, AZ ^b (6/2004)	5	5	1	1	10	10	16	16	100	2	2
Portland, OR (7/2004)	20	20	14	14	18	18	52	52	100	6	2
Denver, CO (8/2004)	18	16	14	14	19	15	51	45	88	4	2
Miami, FL ^c (10/2004)	17	15	12	10	18	13	47	38	81	4	3
Hagerstown, MD ^c (10/2004)	13	9	13	10	20	19	46	38	82	6	4
Nashville, TN (11/2004)	11	10	11	10	20	20	42	40	95	5	2
Middletown, CT (12/2004)	14	13	14	13	20	17	48	43	90	3	3
Houston, TX ^c (1–2/2004)	15	6	15	6	19	18	49	30	61	2	2
Laredo, TX (2/2004)	17	12	14	12	13	13	44	37	86	1	2
Philadelphia, PA (3/2004)	17	15	13	12	18	17	48	44	92	6	2
Total	186	158	148	128	221	204	555	490	88	49	34

^aThe full complement of modified sampling boxes was not ready until the fourth terminal visit.^bDelivery of the sampling equipment in Phoenix was delayed until Wednesday of the sampling week.^cSome hydrocarbon samples were lost in Miami, Hagerstown, and Houston because of analytic problems caused by high water content in the samples caused by high humidity and heavy rain.

Appendix Table B.3. Summary Data and Comparisons with Other Studies

Variables	Current Study ^a		New York ^b Personal	Hong Kong ^c Roadside	Finland ^d		Mexico ^e Outdoors by Home
	Yard Upwind	Driver			Background	Road Center	
Formaldehyde ($\mu\text{g}/\text{m}^3$)	3.3 ($n = 186$)	9.20 ($n = 201$)	20.0	4.65	1.3	12.8	5.5
Acetaldehyde ($\mu\text{g}/\text{m}^3$)	2.4 ($n = 186$)	6.5 ($n = 201$)	11.6	2.11	0.4	3.6	4.3
Formaldehyde: Acetaldehyde	1.4	1.4	1.2	2.2	2.1 ^f	2.6 ^f	1.3
Benzene ($\mu\text{g}/\text{m}^3$)	1.2 ($n = 237$)	2.0 ($n = 196$)	3.9	4.85	—	—	7.2
1,3-Butadiene ($\mu\text{g}/\text{m}^3$)	0.2 ($n = 237$)	0.5 ($n = 196$)	1.0	—	—	—	0.9
Benzene:1,3-Butadiene	6.0	4.0	3.9	—	—	—	8.0

^a Half of the LOD was substituted for the values below the LOD.^b Kinney et al. 2003.^c Ho et al. 2004.^d Viskari et al. 2000.^e Serrano et al. 2004.^f Mean ratios were calculated on matched samples; annual mean concentrations were not the same matched samples.

effects of seasonal and diurnal variations as well as of different sites. To further verify that we have not been losing formaldehyde by our use of sampling tubes made by SKC, we will do concurrent sampling with DNPH-treated Sep-Pak collectors made by Waters.

DUPLICATE SAMPLES

A limited number of duplicate samples were collected to assess repeatability (Table B.4). Differences in air flow between sample boxes can increase the differences in measurements. Concentrations of compounds close to the LOD, such as 1,3-butadiene in yard samples, can show large relative differences with small values.

BREAKTHROUGH TESTS

A small number of breakthrough tests were performed, showing few compounds with breakthrough problems (Table B.5).

LABORATORY SPIKED SAMPLES

A small number of blank tubes were spiked with known amounts of VOCs and analyzed to determine the efficiency of recovery (Table B.6 and Table B.7). All but 1,3-butadiene (79%) showed at least 80% recovery. The SDs were of the same magnitude as those of the duplicate measurements.

BLANKS

As usual, lab and field blanks were collected (Table B.8). On average the field blanks were slightly higher than the lab blanks.

Appendix Table B.4. Relative Percent Differences (RPD) Between VOC Duplicate Samples Overall and for Duplicates with Similar Flow Rates

Compound	RPD Duplicates Overall (n = 7) ^a	RPD Duplicates with Similar Flow Rates (n = 3)
1,3-Butadiene	86	143
2-Methylpentane	34	32
2-Methylhexane	22	8
3-Methylhexane	23	9
2,3-Dimethylpentane	11	9
2,2,4-Trimethylpentane	27	20
Methylcyclohexane	17	10
Benzene	24	19
Toluene	20	11
<i>m&p</i> -Xylenes	15	25
<i>o</i> -Xylene	31	19
Ethylbenzene	10	18
Styrene	29	11
MTBE	25	29

Note: Duplicate samples were lost in Miami, Houston, and Laredo because of the high levels of moisture in the air.

Appendix Table B.5. Number of VOCs Above 10% Breakthrough Observed in Backup VOC Samples Obtained

Compound	Total Number of Samples ^a	N > 10% Breakthrough
1,3-Butadiene	14	0
2-Methylpentane	14	2
2-Methylhexane	14	3
3-Methylhexane	14	1
2,3-Dimethylpentane	14	3
2,2,4-Trimethylpentane	14	2
Methylcyclohexane	14	2
Benzene	14	2
Toluene	14	0
<i>m&p</i> -Xylenes	14	1
<i>o</i> -Xylene	14	0
Ethylbenzene	14	0
Styrene	14	2
MTBE	14	1

Note: Breakthrough samples were lost in Miami, Houston, and Laredo because of the high levels of moisture in the air.

Appendix Table B.6. Percentages Recovered from Analysis of Spiked VOC Lab Blanks

Compound	N	Mean (%)	SD (%)
1,3-Butadiene	20	79	44
2-Methylpentane	20	83	27
2-Methylhexane	20	85	29
3-Methylhexane	20	84	25
2,3-Dimethylpentane	20	89	25
2,2,4-Trimethylpentane	20	89	23
Methylcyclohexane	20	91	22
Benzene	20	84	24
Toluene	20	88	23
<i>m&p</i> -Xylenes	20	90	26
<i>o</i> -Xylene	20	90	26
Ethylbenzene	20	90	26
Styrene	20	86	26
MTBE	20	95	32

Appendix Table B.7. Percentages Recovered from Analysis of Aldehyde and Acetone Spiked Lab Blanks

Compound / Section	N	Mean (%)	SD (%)	Relative SD (%)
Formaldehyde				
Front	25	109	20	18
Back	23	101	22	21
Acetaldehyde				
Front	26	84	19	22
Back	23	78	9	12
Acetone				
Front	25	86	19	23
Back	21	91	21	23

Note: 0.01 to 0.50 µg in the carbonyl form were added to each section.

Appendix Table B.8. VOCs (ng) and Aldehydes (µg) in Lab Blanks and Field Blanks and LODs

Compound Name	Lab Blanks			Field Blanks			
	N	Mean	SD	N	Mean	SD	LOD
1,3-Butadiene	33	0.02	0.10	49	0.01	0.08	0.24
2-Methylpentane	33	0.15	0.34	49	0.33	0.73	2.53
2-Methylhexane	33	0.05	0.18	49	0.07	0.27	0.88
3-Methylpentane	33	0.07	0.23	49	0.12	0.38	1.27
2,3-Dimethylpentane	33	0.02	0.12	49	0.13	0.53	1.72
2,2,4-Trimethylpentane	33	0.07	0.16	49	0.19	0.55	1.85
Methylcyclohexane	33	0.00	0.00	49	0.04	0.16	0.51
Benzene	33	1.89	1.99	49	2.29	2.17	8.81
Toluene	33	1.25	1.79	49	1.33	1.15	4.79
<i>m&p</i> -Xylenes	33	0.88	2.06	49	0.86	1.24	4.57
<i>o</i> -Xylene	33	0.11	0.27	49	0.16	0.27	0.98
Ethylbenzene	33	0.12	0.25	49	0.15	0.20	0.75
Styrene	33	0.28	0.44	49	0.37	1.00	3.36
α -pinene	33	0.08	0.25	49	0.01	0.08	0.26
D-limonene	33	0.00	0.00	49	0.00	0.00	0.00
MTBE	33	0.02	0.10	49	0.02	0.09	0.29
Formaldehyde	34	0.03	0.02	47	0.05	0.06	0.22
Acetaldehyde	34	0.03	0.01	47	0.03	0.04	0.13
Acetone	34	0.13	0.07	47	0.12	0.16	0.60

Note: LOD = $3 \times$ the SD of field blanks + mean blank value.

APPENDIX AVAILABLE ON THE WEB

Appendix C contains supplemental material not included in the printed report. It is available on the HEI Web site, at <http://pubs.healtheffects.org>.

Appendix C. Consent Form, Driver Questionnaire, and Post-Shift Questionnaire

ABOUT THE AUTHORS

Dr. Thomas J. Smith was the principal investigator for this study. He is a professor of industrial hygiene at the Harvard School of Public Health, Boston, Massachusetts, and the director of the industrial hygiene training program. His primary research interest is development of biologically based exposure assessments for epidemiologic studies. He is directing the diesel exposure assessment of the NCI Trucking Industry Particle Study, which is designed to determine cancer risks per unit of exposure. He is also directing a study of 1,3-butadiene metabolism in a laboratory study of human exposures using physiologically based toxicokinetic modeling to estimate personal rates of metabolism. He earned his M.P.H., M.S., and Ph.D. in environmental health from the University of Minnesota, Minneapolis, Minnesota.

Dr. Mary E. Davis was a co-investigator for this study. She is an associate professor in the Department of Urban and Environmental Policy at Tufts University, Medford, Massachusetts. Her research interests are in using economic data to estimate historical exposures in the trucking industry and in risk and policy assessment for environmental hazards. She was a postdoctoral fellow in biostatistics and environmental health at the Harvard School of Public Health. She earned her Ph.D. in economics from the University of Florida, Gainesville, Florida.

Dr. Jaime E. Hart was a co-investigator for this study and was also the project manager for the study and for the NCI Trucking Industry Particle Study. Her research interests are in occupational and environmental epidemiology. She earned her Sc.D. from the Harvard School of Public Health, Boston, Massachusetts.

Andrew Blicharz was a research assistant in the Exposure, Epidemiology and Risk Program at the Harvard School of Public Health, Boston, Massachusetts, during this study. He was also the field team leader for the study and for the NCI Trucking Industry Particle Study. He earned his B.S. in biology from Bates College, Lewiston, Maine.

Dr. Francine Laden was a co-investigator for this study. She is an associate professor of environmental epidemiology at the Harvard School of Public Health and an associate professor of Medicine at the Channing Laboratory, Brigham and Women's Hospital and Harvard Medical School, all of Boston, Massachusetts. Her research interests are in the environmental epidemiology of cancer and respiratory disease. Her current research is focused on analyses of the relationships between organochlorines and non-Hodgkin lymphoma and Parkinson disease, between traffic exposures and lung cancer and cardiovascular mortality in the NCI Trucking Industry Particle Study, and between ambient air pollution and cardiopulmonary mortality in the Nurses' Health Study and the Harvard Six Cities Study. She earned her M.S. in environmental health management and her Sc.D. in epidemiology from the Harvard School of Public Health, Boston, Massachusetts.

Dr. Eric Garshick was a co-investigator for this study. He is an associate professor of medicine at the Harvard Medical School, Boston, Massachusetts. He is also the associate chief of the Pulmonary and Critical Care Medicine Section, VA Boston Healthcare System, and a physician at Channing Laboratory, Department of Medicine, Brigham and Women's Hospital, all of Boston, Massachusetts. His research interests are the health effects of diesel exhaust exposure and traffic-related emissions and the epidemiology of chronic lung disease. He earned his M.D. from Tufts University School of Medicine and his M.P.H. from the Harvard School of Public Health, Boston, Massachusetts.

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ABBREVIATIONS AND OTHER TERMS

BEAM	Boston Exposure Assessment in Microenvironments	GM	geometric mean
BTEX	benzene, toluene, ethylbenzene, and xylenes	GPS	global positioning system
CO ₂	carbon dioxide	GSD	geometric standard deviation
DNPH	2,4-dinitrophenylhydrazine	HPLC	high-pressure liquid chromatography
EC	elemental carbon	ICT	industrial, commercial, and transportation
EPA	U.S. Environmental Protection Agency	IR	Investigators' Report
GC	gas chromatography	LH	long haul
GC–MS	gas chromatography–mass spectroscopy	LOD	limit of detection
GC–MSD	gas chromatography–mass selective detector	MATES	Multiple Air Toxics Exposure
GIS	geographic information system	MSAT	mobile-source air toxic
		MTBE	methyl <i>tert</i> -butyl ether
		NCI	National Cancer Institute
		NHANES	National Health and Nutrition Examination Survey
		NO _x	nitrogen oxides
		OC	organic carbon
		P&D	pickup and delivery
		PAH	polycyclic hydrocarbons
		PAKS	Personal Aldehydes and Ketones Sampler
		PID	photo-ionization detector
		PM	particulate matter
		PM ₁	PM with an aerodynamic diameter ≤ 1.0 μm
		PM _{2.5}	PM with an aerodynamic diameter ≤ 2.5 μm
		QA–QC	quality assurance–quality control
		RFA	Request for Applications
		RIOPA	Relationships of Indoor, Outdoor, and Personal Air
		SD	standard deviation
		TWA	time-weighted average
		UATMP	Urban Air Toxics Monitoring Program
		UV	ultraviolet
		USGS	U.S. Geological Survey
		VOC	volatile organic compound

Research Report 172, *Potential Air Toxics Hot Spots in Truck Terminals and Cabs*, Smith et al.

INTRODUCTION

Motor vehicles and other combustion sources emit many air toxics whose ambient concentrations are not regulated by the U.S. Environmental Protection Agency (EPA*) but that, with sufficient exposure, are known or suspected to cause adverse human health effects. Although some state and local government agencies have performed limited monitoring of air toxics, characterization of ambient concentrations of, and personal exposures to, air toxics has been challenging, in part because of the low ambient concentrations of the individual compounds. HEI has had a longstanding commitment to improving methods for measuring selected air toxics and understanding the resulting exposures and health effects.

The Preface that accompanies this Research Report describes the regulatory actions the EPA has taken to control emissions of air toxics in general and of mobile-source air toxics (MSATs) specifically. As the Preface makes clear, however, better characterization of exposures to air toxics should be undertaken — especially at sites of possible high exposures — before conducting health effects studies. Thus, in 2003, HEI issued Request for Applications (RFA) 03-1, “Assessing Exposure to Air Toxics,” to support research to identify and characterize exposures to air toxics at so-called hot spots, areas where concentrations of one or more air toxics, and exposure of the populations in these areas, are expected to be “higher than those to which the broader public is exposed.” Such areas may be in proximity to one or more pollution sources or may be affected by transient or sustained localized conditions that lead to elevated concentrations of some pollutants.

Dr. Thomas Smith’s 3-year study, “Air Toxics Hot Spots in Industrial Parks and Traffic”, began in January 2004. Total expenditures were \$1,091,100. The draft Investigators’ Report from Smith and colleagues was received for review in August 8, 2007. A revised report, received in September 2010, was accepted for publication in October 2010. During the review process, the HEI Health Review Committee and the investigators had the opportunity to exchange comments and to clarify issues in both the Investigators’ Report and the Review Committee’s Critique.

This document has not been reviewed by public or private party institutions, including those that support the Health Effects Institute; therefore, it may not reflect the views of these parties, and no endorsements by them should be inferred.

* A list of abbreviations and other terms appears at the end of the Investigators’ Report.

In response to this RFA, Dr. Thomas Smith, of the Harvard School of Public Health, and his colleagues submitted an application, “Air Toxics Hot Spots in Industrial Parks and Traffic,” proposing to examine areas in and around truck terminals to identify potential MSAT hot spots. The study would add on to a then-ongoing study of the trucking industry funded by the National Cancer Institute (NCI) to evaluate the associations between lung cancer mortality and exposure to particulate matter (PM) $\leq 2.5 \mu\text{m}$ in aerodynamic diameter (PM_{2.5}), elemental carbon (EC), total carbon, and polycyclic aromatic hydrocarbons (PAHs) (see Smith et al. 2006).

The HEI Research Committee thought that the proposed study would provide a rich source of data on pollutant concentrations in areas around truck terminals and on in-vehicle exposures. In addition, the Committee thought that the linkage with the ongoing NCI study was a strength in that the terminals had already been selected and sampled, so the study could start right away.

This Critique is intended to aid the sponsors of HEI and the public by highlighting both the strengths and limitations of the study and by placing the Investigators’ Report (IR) into scientific and regulatory perspective. A description of other HEI studies funded under RFA 03-1 can be found in the Preface.

SCIENTIFIC BACKGROUND

Air toxics are a large and diverse group of compounds that are generated by multiple sources; understanding exposures to and the effects of air toxics generated by mobile sources is of particular concern to the scientific community and the EPA. Summary information about the concentrations of numerous air toxics — including benzene, acetaldehyde, and formaldehyde, as well as naphthalene and several other PAHs — in various microenvironments relevant to the current study can be found in HEI’s special report on MSATs (HEI Air Toxics Review Panel 2007).

When this study was started, in 2004, it was known that the concentrations of pollutants to which people are exposed vary significantly across locations and microenvironments and that the concentrations of several pollutants (including carbon monoxide, nitrogen dioxide, EC, and

benzene) measured on roads or in close proximity to them decrease with distance from the road. The decay gradients are affected by meteorologic conditions (especially wind direction) and other factors. These studies were included in a review by Zhou and Levy (2007). At the time, very limited information existed about the decay gradients of MSATs.

AIR TOXICS MONITORING PROGRAMS

In the United States, various federal and state agencies have developed programs to characterize exposures to air toxics and define potential hot spots. The EPA started the Urban Air Toxics Monitoring Program (UATMP) in 2001 to characterize the composition, and the concentrations of the components, of toxic air pollutants in or near urban areas. The program includes 24-hour measurements of 75 compounds typically made every 6 or 12 days at 46 sites throughout the United States. The location of the sampling sites was chosen by local agencies and includes commercial, industrial, and residential areas in urban, suburban, and rural settings. The data collected are available in yearly final reports. The 2005 measurements (U.S. EPA 2006a) are included in the current study's IR for comparison with the measurements obtained by the investigators. As indicated in the EPA report (U.S. EPA 2006a), "chemical concentrations measured during the 2005 UATMP varied significantly from monitoring site to monitoring site. As discussed throughout this report, the proximity of the monitoring locations to different emissions sources, especially industrial facilities and heavily traveled roadways, often explains the observed spatial variations in ambient air quality." California's South Coast Air Quality Management District set up an air toxics evaluation program referred to as Multiple Air Toxics Exposure Study (MATES) I, II, and III to quantify the population exposure risk from existing sources of select air toxics starting in the 1980s and continuing until 2005. A new phase of MATES started in 2012. The monitoring component of the program included fixed sites to characterize concentrations at the neighborhood scale and microscale monitoring sites selected to "determine whether localized source emissions cause a significant increase in the concentration of certain toxic air contaminants" (also referred to as "local hot spots") (South Coast Air Quality Management District 2000).

To better define the relationships among indoor, outdoor, and personal exposure concentrations of PM_{2.5} and other air toxics, HEI and the then Houston-based National Urban Air Toxics Research Center co-funded the Relationships of Indoor, Outdoor, and Personal Air (RIOPA) study (Weisel et al. 2005; Turpin et al. 2007). The RIOPA study was conducted in three cities with different weather conditions and air pollution source profiles — namely, Los Angeles,

California, which is dominated by mobile sources; Houston, Texas, which is dominated by large industrial stationary (point) and area sources (with a portion contributed by mobile sources); and Elizabeth, New Jersey, which has a mixture of mobile, point, and area sources. Measured concentrations of air toxics were highly variable for all air toxic species within and across the three cities (Weisel et al. 2005).

CHANGES IN MOTOR VEHICLE EMISSIONS

At the time current study started, in mid-2004, the motor vehicle fleet on U.S. roads had higher emission rates than the current (2012) fleet, because several regulations have been implemented since 2004 and because automotive technology has continued to improve. EPA regulations to reduce exhaust emissions of PM and nitrogen oxides (NO_x) from light-duty vehicles and trucks (both gasoline and diesel-powered) were phased in beginning with model-year 2004 vehicles and were fully phased in by 2009 as part of Tier 2 standards (U.S. EPA 2000). To achieve these emissions reductions and enable the introduction of improved emission-control technologies, the EPA mandated the use of low-sulfur diesel fuel, which became widely available by mid-2006. In 2007, in addition, as part of an ongoing program to reduce emissions of MSATs, the EPA set more stringent exhaust and evaporative hydrocarbon controls and mandated a reduction in the benzene content of gasoline (U.S. EPA 2007). NO_x emission standards for heavy-duty diesel engines were phased in starting with model year 2004 (U.S. EPA 1997) and fully implemented to achieve a 95% reduction compared with pre-2004 standards starting with model year 2010 (U.S. EPA 2001b). PM standards (reducing PM emissions by 90% compared with models from previous years) were phased in starting with model year 2007 (U.S. EPA 2001b). It was expected that the fuel changes and technologies used for reducing PM emitted from diesel vehicles (such as an oxidation catalyst and catalyst-coated PM filters) would effectively reduce emissions of several MSATs (U.S. EPA 2001a). As a result, the current fleet of motor vehicles has lower pollutant emissions than that in use when this study was conducted.

APPROACH AND SPECIFIC AIMS

The main goal of the study was to measure concentrations of selected volatile organic compounds (VOCs) and PM in locations with potentially high levels of air pollution that could make them hot spots for human exposure, that is, at locations around the perimeter of terminals for pick-up and delivery trucks and in truck cabs during daily

runs. The premise underlying the selection of the sampling sites was that locations upwind of the terminals would have lower concentrations than downwind locations. The investigators hypothesized that the upwind locations' concentrations would be influenced by "industrial parks and other commercial zones" while the downwind locations' concentrations would reflect the added contribution from truck traffic inside the terminal and could be representative of exposures in nearby downwind neighborhoods.

The investigators had access to the terminals as part of a then-ongoing NCI-funded study that involved truck drivers, loading-dock workers, and mechanics at 36 truck terminals chosen randomly from major metropolitan areas across the United States (for a description of the NCI study see IR Appendix A and Smith et al. 2006). The NCI study included area and personal measurements of PM_{2.5}, PM ≤ 1.0 μm in aerodynamic diameter (PM_{1.0}), and PM_{1.0} components (EC, organic carbon [OC], and other organic markers of diesel exhaust) in the truck yards and inside the truck cabs (described in IR Appendix A and Smith et al. 2006). At the time of the authors' application to HEI, 15 terminals remained to be visited for exposure assessment. For these terminals, concurrent measurements of air toxics were added. This is referred to as Phase 1 in the IR. The order of sampling the terminals was randomized. Phase 2 consisted of repeat visits to six of the terminals already visited in Phase 1 to make additional measurements.

The specific aims of the study were:

1. To modify the sampling system for PM_{2.5} and add integrated VOC collection capabilities for selected hydrocarbons and aldehydes.
2. To measure the time-weighted average exposure intensity and variation of VOC components by location characteristics at truck terminals across the United States, focusing on three potential exposure hot spots: (a) concentrations nominally upwind of terminals, (b) concentrations nominally downwind of terminals, and (c) in-cab personal exposures of truck drivers.
3. To examine the relationships between VOC exposures and the levels and composition of particulates upwind of trucking activities, downwind of trucking activities, and within truck cabs.
4. To determine the variation in VOC composition and exposure intensity associated with a mix of sources in industrial parks, downwind neighborhoods, and in vehicles observed in the source-apportionment measurements. As part of this aim the authors also conducted analyses to determine the sources of the VOCs.

5. To develop a geographic information system (GIS)-based statistical modeling method that could deal with both the spatial and temporal dimensions of the data.

STUDY DESIGN

The investigators measured VOCs (hydrocarbons and carbonyls) and PM_{2.5} at various locations within each truck terminal:

- At the upwind fence line (also referred to as "terminal background") and downwind fence line of the terminal perimeter at 15 terminals (with repeat visits to six).
- In the docks and repair shops (of the six repeat-visit terminals). Measurements in these two indoor locations were added in Phase 2 of the study (see below) and were not part of the specific aims.

In addition, sampling was conducted in truck cabs during daily pick-up and delivery trips (for a total of 36 trips). Both smoking and nonsmoking drivers were recruited. Smoking status was assessed by means of a questionnaire.

Sampling was conducted as follows:

- Phase 1 overlapped with the sampling done for the NCI study and entailed consecutive 12-hour integrated sampling periods at the upwind and downwind fence-line locations and in the truck cabs for five days in a row at each terminal. The goal was to characterize the concentrations of air toxics, determine the upwind contribution, and evaluate the sources and factors that contributed to the measured concentrations. Phase 1 was conducted between December 2003 and March 2005. Wind direction was predicted before each sampling trip using an online weather information source. Once on location, the investigators placed a weather station downwind of each terminal for a few hours of observations. The upwind and downwind sites were selected on the basis of these data.
- Phase 2 entailed a repeat five-day visit to six of the 15 terminals examined in Phase 1, at approximately the same month as in Phase 1. It was conducted in 2005. (A criterion for selection was dry climate because a loss of aldehyde samples was observed at sites with high humidity.) Time-integrated sampling was repeated at the upwind and downwind locations. Air sampling was added in the docks and shops. Continuous sampling for total VOCs and PM_{2.5} was added at each of the four primary wind directions to allow more flexibility in classifying upwind or downwind locations during sampling. Continuous sampling was

also added in the cabs of trucks equipped with a global positioning system (GPS) unit to allow correlation of the exposure measurements with the route characteristics. (It appears that these measurements were made in two trucks.) Phase 2 focused on defining the within- and between-terminal variability and relationships between the shops and the docks and the upwind measurements, on identifying the VOC sources, and on obtaining better measures of the temporal relationships between measured concentrations and wind direction as well as more detailed data on in-cab exposures and the variables that might affect the pollutant levels.

CHARACTERISTICS OF THE TERMINALS

The 15 terminals sampled for the study were located in 13 states across the United States. Because the larger group of truck terminals (i.e., in the NCI-funded study) was a random sample and the order of sampling was randomized, the 15 terminals sampled in the current study can also be considered a random sample. The majority of the terminals were near one or more interstate highways and industrial areas (see IR Table 2). However, the distances to the highways differed across the terminals. Two of the terminals were located within 500 meters of a highway, three within 1000 meters, and five were within 1000 and 2000 meters; five were more than 2000 meters away.

A schematic representation of what the investigators referred to as a “model” terminal location is provided in IR Figure 2. However, there was little information in the report about land use around the terminals sampled apart from the percentage of industrial land use within 1 kilometer of the terminals. The percentage of industrial land use (designated as “industrial, commercial, and transportation” by the U.S. Geological Survey) around the terminals ranged from 6% to 92%, with a median of 25%. Other types of land use were not reported.

METHODS

For integrated sampling of air toxics, Smith and colleagues used a triple-sorbent tube for hydrocarbons followed by gas chromatography–mass spectrometry and a 2,4-dinitrophenylhydrazine–based cartridge for carbonyls followed by high-pressure liquid chromatography. Continuous total VOC measurements were taken with a photoionization detector (PID). This instrument has different sensitivity to the various VOCs (as shown in IR Table 3).

The compounds measured with the integrated samplers are listed below. The compounds in *italics* were those targeted in the original RFA. Acrolein, crotonaldehyde, and naphthalene were also listed in the RFA but could not be measured with the samplers chosen.

- Hydrocarbons: *1,3-butadiene*, aromatic compounds (*benzene, toluene, xylenes, ethylbenzene, and styrene*), alkane compounds (*n-hexane, trimethylpentane, dimethylpentane, 2-methylhexane, methylpentane, 3-methylhexane, and methylcyclohexane*);
- *Methyl tert-butyl ether (MTBE)*; and
- Carbonyls: aldehydes (*formaldehyde* and *acetaldehyde*) and acetone.

PM_{2.5} was characterized for mass as part of the NCI study by gravimetric analysis; EC, OC, and a larger number of organic species were characterized in personal samples and terminal locations (see IR Tables 14 and 15). Continuous mass measurements were made using a DustTrak PM_{2.5} aerosol monitor.

Wind direction at each terminal was obtained from the weather stations, which were placed at the least obstructed of the fence-line sites.

DATA ANALYSIS

Smith and colleagues use several statistical approaches in analyzing their data:

1. Descriptive analysis. Descriptive analyses of the integrated and continuous monitoring data were conducted as follows:
 - a. The time-integrated upwind and downwind measurements were summarized as means, medians, and standard deviations (SDs) for each compound across all terminals. Downwind contributions were expressed as ratios between the two mean measurements for each terminal.
 - b. The time-integrated measurements at the docks and shops were similarly summarized and compared with those at the upwind locations.
 - c. The continuous measurements (5-minute averages) were graphed over the 12-hour monitoring periods. The terminal contributions were estimated from the difference between the downwind and upwind 5-minute averages for each session. The data were also combined over all the sessions to characterize trends in upwind–downwind concentrations.
2. Structural equation modeling. Analysis by means of structural equation modeling was the primary statistical

tool and was used to identify the indirect effects of intermediate variables (including temperature, wind speed, distance of the terminal to an interstate highway, and regional census variables) on primary dependent variables, which for air toxics were the concentrations of 1,3-butadiene, benzene, toluene, and formaldehyde at the upwind locations and indoor work area.

3. Principal component analysis. Principal component analysis was applied to the upwind measurements to identify the sources of PM, VOCs, and aldehydes at the six terminals visited again in Phase 2 (using the measurements made in both Phase 1 and Phase 2). The ratios of different components of BTEX (benzene, toluene, ethylbenzene, and xylenes) were determined to evaluate the proximity of the terminals to traffic emissions. The authors focused on the benzene-to-toluene ratio because of the different reactivity of the two species (with toluene being more reactive than benzene).
4. Geographic analysis. Land-use data, GIS data, and GPS data for the locations of the terminals and for the specific routes taken by various drivers were used for some analyses. GIS was used to identify land uses and roads around the terminals. GPS tracking of truck routes was used to match the real-time in-cab measurements of pollutant concentrations to specific locations.

DATA QUALITY

Smith and colleagues provided, in an appendix, information on limits of detection (LODs), blanks, duplicate samples, and sample recovery. Air concentrations were calculated after correction for the efficiency of sample recovery and subtraction of field blanks. Four internal standards were used for hydrocarbon analysis; no information was provided for aldehyde internal standards. Recovery was greater than 83% for all species of VOCs and 79% for 1,3-butadiene. Recovery was between 78% and 84% for acetaldehyde and between 101% and 109% for formaldehyde. Sample losses were 36% for the aldehyde samples and 15% for the VOC samples at the yard locations. Not all terminals had the same number of sample analyses; at six terminals, four or fewer samples were analyzed. Duplicate samples were collected to determine repeatability.

The LOD was calculated as three times the SD of the mean of the field blanks plus the mean blank value. The method was different for 1,3-butadiene and MTBE, for which spiked lab blanks were used.

RESULTS

PHASE 1

Pollutant Concentrations at Upwind and Downwind Locations

The ratios of the mean downwind and upwind concentrations for various pollutants by terminal indicated that, overall, there was little or no difference between the concentrations at the two locations. The ratios showed wider ranges for VOCs than for aldehydes and PM_{2.5}. The investigators acknowledged that wind directions were not constant during the 12-hour sampling periods and that this probably contributed to reducing the differences between the upwind and downwind locations.

The results of the measurements at terminal upwind and downwind locations showed that the VOCs present at the highest concentrations in all locations were toluene and formaldehyde, followed by acetaldehyde and acetone (see IR Table 7).

Concentrations at terminal upwind locations were generally lower than those at indoor locations. The two indoor locations differed in pollutant concentrations; the shops had higher concentrations of xylenes, alkanes, acetone, and PM than the docks, and the docks had higher concentrations of 1,3-butadiene, benzene, and carbonyls (shown in IR Table 8). All the alkane and aromatic hydrocarbon concentrations at the yard upwind locations and at the loading docks were weakly correlated with EC ($r = 0.54$ – 0.62). In the shops the correlations were more variable (see IR Table 9) and generally lower. A low correlation is generally considered indicative of different pollutant sources.

Source Characteristics

Source apportionment analysis of the upwind measurements across the six repeat-visit terminals identified three factors that explained 80% to 92% of the total variability: a primary factor consisting of alkanes and aromatics (responsible for 46% to 75% of the variability) and two smaller factors, one consisting of formaldehyde, acetaldehyde, and sometimes acetone, and the other consisting of varying components (sometimes formaldehyde and benzene, sometimes alkanes and styrene). The primary factor was attributed to traffic sources upwind of the terminals. The aldehydes in factor two and sometimes in factor three were considered to be the products of photochemical reactions. The benzene and alkanes were considered to be contributed by gas stations and auto-body shops.

The ratios of median concentrations of BTEX components at the upwind locations were similar to those found along major roadways in other studies (see IR Tables 16 and 17). The investigators found a toluene-to-benzene ratio of 3.3 for the upwind locations, 3.1 for the truck drivers, and 4 for the work areas. Based on their previous work, the authors stated that a ratio of approximately 2 was expected for sampling locations near traffic sources (Smith et al. 2001).

Results of Structural Equation Modeling

Higher temperatures were associated with higher concentrations of formaldehyde and lower concentrations of 1,3-butadiene. Wind speed was inversely correlated with the concentrations of all four pollutants. Distance to an interstate highway was significantly and inversely associated only with toluene and benzene (formaldehyde also decreased with increasing distance, but not statistically significantly). An analysis by census regions (i.e., Midwest, Northeast, and West) showed much variability across the regions, with higher concentrations of benzene in the West and of formaldehyde in the Northeast; the reason for this regional pattern was not clear.

Elevated upwind concentrations of benzene and 1,3-butadiene were associated with elevated concentrations at the docks and, to a lesser extent, in the shops.

In-Cab Measurements

The concentrations of benzene, MTBE, styrene, and hexane measured in the cabs of the nonsmoking drivers (the majority of the drivers studied) were higher on average than those measured at the upwind locations and indoor work locations. There were some differences between nonsmokers and smokers in the in-cab concentrations of VOCs and carbonyl species.

The correlations of in-cab concentrations of EC and PM_{2.5} with individual VOCs and carbonyls (shown in IR Table 20) were generally poor, with r 's < 0.5 (for non-smoking drivers). The lowest correlations were for aldehydes, acetone, and hexane, suggesting that the species originated from sources different from those of the PM_{2.5}.

Analysis of the effects of open or closed windows on in-cab concentrations showed that when the windows were "predicted to be open" there were significantly lower concentrations of aldehydes and higher concentrations of PM_{2.5} and 1,3-butadiene (data not shown). Whether the windows were open or closed was predicted based on in-cab carbon dioxide (CO₂) concentrations.

The authors mentioned that in-cab exposures did not vary much between trucks of different production years or models but that EC concentrations were correlated with truck age.

PHASE 2

Real-Time Upwind and Downwind Concentrations

The continuous PM_{2.5} and total VOC data from the four fence-line locations per terminal at the six repeat-visit terminals were used to determine terminal contributions downwind of the terminal over time. Here, unlike the time-integrated results in Phase 1, real-time analyses combining data from all six terminals showed significant upwind-to-downwind differences for about 60% of the sessions. An example of these differences during one session is provided in IR Figure 7.

Real-Time In-Cab Measurements

From the analyses of the real-time measurements of PM and VOCs during work shifts in conjunction with GPS data, the investigators were able to trace the concentrations of these pollutants in space and time. They noted that the highest values occurred when a truck was in a terminal, stopped in traffic, or at a delivery site.

Repeat-Visit Analysis

The comparison of terminal upwind measurements for the first and second visits to the six repeat-visit terminals showed that the concentrations were more stable at the loading dock than at the upwind sites, where the differences were more pronounced (with several species significantly higher and some significantly lower) (see IR Table 23). The structural equation modeling analysis showed that meteorologic differences (such as differences in wind speed, temperature, or humidity) played important roles in the differences between the two visits.

COMPARISONS WITH OTHER STUDIES FOR HOT-SPOT DETERMINATION

The investigators made various comparisons to assess whether terminal areas were potential exposure hot spots and their conclusions depended on the comparisons being made. They compared the mean or median concentrations measured at the terminal upwind locations with those measured by the EPA air toxics monitoring network (shown in IR Table 10), those reported in various exposure studies conducted in urban areas in the United States and abroad (see IR Table 11), and the EPA's screening values for noncancer and cancer risk (see IR Table 26), which the Agency developed as a screening methodology for identifying "chemicals and geographic locations that should be the focus of more rigorous risk evaluation" (U.S. EPA 2006b). The values were calculated using the unit risk value for cancer-causing compounds and the inhalation

reference concentration for noncancer health effects for all compounds as a starting point, with various corrections that resulted in more conservative (i.e., health protective) values. The EPA explains that a “screening value is used to indicate a concentration of a chemical in the air to which a person could be continually exposed for a lifetime (assumed to be 70 years) and which would be unlikely to result in a deleterious effect (either cancer or noncancer health effects)” (U.S. EPA 2006b).

The authors report that the means and medians of upwind concentrations were very similar to the mean concentrations of pollutants (which included many of the air toxics measured in the current study) measured by the EPA across all air toxics monitoring sites, with the exception of formaldehyde, which was lower in the current study. The comparison with the concentrations measured by other investigators (IR Table 11) focused on measurements obtained in the three RIOPA cities (Weisel et al. 2004), inner-city neighborhoods in Minneapolis (Adgate et al. 2004), in Los Angeles and New York City (Sax et al. 2004; Kinney et al. 2002), and Brisbane (Hawas et al. 2002). The authors noted that the 1,3-butadiene and aldehyde concentrations were similar and that all the aromatics (such as benzene, toluene, and xylenes) were lower in the current study. They concluded from all these comparisons that there was “no evidence of a toxic hot spot.”

As for the comparison with the EPA screening values, the investigators found that 100%, 93%, 61%, and 6% of the upwind mean concentrations of formaldehyde, acetaldehyde, 1,3-butadiene, and benzene, respectively, exceeded the screening values for cancer risk. They concluded that “all three types of testing sites — upwind and downwind fence-line locations and inside truck cabs while in heavy traffic — met the established definition for a hot spot by having concentrations of pollutants that exceeded the EPA’s screening values.”

HEI HEALTH REVIEW COMMITTEE EVALUATION

In its independent review of the study, the HEI Health Review Committee thought that a major strength of the study was to document concentrations of air toxics in various environments in and around truck terminals and inside truck cabs. The Committee noted that the terminals were not selected to meet the initial hypothesis of there being industrial areas upwind of the terminals and neighborhoods downwind of the terminals. In addition, the upwind location was defined operationally as being upwind with respect to the prevailing wind direction, whereas in fact wind direction proved to be variable over

the course of a day and from day to day. The Committee noted that the determination of prevailing wind direction requires collecting data for longer periods of time but acknowledged that this would have not been feasible for the current study.

The Committee agreed with the investigators’ main conclusion about the relation between the upwind–downwind concentrations, that is, that “the downwind fence-line concentrations near the terminals, on average, were not significantly elevated compared with the upwind concentrations in the 12-hour [time-weighted average] samples.” The Committee agreed that this was likely related to the fact that the influence of shifting wind directions and speeds could not be adequately taken into account. With time-integrated sampling the analyses of the continuous total VOC measurements made during Phase 2 provided a more detailed pattern of concentration variations in relation to changes in wind directions. Although these data were analyzed only to a very limited extent in relation to actual wind direction and were limited to total VOCs, they pointed to the importance of wind direction in determining the impact of pollutant sources and to the existence and variability of localized elevated pollutant levels that could affect human health.

The source apportionment data indicated that the major source of the VOCs was traffic, as would be expected from truck emissions in and around truck terminals. The use of structural equation modeling to test relationships between the various measured variables was innovative.

With regard to the in-cab measurements, the Committee agreed that the average concentrations of benzene, MTBE, styrene, and hexane inside the cab during a work shift were higher than those at the upwind sites and the indoor terminal locations. Based on the differences between windows opened and closed, the authors suggest that some of the pollutants (such as aldehydes) are generated within the truck’s own cab, and that others originate from the surrounding traffic. The Committee thought that this explanation was plausible. The finding of the structural equation modeling that whether cab windows were open or closed was the major determinant of the elevated concentrations of some of the pollutants inside the cab is not novel. The Committee noted that the investigators determined whether or not the windows were open based on CO₂ concentrations inside the cabs, which provided an indication of air exchange regardless of the mechanism (window status or ventilation system), but they did not directly monitor the degree of ventilation.

DATA QUALITY

Overall, the measurement methods employed were appropriate for most of the pollutants measured. However, the method for sampling carbonyls, which used 2,4-dinitrophenylhydrazine as the sorbent, was not adequate for measuring acrolein and crotonaldehyde (Zhang et al. 2000), two important MSATs. The continuous VOC monitor provides a value for total VOCs but has a variable sensitivity to the species measured. Thus, the Committee agreed with the investigators' statement that the PID monitor's "total VOC measurements were not equal to the sum of the weighted individual measured components."

The quality of the data the investigators collected was good. Spiked samples were used to determine and correct for recovery; however, details about the internal standards used to determine the recovery of carbonyls and when they were added to the procedure were not provided.

The LODs were determined for most compounds using the SDs of the field blanks, which is a common and appropriate practice. Paradoxically, for some VOCs (toluene and *m&p*-xylanes) the lab blanks actually had higher SDs than the field blanks; if the lab blanks had been used to calculate the LODs for toluene and *m&p*-xylanes, they would have been 6.6 and 7.3 ng, respectively, rather than 4.8 and 4.6 ng. The percentage of values below the LOD was not provided. However, the report mentioned that a "large number" of 1,3-butadiene yard samples were below the LOD.

HOT-SPOT DETERMINATION

The Review Committee considered whether the fence lines of the truck terminals studied in the current study were hot spots for air toxics based on the comparisons made by the authors. The Committee thought the comparison with the EPA screening values, their primary method for answering this question, was not a very discriminating criterion for hot spot determination, because these screening values (which are based on compound unit risk estimates for chronic exposures with an added safety factor) can be often exceeded in many urban areas as can be observed by comparing the mean concentrations measured at the EPA air toxics monitoring sites (reported in IR Table 10) with the EPA screening values. The Committee thought that comparing the measurements made in this study with those made by others in urban locations and inner-city neighborhoods was more meaningful; such comparisons did not support defining the terminals as hot spots. However, the Committee noted that comparing measurements across studies can be problematic, because of the different sampling and analysis methods, protocols, types of sampling sites, and meteorologic conditions.

Finally, as planned, the investigators compared the fence-line upwind measurements with the downwind measurements, primarily using time-integrated measures. This comparison was useful, but because of the difficulty in accounting for meteorologic conditions, it turned out to be not very informative.

The Committee thought that a limitation of the study as a hot-spot study was the lack of parallel measurements at background sites (i.e., sites that were at an appropriate distance from the terminals and not impacted by local sources) and of discussion of the local context of the terminals (such as the quality and quantity of the sources within the industrial parks in which the truck terminals were embedded and the source strength of the terminals themselves). Although the Committee thought that the assessment of the variations in pollutant concentrations within the terminals was informative, it did not think that the upwind site at each terminal represented an adequate "regional" background site (as these sites are sometimes referred to in the IR) and cautioned against interpreting the upwind concentrations as representative of regional background concentrations.

The continuous and time-weighted average measurements in the truck cabs did document elevated concentrations of a range of components compared with the fence-line measurements. Although the investigators concluded that "driving in heavy urban traffic can put someone in a local hot spot of traffic emissions," the Committee thought that these should be considered occupational exposures because they were measured during activities related to the pick-up and delivery of goods over a work shift. In addition, the Committee did not think the term "hot spot" should be used for in-vehicle exposures. Many studies have documented that being in traffic on busy roads leads to increased exposures to traffic-related air pollutants, and the Committee considered that this part of the study contributed to the literature on in-traffic exposures, rather than to the literature on hot spots — specific areas with increased concentrations of criteria pollutants and toxic air pollutants.

CONCLUSIONS

In summary, Smith and colleagues collected and analyzed detailed data on concentrations of air toxics in various environments in and around truck terminals and in truck cabs. They used appropriate methods for data collection and analysis and paid attention to data-quality issues. They focused on measuring VOC concentrations at two sites around the fence line of each terminal, with one site

operationally defined as being upwind and the other as being downwind of the terminal. They hypothesized that concentrations at the upwind sites would be affected by upwind commercial and industrial activities and that the downwind sites would reflect the added contribution of the terminal and could be representative of the exposure in downwind neighborhoods. The Committee agreed with the investigators' main conclusion about the relation between the upwind and downwind concentrations, that is, that "the downwind fence-line concentrations near the terminals, on average, were not significantly elevated compared with the upwind concentrations in the 12-hour [time-weighted average] samples." The Committee agreed that this was likely related to the fact that the influence of shifting wind directions and speeds could not be adequately taken into account.

The investigators made several comparisons for hot-spot determination and concluded that the terminals were hot spots when compared with EPA screening values. In the Committee's view, comparison using this criterion is problematic, however, as noted above, because the screening values are often exceeded in many urban areas. Comparisons with measurements reported in other studies did not support defining the terminals as hot spots, and comparison of upwind and downwind measurements also showed little or no difference, because of shifting wind patterns. Measurements at appropriately selected background sites would be needed to establish exactly how "hot" the terminal fence-line locations were at any given time. Overall, the Committee noted that the study does not provide conclusive evidence as to whether the truck terminals were pollution hot spots, but pointed out the existence and variability of localized elevated pollutant levels that could affect human health. The measurements represent potential exposures of workers who work at the terminals frequently and for prolonged periods of time.

The authors also measured VOC concentrations inside the truck cabs during a work shift and concluded that driving in traffic was a hot spot. The Committee agreed that concentrations in the truck cabs were higher than those measured at the terminals but stressed that the term hot spot should not be used for elevated air toxics concentrations related to being in traffic. There is of course extensive literature documenting the fact that being in traffic, or being in close proximity to traffic, leads to increased exposures to traffic-related air pollutants (HEI 2010). Thus, this aspect of the study contributed to the literature on traffic rather than to the literature on hot spots — specific areas with increased concentrations of toxic air pollutants derived from elevated emissions from various sources.

Overall, this study provides useful information on measurements of a series of air toxics at truck terminals. It also illustrates the challenges encountered in defining and documenting air pollution hot spots without accounting for the role of meteorologic conditions or establishing adequate background sites for comparison.

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