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Air Quality Modeling of Hazardous Pollutants: Current Status and Future Directions

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ABSTRACT
The current requirements and status of air quality modeling of hazardous pollutants are reviewed. Many applications require the ability to predict the local impacts from industrial sources or large roadways as needed for community health characterization and evaluating environmental justice concerns. Such local-scale modeling assessments can be performed by using Gaussian dispersion models. However, these models have a limited ability to handle chemical transformations. A new generation of Eulerian grid-based models is now capable of comprehensively treating transport and chemical transformations of air toxics. However, they typically have coarse spatial resolution, and their computational requirements increase dramatically with finer spatial resolution. The authors present and discuss possible advanced approaches that can combine the grid-based models with local-scale information.

INTRODUCTION
Hazardous air pollutants (HAPs), also called air toxics, contribute to a wide variety of human health and ecological effects. The U.S. Environmental Protection Agency (EPA) defines air toxics as "those pollutants that are known or expected to cause cancer or other serious health effects or adverse environmental effects." Air toxics involve a large number of chemical species that can be directly emitted into the atmosphere from stationary and mobile sources (primary air toxics), chemically formed in the atmosphere (secondary air toxics), or both. Some air toxics are chemically nonreactive, whereas others are reactive with slow or rapid decay away from the source. Air toxics emitted in significant quantities from isolated sources may have primarily local impacts. Air toxic pollutants, such as benzene and diesel particles, have been shown through model simulations to have a significant regional component, although there may also be significant "hot spots" associated with localized sources. Other air toxics, such as some forms of mercury, have a long atmospheric residence time and, consequently, are treated as global pollutants. These different characteristics influence the relative importance of the processes that govern air toxics concentrations (i.e., emissions, transport, transformation, and removal) and, consequently, affect the modeling approaches.

Air toxics are regulated in the Clean Air Act under Section 112 and include hundreds of chemical species. Because of the large number of air toxics, the relatively small number of toxics measured and the sparse nature of routine monitoring networks, many regulatory agencies rely on air quality models to estimate ambient concentrations and use monitoring data where available to evaluate model performance. Air quality models are useful for assessing baseline ambient concentrations, analyzing the relative importance of various emission sources, and testing emission reduction strategies. These assessments typically involve application of different models depending on program objectives: national, regional, urban, or local scale. These modeling applications focus primarily on outdoor "ambient" concentrations. Indoor concentrations can be both lower because of filtering of the outdoor air intake and scavenging of air toxics on surfaces or higher for those air toxics that are generated by indoor sources (e.g., benzene and formaldehyde). The focus here is on outdoor atmospheric concentrations.

Significant progress has been made in air toxics modeling over the past few years and it is, therefore, of interest to assess the current status of air toxics modeling. Presented here is an overview of the current status of simulating atmospheric concentrations of air pollutants and also a discussion of the existing challenges in air toxics modeling and recommendations for some possible approaches to address those challenges.

IMPLICATIONS
Air toxics models are being used by the general air pollution community in understanding not only the magnitude of the air pollution problem, but also in developing emission control policies and regulations. However, there are multiple approaches in estimating pollutant concentrations on various geographic scales. These approaches are discussed and recommendations for improvements are made.

MODELING AT VARIOUS SPATIAL AND TEMPORAL SCALES
Air quality models are useful for simulating atmospheric processes both on regional scales and localized neighborhood scales. Generally speaking, there are two major

types of air quality models: source-based dispersion models and grid-based chemical transport models. Local-scale dispersion models use either plume or puff representations of the emitted pollutants. They typically do not take into account atmospheric chemical reactions or they do so using simplified representations, such as first-order pollutant decay. For local scales, most methods used to estimate concentrations attributed to individual sources or source groups are steady-state Gaussian plume models. The typical range of such models is within 50 km from the source, and the maximum impact ranges from a few hundred meters to a few kilometers from the source. To identify the location and magnitude of the maximum concentration, a dense network of model receptors is essential. The temporal resolutions range from an hour to an entire year (annual average). The computational burden can become excessive if local scale models are applied to urban scale domains (e.g., 100 km $\times$ 100 km) involving thousands of emission sources and receptors.

For local-scale models that do not account for chemical reactions or long-range transport, other methods to estimate these components are used and are added to the modeled estimate. For example, the Ozone Isopleth Plotting Program Research (OZIPR) model[1] has been applied to estimate the contribution of secondary air toxics, such as acetaldehyde, acrolein, and formaldehyde. Contributions of these secondary air toxics are then added to the primary air toxics concentrations obtained with the local-scale model. For nonreactive pollutants, concentrations because of background must be added to the modeled concentrations. Background is defined as that part of the total concentration that is not accounted for explicitly in the modeling analysis, which includes the long-range transport of air toxics from distant sources (i.e., located outside the local modeling domain), noninventoried anthropogenic emissions, and natural emissions.

Finally, most local-scale applications have used steady-state Gaussian plume models where the plume center line is assumed to be a straight line aligned with the wind direction observed at a single meteorological station. Such an assumption may not be appropriate in areas with very complex wind fields. Nonsteady-state puff models avoid this shortfall, because they allow the use of different wind speeds and directions in the modeling domain. Different models are available based on the user's requirements and availability of modeling expertise, as well as the complexity of the meteorological fields in the modeling domain. The following models are typically used for local-scale applications (spatial scale ranging from tens of meters to several kilometers) in the United States.

The Industrial Source Complex (ISC) Short Term, Version 3 (ISCST3), is a steady-state Gaussian plume dispersion model applicable for estimating impacts from point, area, and volume sources for distances up to ~50 km using an hourly resolution.[2] The ISC Long Term (ISCLT) is a version of ISC that uses annual statistics of meteorological conditions for estimating annual-average concentrations.[2] The American Meteorological Society (AMS)/EPA Regulatory Model (AERMOD) is a steady-state Gaussian dispersion model that replaces ISCST3.[3] The California Line Source Dispersion Model (CALINE) is a steady-state dispersion model used to estimate air quality impacts near transportation facilities.[4] California Puff Model (CALPUFF) is a non-steady-state air quality model; it is compatible with the meteorological model (CALMET).[5]

Grid-based models are used to simulate the transport and formation of ozone (O_3) , acid rain, particulate matter (PM), and other pollutants formed from chemical reactions among precursor species that are emitted from hundreds or thousands of emission sources. Such models may be set up to apply to a wide range of scales ranging from global to urban. Typically, regional scale models are usually applied over hundreds of kilometers using an array of grid cells with a horizontal resolution of several kilometers. These three-dimensional grid models require heavy computational resources and are typically applied for multiday periods, up to an entire year. With expanded chemical mechanisms, these models have also been used to model the transport and transformation of air toxics. These regional-scale models can directly simulate chemically reactive species and address long-range transport.

However, unlike local-scale models, regional-scale grid-based models do not have the spatial resolution needed to correctly estimate concentrations very near the source. Because all of the emissions located within each grid cell are evenly distributed throughout the grid cell, these models are not able to simulate the effects from individual sources that occur within the source grid cell. Reducing grid cell size to correspond to the size of the area of interest is possible, but currently there are technical limitations to reducing grid size below ~1 km.

The following grid models are typically used for regional scale applications (spatial scale ranging from a few to thousands of kilometers): the Community Multiscale Air Quality (CMAQ) model[6] and the Comprehensive Air Quality Model with extensions (CAMx).[7]

Required Temporal and Spatial Resolutions

The required temporal resolution in modeling results is driven by the dose response needs of the particular air toxics that need to be assessed. Exposure to air toxics can be considered chronic or acute in nature. Acute impacts, such as respiratory irritation, are caused by short-term exposure (on the order of minutes or hours). Chronic impacts, such as development of cancer or chronic organ damage, occur because of a long-term exposure (on the order of years or decades). The need for flexibility in the temporal exposure duration requires modeling methods that can assess both short-term and long-term exposures to either an individual or a population. The need to assess spatial gradients in pollutant concentrations can be used to both assess the exposed population and locations of maximum exposed individuals.

The Role of Ambient Monitoring

Ideally, ambient air monitoring could be used to determine airborne concentrations of air toxics anywhere across the country. However, some analytical methods currently cannot detect levels of concern for certain air toxics pollutants, such as acrolein, arsenic, and hexavalent chromium, listed by EPA as risk drivers. Monitors are generally available only in urban areas, because, it is not

economically feasible to establish and maintain an extensive network of monitors in all communities. Thus, air quality models are used as a more practical solution to estimate ambient concentrations. Moreover, models are useful in developing and assessing control strategies. The nationwide monitoring network for HAPs is not as extensive as for criteria pollutants. A number of HAPs are monitored at the Photochemical Assessment Monitoring Stations sites, and a number of state, local, and tribal HAP monitoring programs are in place. These programs vary in terms of the species measured and the frequency and quality of measurements. In 2001, EPA began a pilot monitoring network of 10 locations for a smaller set of HAPs.

The Role of Emissions Inventories

Emissions inventories are critical inputs to dispersion models. EPA develops nationwide estimates of air toxic emissions every 3 yr and reports these data in the National Emissions Inventory (NEI)[8] as annual average emissions for point, area, and mobile sources. These inventories are estimated based on emission factors or source activity data. For point sources, the NEI are tabulated individually and include source location (stack coordinates) and stack parameters such as height, diameter, flue gas exit velocity, and temperature that are needed for modeling. When these parameters are not reported directly by the source, default values are used in the NEI. Area sources are smaller stationary sources of which the emissions are not individually tabulated in the point source inventory and reported on a county basis (e.g., dry cleaners). Mobile source emissions consist of two major subcategories: on-road mobile sources (e.g. cars, trucks, and buses) and nonroad mobile sources (e.g. aircraft, watercraft, railway engines, construction equipment, farm equipment, garden equipment, etc.). Emissions from natural sources (e.g., vegetation) are treated as area sources separately from anthropogenic emissions.

Emissions data must be processed for subsequent air quality modeling. For Gaussian models, the emissions preprocessor Emission Modeling System for Hazardous Air Pollutants (EMS-HAP)[9] is used. EMS-HAP spatially allocates area and mobile source emissions to individual areas, such as census tracts or 2 km $\times$ 2 km grids, or for on-road mobile sources, to road segments. Several types of spatial surrogates, such as population, industrial land use, and vehicle miles traveled, are used to distribute the emissions across the modeling domain. In addition, EMS-HAP temporally allocates the NEI emissions, which represent annual average emissions to desired time periods, such as 3-hourly, day of week, or seasonal values, to provide more realistic variations. The influence of all the assumptions used in the emission preprocessors on model performance can be significant and must be considered. For grid models, emissions models include Sparse Matrix Operator Kernel Emissions,[10] which processes the NEI to provide a gridded emission inventory with a 1-hr temporal resolution.

Allocating county-level emissions to census tracts or grids using some sort of allocation surrogates may be adequate for a more regional scaled assessment. However, such an approach may not adequately capture localized parameters, such as average speeds on specific roadways, vehicle activity, or construction site activity. Thus, more highly resolved (~100 m) emission factor and activity data may be needed.

CURRENT APPLICATIONS AND KEY ISSUES IN AIR TOXICS MODELING

Different approaches are currently being used to model air toxics concentrations in the ambient atmosphere. Described below are the major approaches used to model air toxics at various scales: national, regional (includes several states; urban basin or larger) urban (city or part of city), and local (neighborhood; tens to hundreds of meters or a few city blocks).

National Scale Assessments

The National Air Toxic Assessment (NATA) is designed to help EPA, state, local, and tribal governments and the public to better understand the air toxics problem in the United States.[11] NATA is designed to identify air toxics of greatest concern, characterize the relative contributions from different sources, set priorities for the collection of additional data, and establish a baseline for measuring progress in reducing risk from outdoor sources. To develop the national-scale estimates of annual average ambient concentrations of air toxics, EPA used the Assessment System for Population Exposure Nationwide (ASPEN) model.[12,13] ASPEN uses a Gaussian model formulation (ISCLT) and climatological data to estimate annual average pollutant concentrations for each census tract within the modeling domain. The ASPEN concentration predictions are designed to represent the spatial average concentration over the tract, not the concentration at the centroid. This is accomplished by using two different approaches to generating estimates, depending on whether the emission source is inside of the tract (spatial averaging of concentrations at modeling receptors) or outside the tract (interpolation to the tract centroid). A simplified empirical approach is used to estimate the secondary contribution of reactive pollutants. Model-to-monitor comparisons showed that the model performed within a factor of two at most sites for inert gases, such as benzene, but underpredicted for metals.

In general, NATA estimates that most Americans face some cancer and noncancer risks above the defined levels of concern from exposure to air toxics. Larger urban areas appear to carry larger risk burdens than smaller urban areas and rural areas, because the emissions of air toxics tend to be higher in areas where more people live. This trend is not universal, however, and can vary by pollutant, according to its emission characteristics. Additionally, NATA has not focused on the identification of geographic areas or populations that have significantly higher risks than others. Rather, it has been focusing on characterizing average risks across the country. Generally, the highest localized inhalation exposures and risks are not captured by this national-scale approach. It is important to use local-scale assessments to characterize exposures and risks within a particular urban area or neighborhood or very close to specific sources. Although uncertainties in emissions inventories, air quality models,

exposure models, and dose-response relationships are inherent in NATA-type analyses, the modeling results are best when used in a relative rather than absolute manner.

Regional Scale Assessments

Regional-scale assessments are useful to understand the long-range transport of air toxics and to provide quantitative information on regional background concentrations. Grid-based models simulate air toxics concentrations at global, continental, regional, and urban scales in a number of different studies. Some of the models used recently to simulate air toxics concentrations in the United States include CMAQ, CAMx, and the Urban Airshed Model (UAM) for toxics.[14] All of these models treat both gaseous and particulate species.

In 1999, the California South Coast Air Quality Management District conducted the Multiple Air Toxics Exposure Study II (MATES-II), which focused on the entire Los Angeles basin. The study is notable because it was the first regional-scale assessment to include limited, focused subregional emissions inventories and communities. Results from these subregional assessments suggested that air pollutant concentrations in studied communities were dominated by regional rather than local sources.[15] MATES-II was beneficial in the understanding of subregional assessments but had some limitations, particularly regarding the emission inventories. California South Coast Air Quality Management District conducted a similar study, MATES-III, in early 2004.[16] CAMx was also applied with the emissions and meteorological data developed for the MATES-II study.[17] The California Air Resources Board has applied CMAQ and CALGRID[18] air quality models to a much larger domain in Southern California (that encompasses the MATES-II domain) also for 1998.[19]

CMAQ is currently being applied to the contiguous United States with a 36-km resolution for 20 gaseous air toxics using two different chemical mechanisms, CBM-IV and SAPRC99.[20,21] Regional grid modeling studies using CMAQ at finer resolution have been performed for Philadelphia and Houston.[22,23] Seigneur et al.[24] applied CMAQ over a 4-day period in 1995. This application for benzene and diesel particulates was performed in the Eastern United States using 12-km $\times$ 12-km grid cells, with a nested domain over the Northeastern United States with 4-km $\times$ 4-km grid cells. The study used MM5 meteorological fields and an emissions inventory developed using Sparse Matrix Operator Kernel Emissions.

Predictions from regional models were typically within a factor of two of the observed concentrations on average (although there have been cases where model performance was worse, e.g., chromium in MATES-II, possibly because of uncertainties in the emission inventories). However, for air toxics that have local impacts, regional models can explain only a fraction of the variance in the observations (e.g., 25% with a 4-km resolution for the CMAQ simulation of benzene in New York as shown in Seigneur et al.[24]).

Urban Scale Assessments

As mentioned before, air toxics concentrations tend to be greater in urban areas than in rural areas. Consequently, many air toxics studies have focused on the urban scale.

EPA, as part of its uncertainty analysis initiatives for NATA, is conducting several urban scale modeling studies using ISCST3. One example is a study conducted for the Houston urban area, which contains a wide range of sources. Among the air toxics modeled is benzene, a stable, slowly reactive gas that can be, therefore, modeled using ISC. Modeling this large number of air toxics sources, addressing chemical transformations, and estimating background concentrations posed significant technical challenges.[25] A sensitivity study was conducted to investigate the effect of different approaches used for modeling mobile sources.[26] For road links modeled as area sources, ISCST3 produces results similar (within factor of two) to those from CALINE when both models use the same input data. Kinnee et al.[27] applied ISC using both gridded and link-based emissions to evaluate the effect of improved spatial allocation of emissions on ambient modeled benzene concentrations. Several conclusions can be drawn from the results of these studies, described below.

First, significant variations in air toxic concentrations occurred across the city, and for the five air toxics studied, highest concentrations occurred near the highest emitting sources. Therefore, detailed spatial allocation of emission sources is necessary for modeling on a local scale. Second, increasing the receptor density near high-emission sources changed the location of maximum concentrations. These results illustrated that steep concentration gradients can occur near high-emission sources and the importance of receptor placement and density for good model performance. Third, allocating on-road mobile emissions to road segments improved the agreement between modeled concentrations when compared with the observations and also resulted in higher estimated concentrations in the urban center. Last, it is important to refine national emissions inventory (better emissions amounts, source locations, and source characteristics) for input into local air quality model applications.

Many other urban- and community-scale modeling assessment projects are now under way, such as modeling studies in California (Barrio Logan and Wilmington) and the Philadelphia modeling study. Several urban-scale applications of plume dispersion models have been conducted. For example, Pratt et al.[28] applied ISCST3 to simulate the ambient concentrations of nine VOCs in the Minneapolis-Saint Paul, MN, metropolitan area. They recommended finer spatial resolution for major highways to better characterize concentrations. Cohen et al.[29] applied CALPUFF to Portland, OR, assigning emissions to specific roadway links. The model indicated that the zone of influence around a roadway for benzene was between 200 and 400 m. The results suggest that roadway links should be included to capture localized impacts of air toxics. ENSR[30] compared two models for the Houston area and found emissions to be the dominant source of uncertainty. Wheeler et al.[31] applied ISCST3 to three cities (Detroit, MI; Seattle, WA; and Cedar Rapids, IA). They found that emissions (in terms of both magnitude and spatial resolution) and regional background concentrations were major sources of uncertainty. Their conclusion underscores the need to address both local and regional aspects of air toxics jointly.

Neighborhood Scale Assessments

Typical community health assessments of environmental justice require finer-scale air toxic modeling. Localized air pollution impacts from sources such as heavily traveled roadways, warehousing facilities, or industrial or commercial facilities located near residences could reveal "hot spots" of concentrations that are higher than predicted by national-, regional-, or urban-scale models.

The neighborhood assessment studies are applied research projects that require more accurate estimates of all key model inputs than what are available in national-scale and urban-scale assessments. These refined inputs include a detailed emissions inventory with facility-specific emission rates, stack parameters and building dimensions, on-site meteorological observations to define local atmospheric transport, ambient monitoring data that can be used to estimate background concentrations because of long-range transport and to evaluate model simulation results, and the selection of receptor locations of special interest, such as schools or hospitals.

Key Limitations of Local-Scale Dispersion Models

One potential limitation of local dispersion models is their inputs (i.e., emissions, source characteristics, and meteorology). The uncertainties associated with the emissions magnitudes typically dominate the uncertainties associated with the modeling results. Sax and Isakov[32] showed that meteorology, spatial distribution of sources, and the model formulation (user's selection of model parameters) also contributed to the uncertainty in air toxics concentrations, but they were not as important as the emissions. A similar conclusion was reached in the sensitivity analysis of ISCST3 and AERMOD for the Houston area.[30] The study found that the simulated benzene concentrations differed between the two models by ~35% on average. On the other hand, uncertainties in emissions were estimated to lead to uncertainties in the predicted concentrations of a factor of 2.6 (for AERMOD) to 2.8 (for ISCST3). The other urban-scale modeling studies mentioned above also cited emission uncertainties as a dominant concern.

Dispersion models, such as ISC and AERMOD, involve the assumption of steady-state meteorology. This assumption is appropriate for short periods (e.g., 1 hr) over which atmospheric conditions can be considered not varying. However, over longer periods, wind speed, wind direction, atmospheric stability, and mixing height may vary, and these assumptions may not be valid. In such cases, puff models that take into account the transient character of the meteorology are more appropriate. As mentioned above, such models cannot simulate properly the concentrations of highly reactive chemical species, such as aldehydes.

Key Limitations of Regional Models

The spatial resolution of the grid is the major limitation of grid-based models for assessing the local impacts of air toxics. Several approaches have been used to address this limitation; they are discussed in the following section. Uncertainties associated with emissions can be the dominant factor affecting model performance for some air toxics (e.g., chromium in the MATES-II applications).

RESOLVING FINE SCALE: COMBINATION OF REGIONAL AND LOCAL MODELING

Regional Grid Modeling with Subgrid Spatial Variability

Figure 1a depicts the overall concept of this approach. Because the regional grid model cannot resolve all of the local details in air toxics concentrations within the grid, some parameterization of the subgrid spatial variability (SGV) is used to represent the nonresolved features in a grid cell. In Figure 2, the pollutant concentration is the model value of a grid cell of some prescribed cell size (indicated here as 12 km). Inherent finer-scale detail is evident; a description of this subgrid variability would provide interesting and useful information for some applications. In this example, the SGV for the 12-km cell was obtained using results from model simulations at finer resolution (1-km grid size). A nested grid approach was used in this case. Spatial variability can be depicted with concentration histograms that are derived from this 1-km simulation. The following example briefly illustrates these concepts.

Figure 3 depicts various levels of details in CMAQ simulations for the Philadelphia area at 36-, 12-, 4-, and 1.3-km grid size for formaldehyde.[22] Modeled concentrations have fine-scale spatial features that only become apparent as the spatial resolution of the simulation becomes smaller. Application of grid models for operational applications would probably use coarser grid resolution, such as 12 km, with resulting loss of fine-scale details. The details seen in the 1.33-km results would, thus, be subgrid features at 12-km grid resolution. Using results of the smallest grid size, concentration histograms were created using values of 81 cells of 1.33-km size for each 12-km cell to represent its SGV. The histogram can be described as a probability density function (PDF) shown in Figure 4.[23]

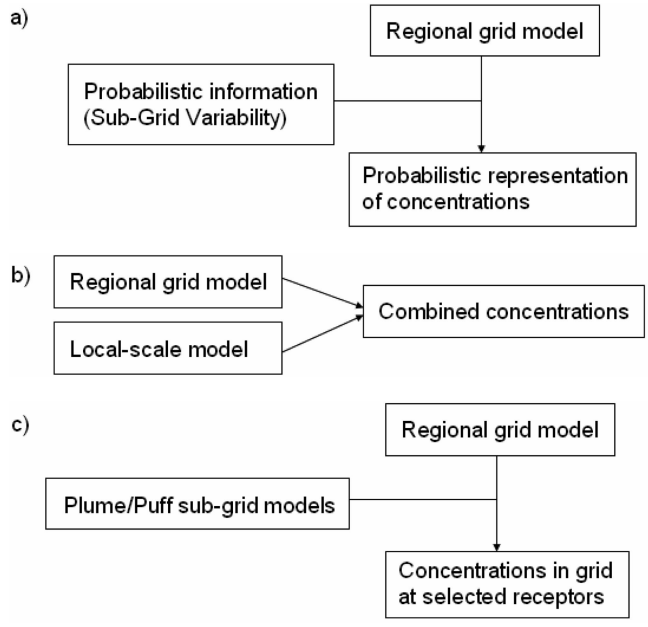

Figure 1. Schematic descriptions of three regional/local modeling approaches: (a) probabilistic approach: regional grid model with subgrid variability; (b) deterministic approach: separate regional grid model and local plume model; and (c) hybrid approach: regional grid model with subgrid scale treatment of selected sources.

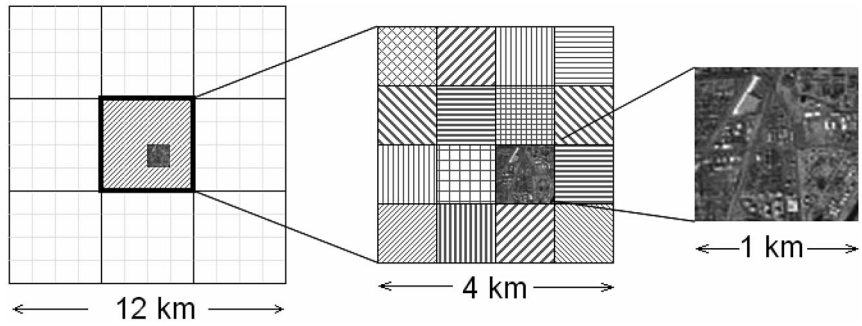

Figure 2. Conceptual approach to estimate subgrid variability in gridded modeled concentrations pollutant concentration. In this example, nests of 4 and 1 km are obtained for an arbitrary 12-km grid cell.

The challenge is to estimate SGV to complement the coarse grid air toxics simulations in routine applications. The issue is to obtain PDFs for each grid cell and pollutant. One approach is to run the model at fine scales for a variety of model scenarios and subsequently to build parameterizations from these results, thus serving in an a priori, off-line basis. However, it is recognized that SGV depends on the air toxic species and varies with time and location. Also, modeling at fine scales is computationally intensive, and, moreover, the input requirements and model physics must be appropriate at the commensurate fine scales for simulating meaningful results. In principle, the PDFs should encompass all variability within the grid cells, even those at grid sizes smaller than shown in the example. The PDFs can be estimated by the use of modeling tools, such as computational fluid dynamics and large eddy simulations. Preliminary results obtained for Houston and Philadelphia have shown that this new approach offers promise, but is still in a research stage.

Separate Regional Grid Modeling and Local Plume Modeling

Figure 1b depicts the overall concept of a hybrid approach, where a regional grid model and a local plume model are run independently. The regional grid model provides the regional background concentrations, and the local plume dispersion model provides the air toxics concentrations because of local emission sources. Then, the results of both model simulations are combined to provide the total ambient air toxics concentrations. Care is required to avoid counting air toxics emissions twice when combining the two simulations. As an example, this methodology was used in the Wilmington Air Quality Modeling Study. The Wilmington study domain is shown in Figure 5. In this figure, mobile sources (road segments) are shown as black lines, and stationary sources are shown as black dots. Census tracts are also shown in the figure as gray polygons. The ISCST3 dispersion model was used to simulate ambient average concentrations on a local scale;

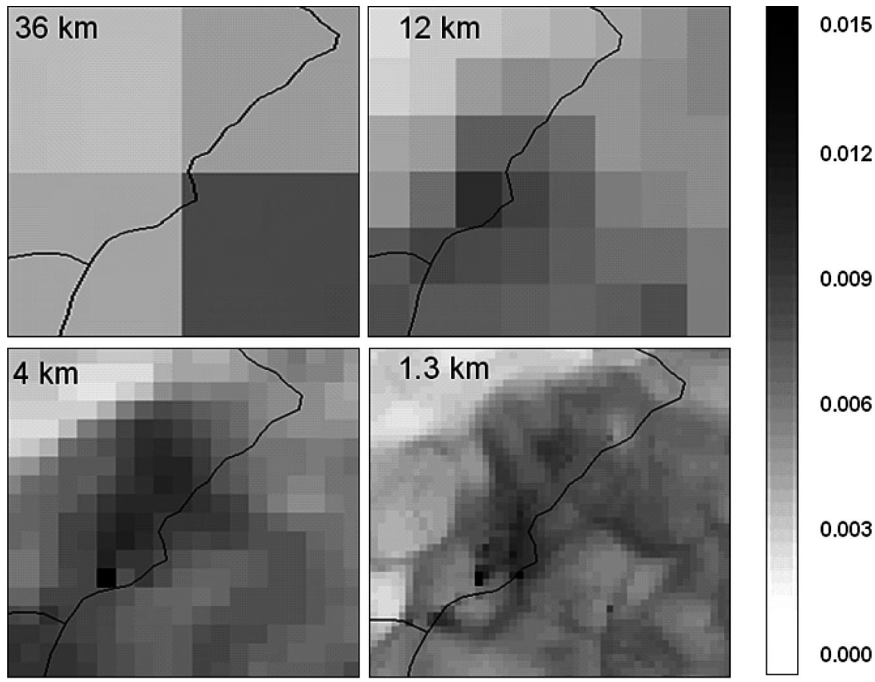

Figure 3. Formaldehyde concentrations from CMAQ in Philadelphia at 36-, 12-, 4-, and 1.3-km grid size.

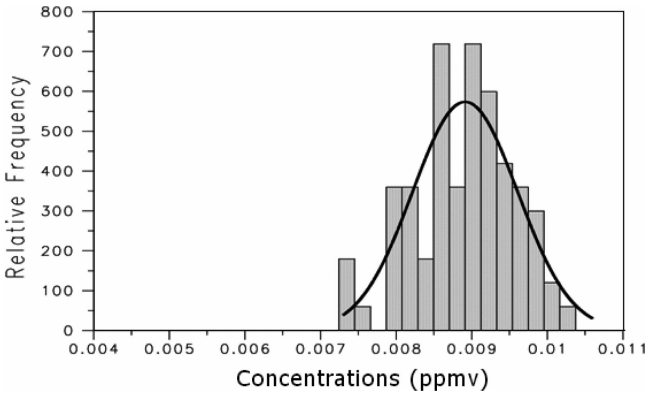

Figure 4. Relative histogram for formaldehyde with fitted normal PDF, 6:00 p.m., Philadelphia.

a regional-scale model, CALGRID, was used to account for photochemistry and long-range transport.[19] The locations of 4-km × 4-km model grid cells are shown in Figure 5 as dashed lines.

However, combining the results from microscale and regional-scale models is not straightforward. Including the same emission sources in both regional-scale modeling and microscale modeling and adding the modeling results will "double count" the impact of these sources. To avoid double-counting, a "zero out" approach has been used. Two annual model simulations have been conducted: one for the base case and another one excluding all of the local emissions from those grid cells in the modeling domain. The difference between the baseline simulation (that uses all emissions) and the new simulation provides an indication of how local emissions impact local concentrations. Figure 6 shows that a contribution from regional background in Wilmington is significant. The impact of local sources is more noticeable for inert pollutants, such as benzene, and less for reactive pollutants, such as formaldehyde.

Results from the ISCST3 dispersion model were combined with the regional background simulated by a grid-based model, CALGRID. The ISCST3 dispersion model was applied to reveal hot spots of concentrations caused by individual sources within the modeling domain, using locally derived emissions from mobile sources. Emissions were allocated to roadway locations (roadway links) using the Southern California Association of Governments travel demand model and default fleet average emission factors developed using the EMFAC2002 model[33] and modeled by ISCST3, treating individual links as area sources. An example of modeling results for formaldehyde is shown in Figure 7. The figure displays modeled concentrations, generated using ISCST3 for stationary sources (dots) and roadway sources (lines) in the modeling domain, and adjusted for the regional background from CALGRID simulation results. The results demonstrate concentrations gradients near major roads and stationary sources.

Regional Grid Modeling with Subgrid Scale Modeling of Selected Sources

Figure 1c depicts the overall concept of this approach, where emissions from local sources are treated with a local dispersion model that is imbedded within the regional grid model. This approach has been used to simulate the emissions from large point sources[34] but can be extended to other types of sources, such as line sources (e.g., roadways) and area sources. This approach treats regional and local sources within the same modeling system, avoiding the issue of double-counting emissions noted in the previous section. The mix of dispersion models not only handles local sources appropriately but also calculates concentrations at specific receptor points. The subgrid scale modeling approach based on puffs is likely to be more flexible for air toxics applications that will involve many different sources with overlapping areas of impact than an approach based on plumes (the puff modeling approach can handle overlapping plumes and wind shear by merging and splitting of the puffs).

Figure 8 presents an example of the application of a subgrid scale modeling approach for two air pollutants, O_3 and nitric acid (HNO_3). In this application,[34] CMAQ was used as the grid-based Eulerian model (with a 12-km horizontal resolution), and a reactive puff model, SCICHEM, was used as the subgrid scale model. The resulting model with subgrid scale treatment of large point sources is referred to as CMAQ-advanced plume treatment. The puff model had previously been satisfactorily evaluated using aircraft data collected during the 1995 Nashville Middle Tennessee Ozone Study as part of the Southern Oxidants Study.[35] Figure 8 shows the differences in ground-level concentrations of O_3 and HNO_3 between simulations with and without the subgrid model. The subgrid model was applied to emissions from 30 major point sources for a July 1995 episode (i.e., with all emissions released within the grid system). Differences in air pollutant concentrations are significant, thereby highlighting the fact that it is essential to correctly represent the subgrid processes of emissions from major sources.

CONCLUSIONS

Combining Regional and Local Models

As discussed above, a correct appraisal of many air toxics will require a combination of regional and local scale modeling. Presented here are three different approaches to address this multiscale problem. These three different approaches are not mutually exclusive and can be collectively exhaustive.

The regional grid modeling approach with subgrid spatial variability (Figure 1a) is designed to address toxics concentrations taking into account their variability within an area. It is well suited for use in combination with population activity patterns, which are by nature aleatory.

The other two approaches combine a regional grid model with local plume/puff models. The approach that uses these two types of models separately and then combines results to calculate the sum of regional and local concentrations (Figure 1b) can potentially be computationally more efficient, particularly if the air toxics are treated as chemically inert. It is, however, cumbersome to properly treat reactive air toxics.

The approach that uses the local-scale model imbedded within the regional grid model (Figure 1c) offers a more accurate formulation of the multiscale problem and

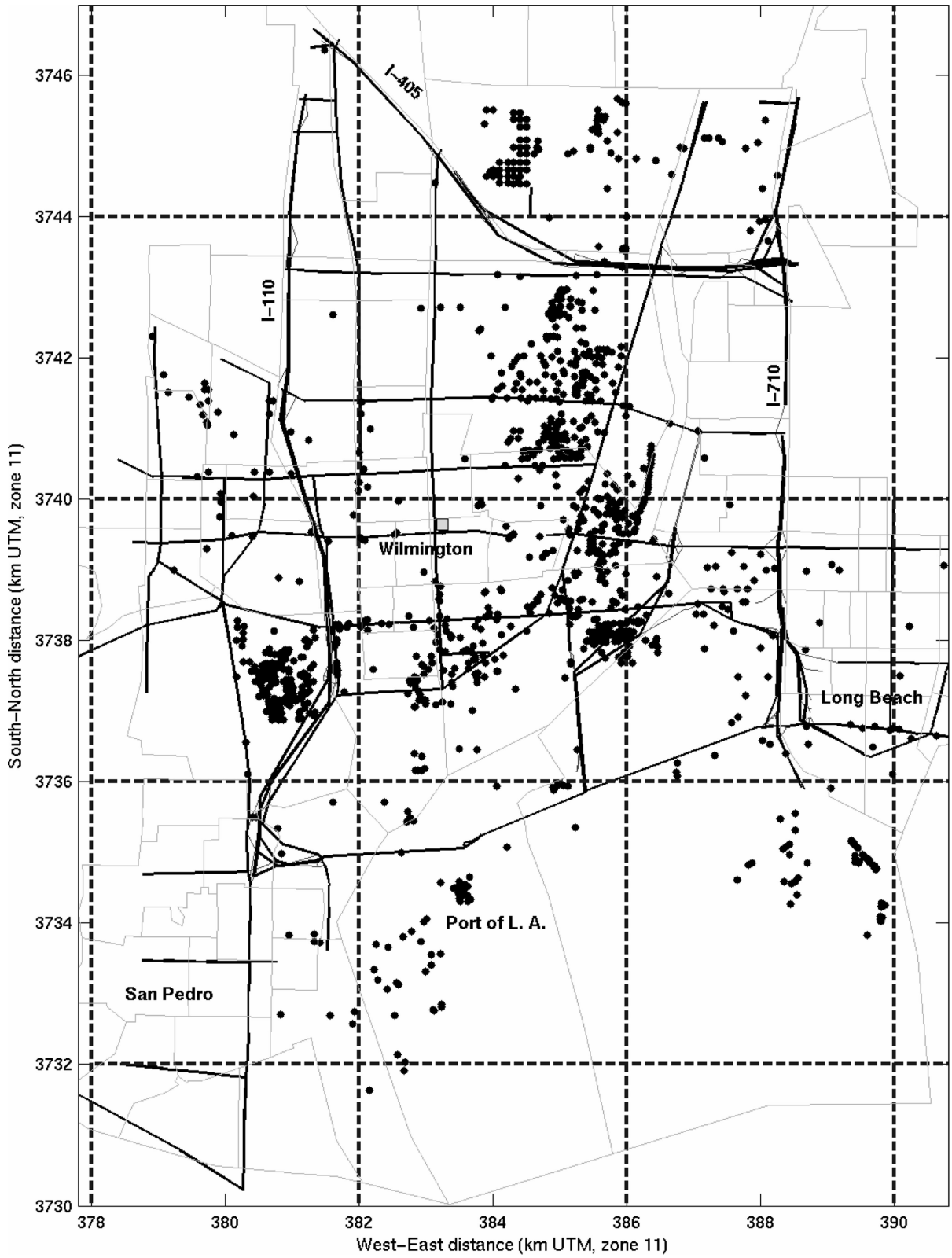

Figure 5. Schematic map of the modeling domain in Wilmington, CA.

also allows a better treatment for chemically reactive air toxics. The modeled air toxics concentrations would need to be calculated: (1) in the grid, and (2) at selected receptor points. The concentrations resolved by the grid system would then be added to the puff concentrations to obtain the total air toxics concentrations at each receptor. The computational limitations of this latter approach could, however, prevent its application to a very large number of sources. Consequently, a possible overall approach could be to use the original grid modeling approach as a screening

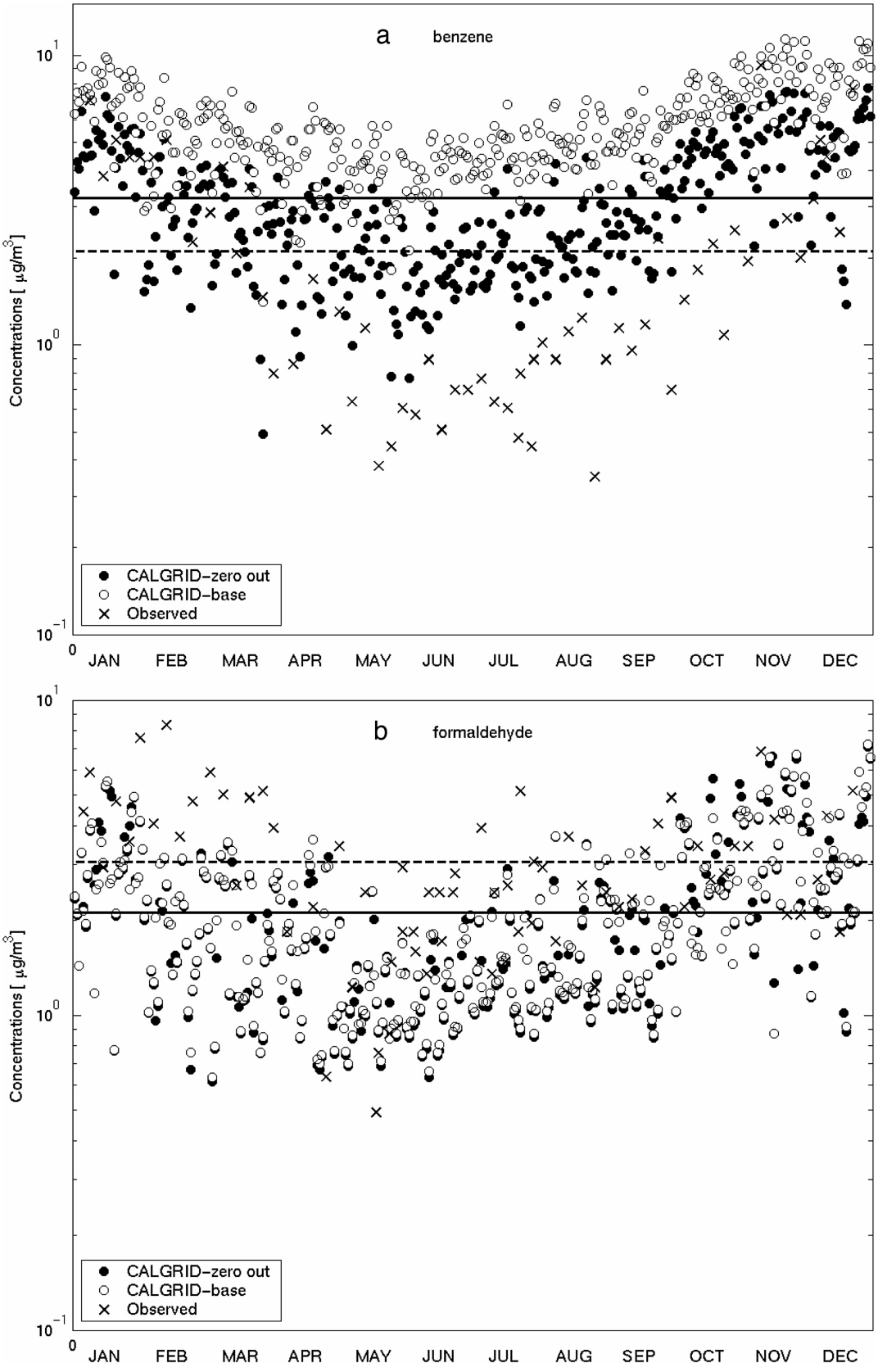

Figure 6. Regional background of benzene (a) and formaldehyde (b) in Wilmington, CA, from CALGRID. (Solid and dashed lines represent annual-average concentrations from CALGRID and from observation, respectively).

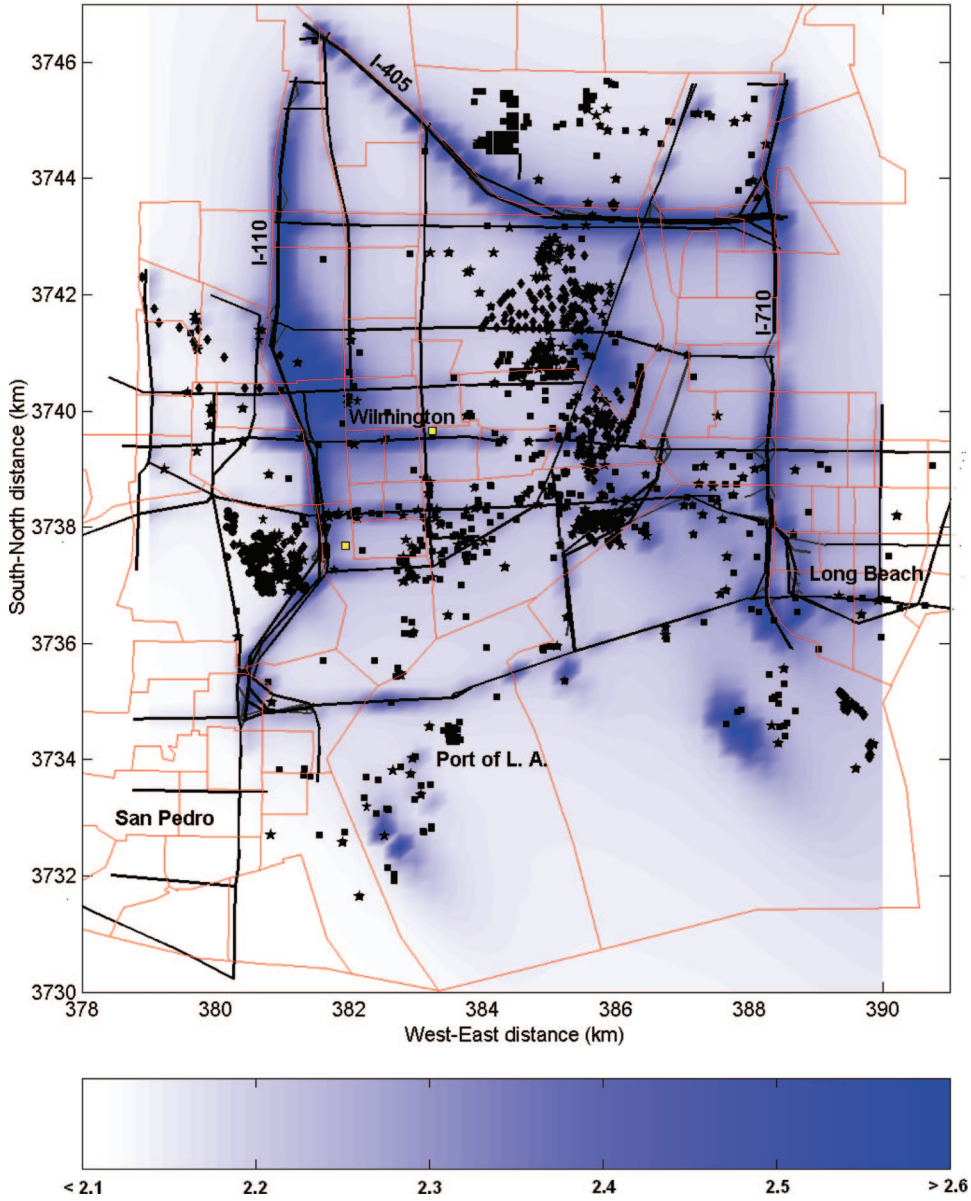

Figure 7. Modeled formaldehyde concentrations in Wilmington, CA, from local stationary and mobile emissions sources, adjusted for regional background from CALGRID

tool and the latter, more accurate approach, as a tool concentrating on the air toxics sources and receptor areas of most interest for air toxics.

Improving Model Inputs

Air toxics modeling studies have shown that a major contribution to the uncertainty in the model simulation results originated from the model inputs rather than from the model formulation. It is important to improve the air toxics emission inventories by providing a better quantification of the magnitude of the air toxics emissions and their temporal (1 hr) and spatial (~100 m) distribution and, for particulate-bound air toxics, the particle size distribution of the air toxics (fine PM vs. coarse PM). Use of local meteorology can also be important in areas with complex terrain, coastal areas, and urban canyons.

Evaluating Model Performance

Model performance evaluations are needed before the use of air toxics models for regulatory or policy applications. However, there is only a limited amount of available ambient monitoring data. Similarly, emissions and meteorological conditions vary considerably during a 24-hr period, and it is difficult to use these data for diagnostic model evaluation. It is desirable to evaluate the ability of air toxics models to resolve proper time scales. Hourly and 3-hr average data are recorded in the Photochemical Assessment Monitoring Stations network; however, this network only operates during the O_3 season. Because most air toxics data consist of 24-hr averages, this does not provide a full data set for proper model evaluation, and there is a dire need for air toxics monitoring data with better temporal resolution.

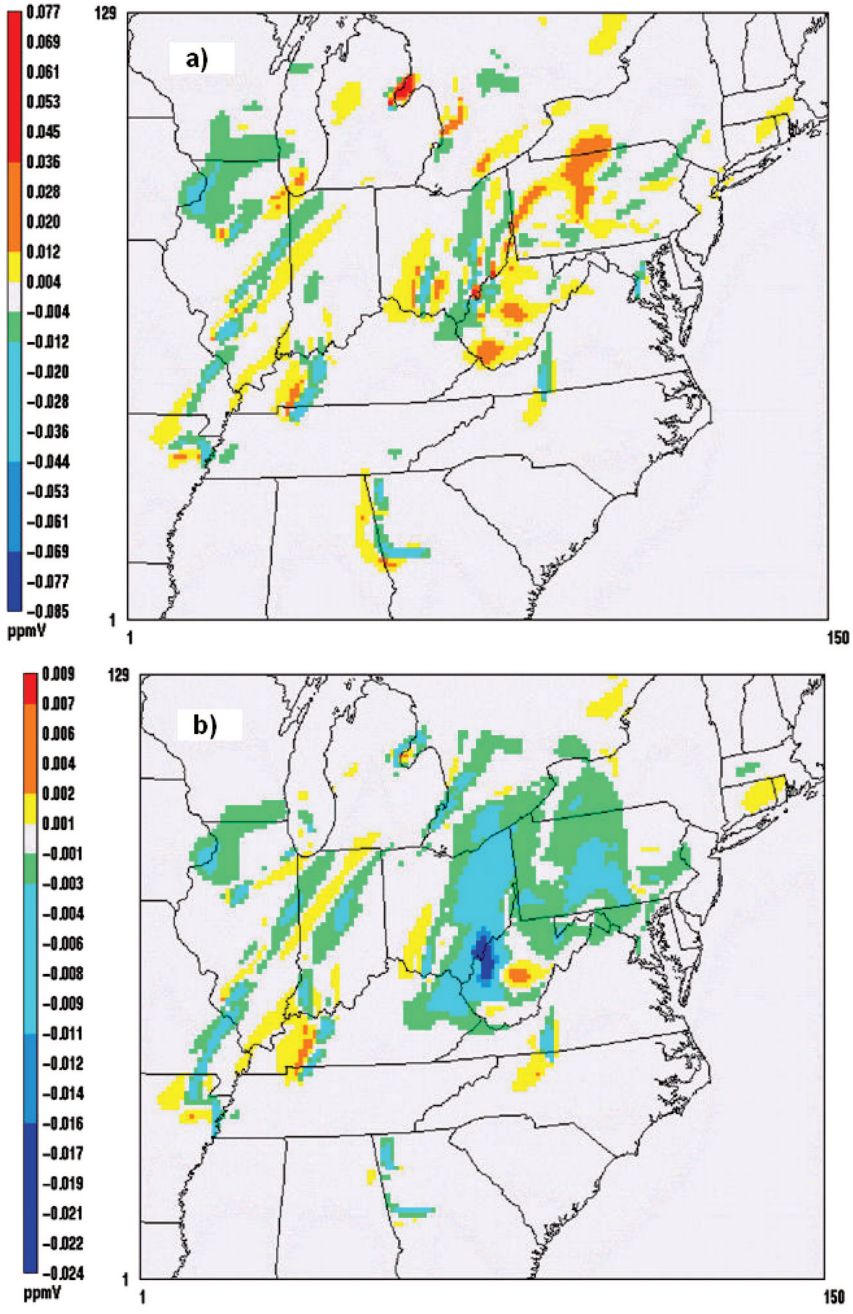

Figure 8. Differences in hourly ground-level concentrations (3:00 p.m., July 13) of O_3 (a) and HNO_3 (b) with and without subgrid scale treatment of 30 major point sources.[34] (Source: Karamchandani, P., Seigneur, C., Vijayaraghavan, K., and Wu, S.-Y., Development and application of a state-of-the-science plume-in-grid model, *J. Geophys. Res.*, 107, 4403–4415 (2002), Copyright (2002) American Geophysical Union, reproduced by permission of American Geophysical Union).

Treating Uncertainties

Assessing uncertainties is an integral part of the health-risk assessment process.[36] It is, therefore, desirable to incorporate some treatment of uncertainties in the entire air toxics modeling process: emissions and meteorological inputs, model formulation, monitoring data, and exposure and risk. This process has been implemented systematically, for example, by Sax and Isakov.[32] However, characterizing uncertainty for reactive pollutants is more complicated[37]; therefore, improved methods for quantifying uncertainty are needed.

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DISCLAIMER

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